Behaviour of Tritium in the Environment

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BEHAVIOUR OF TRITIUM IN THE ENVIRONMENT


INTERNATIONAL ATOMIC ENERGY AGENCY VIENNA, 1979
FOREWORD

There is considerable interest in the behaviour of radionuclides of global character that may be released to the environment in the process of producing nuclear power. Tritium is of particular interest because of its direct incorporation into water and organic tissue.

Progress has been made over the past few years in the assessment of tritium distribution, in detection methods and in biological studies. Several meetings have given scientists an opportunity to present papers on tritium, but no symposium devoted specifically to tritium has been organized by the International Atomic Energy Agency since 1961.

In addition, a five-year IAEA co-ordinated research programme on the behaviour of tritium in different types of ecosystems came to an end in 1978. A symposium was required to provide a good opportunity for the participants in the co-ordinated programme to give information on the residence time, pattern of movement and distribution of tritium in typical ecosystems, its incorporation into biological compounds and its transfer along food chains, in order to determine the biological significance of this radionuclide in various climatic conditions and ecosystems.

The occasion for this wide exchange of information was provided by the international symposium on the Behaviour of Tritium in the Environment held from 16 to 20 October 1978 in San Francisco. The symposium, organized by the IAFA and the OECD Nuclear Energy Agency, in co-operation with the United States Department of Energy and the Lawrence Livermore Laboratory, was attended by about 220 participants.

The text of the papers, together with a record of the discussions, is published in these Proceedings.
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DISTRIBUTION OF TRITIUM

Session 1 and
Session 2, Part 1
ATMOSPHERIC HT AND HTO: V. DISTRIBUTION AND LARGE-SCALE CIRCULATION

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Abstract

ATMOSPHERIC HT AND HTO: V. DISTRIBUTION AND LARGE-SCALE CIRCULATION.
The two major chemical forms of atmospheric tritium are water vapour (HTO) and hydrogen gas (HT). These forms have quite different sources, distributions and sinks. The chemical conversion from HT to HTO in the atmosphere proceeds with a characteristic time of 6.5 years. Combined with the radioactive decay, a net lifetime of 4.8 years is estimated for atmospheric HT. HT is released predominately at the surface in mid- to high latitudes in the northern hemisphere. A negative gradient southward has been found from aircraft transects and from sampling at surface stations. After many years of a relatively constant global inventory of 1.1 kg of tritium gas, the HT mixing ratios decreased during 1977, with the sharpest drop at high latitudes. The estimated decline in annual production was 100 g. At the end of 1977, the atmospheric HT burden was 1.0 kg, and the estimated annual release was 200 g. An unknown portion is present as T₃ gas. The effect of T₃ is to decrease the net lifetime to 3.7 years. In the troposphere, the cycle of HTO has been treated exhaustively by others. The stratospheric distribution of HTO has been sampled from aircraft, and found to increase rapidly with height above the troposphere. An annual cycle has been observed, in which the lower stratosphere is depleted during the spring, and replenished by subsidence from higher levels during summer and fall. The effects of a nuclear test by the People’s Republic of China in November 1976 have been clearly observed in the stratospheric HTO; however, no HT deposition was found. Presumably, the HTO at higher levels was originally deposited by the large nuclear weapons tests of the 1960s. An estimated 5 kg of tritium are now present in the stratosphere below 19 km.

1. INTRODUCTION

Large-scale releases of anthropogenic tritium to the atmosphere began with the detonation of the Ivy-Mike thermonuclear device on 31 October 1952. Estimates of the tritium releases from thermonuclear tests, up to 1962, have been summarized by Miskel [1] and others. Subsequently, as a result of the test ban
treaty, testing in the atmosphere declined greatly, and consequently the tropospheric content of HTO. Other sources of man-made tritium in various chemical forms are discussed by Crowson [2] and include nuclear reactors, reactor fuel element reprocessing, and tritium production for civilian and military use. A general review has been made by Jacobs [3].

Most of the early interest in world-wide tritium has been focused on tritiated water, and the first large-scale applications of tritium to meteorology and hydrology used the tritium content of rainwater as the data base. A world-wide network of rain collecting stations has been operated by the International Atomic Energy Agency (IAEA) in co-operation with the World Meteorological Organization [4–8]. Krey and Krajewski [9] successfully used the 1963–65 data [4, 5] to estimate water lifetime in the atmosphere, in spite of rain samples being rather deficient in representing tritium content of the troposphere, according to Östlund and Mason [10]. Vapour data were used for studies of mid-latitude tropospheric water transport by Ehhalt [11, 12].

2. DEFINITIONS

2.1. Speciation

The two major chemical forms of atmospheric tritium are tritiated water vapour (HTO) and tritiated hydrogen gas (HT, T₂ or possibly DT). The former species follows the water cycle in the atmosphere, having a residence time which varies from a few days in the troposphere to years in the stratosphere [13]. The HT, on the other hand, is well-mixed globally, in areas away from nuclear industry, over the entire troposphere and the lower stratosphere [14]. Conversion from HT to HTO in the atmosphere occurs quite slowly.

2.2. Units of measurement

It is necessary to distinguish carefully between specific activity and mixing ratio. Both types of units are in common use, and frequently lead to confusion.

2.2.1. Specific activity

These units are measures of the isotopic abundance of the species in question. Common units for tritium are the tritium unit, defined as \([T]/[H] \times 10^{18}\), the disintegration per unit time per unit mass (or volume) of substance which may be either water or hydrogen gas, and the picocurie per unit mass or volume. Such units are chiefly appropriate to hydrology or oceanography.
2.2.2. Mixing ratio

Mixing ratios are measures of the number of atoms of the isotope under study contained within a unit mass of air. Common units are tritium atoms per mg of air, disintegrations per minute per standard cubic metre of air, and picocuries per standard cubic metre of air. These are, in meteorological terms, conservative units. That is, they do not vary under expansion or compression of the air.

3. EXPERIMENTAL METHODS

3.1. Sampling systems

The principle of our air sampling system is outlined in Fig.1. Air at a measured flow rate, from 1 to 150/1 min., is first passed through a bed of Molecular Sieve, called a sieve trap, where all the water vapour is removed. At this point, tritium-free hydrogen is added, so that the total hydrogen concentration of the air is between 0.1% and 0.5% by volume. The dry mixture of hydrogen and air is now passed through a bed of colloidal palladium, carried on Molecular Sieve No.4A, called the combustion trap. By this catalyst the hydrogen is converted to water, which is immediately adsorbed on the sieve bed supporting the catalyst. At some ground locations, the additional stage shown in Fig.1 has been added for determining tritium in atmospheric hydrocarbons. A methane carrier is added, and the air is passed through a catalytic combustion furnace containing Hopcalite (MSA, Inc.) catalyst at 400°C. The resulting water is adsorbed on a third sieve trap.

Each run thus yields two or three samples of water, each adsorbed on Molecular Sieve. The sample in the first trap consists of the original H₂O, HDO and HTO of this particular air sample. From this one can determine the true amount of HTO per kg of air and the TU value of the moisture (if larger than 0.3 g/kg). The second trap contains all the tritium that originally occurred as HT, DT and T₂, now in the form of HTO. From this we obtain a true measure of the quantity of tritium, as number of T-atoms, in the form of molecular hydrogen, in the air sample. Information on the specific activity and D/H ratio of the original atmospheric hydrogen gas is lost in the dilution by H₂ carrier. The third trap contains the tritium originally in the form of tritiated hydrocarbons, now in the form of HTO.

3.2. Sample recovery

To recover the adsorbed water, the sieve trap or combustion trap is placed in a furnace and connected to a vacuum line through a freeze trap kept at dry ice
FIG. 1. Atmospheric HTO/HT tritiated hydrocarbon sampler.
temperature. When most of the sample has been removed, a known small quantity of tritium-free water is introduced into the trap to displace any remaining sample. The dilution caused by the flush is accounted for in the calculations. We have reported elsewhere the procedures for reconditioning the traps for further use [15].

3.3. Sample counting

The sample activity is determined by low-level proportional gas counting. The procedures and equipment have been described fully elsewhere [16].

3.4. Calculations

The data necessary for calculation of atmospheric mixing ratios are the quantities of sample recovered from each of the two or three traps, the activities of those samples, and the standard volume or mass of the air sampled. Note that the dilution factor is not needed for this calculation. The basic equation is

$$\omega = \frac{A \times Q}{M} \times F$$

where \( \omega \) is the mixing ratio of the species
A is the activity of the sample water
Q is the quantity of the sample water
M is the mass of the air sampled
F is the conversion constant for the units employed

For example, if \( \omega \) is desired to be in T-atoms per mg of air, A is in tritium units, Q is in grams of water, and M is in kg of air, F is 0.06684.

The analysis of the errors is straightforward, and is derived from the estimates of the errors in the above parameters. Typical accuracy is ±3%.

4. RESULTS

4.1. Ground-level station operation

Operation of atmospheric HT and HTO samplers began in Miami in 1968, and has been continuous since late 1969 at a rate of 2 or 3 samples weekly. The atmospheric hydrocarbon stage was added in late 1975. A sampler was installed in Fairbanks, Alaska, in late 1970, and has also operated continuously since then. A sampler was operated at the Mauna Loa Observatory on Hawaii from mid-1971 until the end of 1977. A sampler has been operated at Baring Head, near
Wellington, New Zealand, since early 1975. Finally, a sampler including the atmospheric hydrocarbon stage began operation at the South Pole in December 1977. Data for all stations through 1976 are available in unpublished reports [17–19].

4.2. Airborne sampling

Numerous flights have been made with various types of aircraft, beginning with DC-6, and including C-135 (similar to Boeing 720), Sabreliner, C-130 (Hercules) and WB-57F research aircraft. Data for most flights through 1976 are also contained in the above references [17–19], and interpretations of the data have been published [13, 14].

4.3. Tropospheric HT data

The mixing ratios of HT found by the stations identified in paragraph 4.1 above are shown in Fig. 2 as annual averages from the beginning of each station operation through 1976, and seen in this presentation. Fluctuations are the greatest at Fairbanks, and decrease towards the south.

From previous work [14], we have shown that the production of HT, assumed to be all in the northern hemisphere, can be described by

$$J_n = \lambda_n M (C_n - C_s) + (\lambda_r + \lambda_c) M C_n$$

where $\lambda_n$, $\lambda_r$, $\lambda_c$, $M$, $C_n$, $C_s$ are:

- $\lambda_n$: is the HT mixing ratio, T atoms/mg air
- $J$: is the production of HT, T atoms/year
- $\lambda_r$: is the radioactive decay constant for tritium
- $\lambda_c$: is the chemical decay constant for hydrogen gas
- $\lambda_e$: is the air exchange coefficient between hemispheres
- $M$: is the hemispheric atmospheric mass, mg

Differentiating with respect to time, we get

$$\frac{dJ_n}{dt} = (\lambda_c + \lambda_r + \lambda_e) M \frac{dC_n}{dt} - \lambda_c M \frac{dC_s}{dt}$$

Using the observed differences in mean annual mixing ratios as approximations of the time derivatives, we calculate $\Delta J_n$ to be $-100$ g. The annual production in 1977 is estimated from the first equation to be $200$ g. Values of the constants were estimated in the work cited above [14]. The estimation will be reviewed
FIG. 2. Tropospheric HT mixing ratios 1968–78.

FIG. 3. Atmospheric hydrogen residence time versus interhemispheric air exchange time.


FIG. 7. Stratospheric HTO mixing ratios, T-atoms/mg air 1976 (August).
briefly. In 1971 and 1972, tropospheric HT mixing ratios in both hemispheres were relatively constant. In each year, sampling flights were made in the upper troposphere from California to New Zealand. Significant interhemispheric differences in the HT mixing ratios were detected in both years. Assuming a steady state, the source and loss terms for the southern hemisphere can be equated as follows:

\[ \lambda_c MC_n + J_s = (\lambda_c + \lambda_r + \lambda_c) MC_s \]

where the terms are as explained above. Since \( J_s \) can be neglected, the equation can be solved

\[ \lambda_c = \left[ \lambda_c (C_n - C_s) / C_s \right] - \lambda_r \]

Using the data of 1972, a value of 0.155 year\(^{-1}\) was estimated for \( \lambda_c \), or a residence time of 6.5 ± 0.8 years. Use of the decay constant of tritium of 0.0565 year\(^{-1}\) is only valid for the HT molecule. If only T\(_2\) molecules were present, the effective decay constant would be twice that, 0.113 year\(^{-1}\), and the chemical decay constant would be 0.099 year\(^{-1}\), or a residence time of about 10 years.

Using only our data, we can parametrize $\lambda_\sigma$ and $\lambda_c$ since the other factors are well known. Figure 3 shows that relationship between the reciprocals of the two quantities, based upon 1972 data. Note the effect of $T_2$ as stated above.

Using only the data shown in Fig.2, and assuming the HT to be well-mixed vertically up to 19 km, the estimated annual inventories of atmospheric HT have been calculated and are shown in Fig.4.

4.4. Stratospheric HTO

The following discussion is restricted to HTO in the upper troposphere and lower stratosphere. Although our airborne sampling systems collect both HT and HTO, almost all flights have found HT to be well-mixed vertically. Figure 5 shows the HTO mixing ratios found on six sampling flights over Boulder, Colorado, during 1972. The shaded area includes all HT mixing ratios found on the six flights, illustrating the well-mixed situation.
By contrast, the HTO mixing ratios are seen to decrease with height in the upper troposphere to a minimum somewhere below the tropopause, then increase in a nearly exponential manner until heights well above the tropopause are reached. This feature has been used to estimate the vertical flux of water in this region [13].

More recently, several series of flights have been flown under the auspices of Project Airstream, which is a seasonal inventory of stratospheric radionuclides. These flights sample at four levels above the tropopause, and below 19.2 km, over a latitude range from 0° to 75° north. Series of flights are flown in April, July and October of each year. Figures 6, 7, and 8 show HTO mixing ratios found on the three series of 1976. This year was chosen because of the absence of significant man-made perturbations. A cycle is seen that includes relatively high mixing ratios in the spring, depletion and transport southward during the summer, and finally subsidence of large amounts of HTO from higher levels in the fall. The effect of the Hadley cell can be seen in all seasons, the HTO being depleted rapidly in that region by vertical circulation.

An atmospheric fusion test was conducted by the People's Republic of China on 17 November 1976, which was after the conclusion of the fall airstream cycle. The cycle for the spring of 1977 was impaired by equipment problems, and the next complete set of samples was taken in July 1977. Figure 9 shows the data obtained. Note that an order of magnitude increase appears in the HTO mixing ratios. Although it does not appear in the figure, the HT mixing ratios were unperturbed.

5. SUMMARY AND CONCLUSIONS

The predominant tritiated species in the atmosphere are HTO and HT. They must be treated separately, since conversion is a very slow process. Sampling of the atmosphere for both species has been conducted for the last 10 years, using both surface stations and aircraft. The resulting data have been used to estimate the lifetime of hydrogen in the atmosphere, the annual quantities of HT released, and the cycles of water and the HTO in the stratosphere.

The inventories of atmospheric tritium at the end of 1977 were 1.0 kg of HT which is rather uniformly distributed, and 5.3 kg of HTO. Most of the HTO, 5.1 kg, resides in the stratosphere, and 0.2 kg is in transit through the troposphere to the ocean surface.

ACKNOWLEDGEMENTS

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REFERENCES

DISCUSSION

L. FARGES: Do you have any idea of the importance of the $J_z$ term in your set of equations?

A.S. MASON: The present population of reactors and reprocessing plants is located almost entirely in the northern hemisphere, and we therefore consider that $J_z$ can be neglected in these estimates.

J.K. MIETTINEN: Have you estimated the rate loss of tritium from the atmosphere into space? This is after all the only loss that occurs.

A.S. MASON: According to present estimates of thermal escape, the loss appears to be negligible. Particles of more than two atomic mass units are not lost at rates sufficient to affect their overall budgets.
EVIDENCE OF PULSED DISCHARGES OF TRITIUM FROM NUCLEAR ENERGY INSTALLATIONS IN CENTRAL EUROPEAN PRECIPITATION

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Abstract

EVIDENCE OF PULSED DISCHARGES OF TRITIUM FROM NUCLEAR ENERGY INSTALLATIONS IN CENTRAL EUROPEAN PRECIPITATION.

The average geographical pattern of bomb tritium in Central European precipitation is derived on the basis of tritium observations regularly available since the early 1960s. The predominant feature is a steady increase in the tritium concentrations from the European west coast (coastal reference station: Valletta, 51°N, 10°W) towards the southeast, of up to a factor 3.5. This pattern is found to be constant in time up to 1970. Thereafter, the pattern changed in that larger and more scattered continental tritium concentrations are found relative to those at Valletta. The interpretation of this observation is that tritium in Central European precipitation until 1970 was dominated by bomb tritium, whereas the tritium excess observed for the inland stations since then results from tritium releases from European nuclear energy installations located in the western part of Europe and from use of tritium in consumer goods. The excess tritium concentrations are compatible with order-of-magnitude estimates of the tritium released by these sources. Furthermore, the time variation of tropospheric strontium-90 does not resemble that of tritium (which excludes the tritium originating from the stratosphere and/or from contemporary nuclear weapon testing) whereas krypton-85 (a product of nuclear fission) shows pulsed excesses very similar to what is observed for tritium. The strongest discharge event so far was observed in February 1975. Huge tritium pulses up to a factor of 40 higher than the local background of bomb tritium were simultaneously observed over a good part of Central Europe, and a simultaneous 85Kr pulse was observed at Freiburg. The excess tritium apparently increased towards the west and was maximal at Thonon (47°N,7°E). A tritium discharge of about 25 kCi is estimated for this month, whereas the excess tritium to krypton-85 ratio is near unity.

INTRODUCTION

Tritium is found in the environment mostly as radioactively labelled water HTO. Most of its present inventory has been provided by the atmospheric

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nuclear weapons testing between 1952 and 1962, the predominant injection into
the troposphere and, mostly, in the stratosphere, having occurred in 1961–62.
From there it has followed the hydrologic cycle and has gradually been entering
continental waters and the oceans, where today more than 95% of the world
inventory of bomb tritium is found [1]. Information on tropospheric tritium is
available on a large scale in the form of measured tritium concentrations in pre-
cipitation samples, largely from the IAEA/WMO Isotopes-in-Precipitation
Network [2–6]. Although precipitation samples have the disadvantage of being
discontinuous samples of tropospheric HTO, they have been preferred to water
vapour samples because of their great advantage of easy sampling [7–10]. Bomb
tritium has been distributed globally in the atmosphere, and therefore exhibits a
distinct geographical pattern in precipitation [9].

In recent years, nuclear energy installations for both civilian and military use
of nuclear energy have become an additional source for tritium [11]. Most of the
tritium produced in nuclear reactors does not enter the environment until the
nuclear fuel is reprocessed. A typical release rate during reprocessing is 660 Ci
tritium per tonne uranium [12]. With a reprocessing capacity of about 1000 t
annually [13], European reprocessing plants represent a powerful regional source
of tritium. Unfortunately, information about their actual work load and tritium
discharges is scanty. Another factor is that shore-based plants apparently dispose
of most of their tritium into the sea [14]. In addition, similar amounts of tritium
are released in the production, use and final disposal of consumer goods containing
tritium [15–17]. Whereas these discharges must be expected to affect the environ-
mental tritium concentrations, the problem is that the discharges may well be
masked by the 'natural' variability of the bomb tritium, so that the identification
of such discharges is difficult.

It is therefore the aim here to determine as well as is possible the geo-
graphical and time pattern of bomb tritium in Central European precipitation.
This information will then be used to identify tritium not originating from nuclear
weapon testing. Finally, the origin of this tritium is discussed, taking into account
also simultaneous measurements of $^{85}$Kr, which is almost exclusively a result of the
reprocessing of nuclear fuel [18], and of $^{90}$Sr, which virtually has a nuclear weapons
origin only.

2. TRITIUM IN WESTERN AND CENTRAL EUROPEAN PRECIPITATION

2.1. Data treatment

Routine tritium measurements in Central European precipitation have become
available since the late 1950s. In the early and mid 1960s, about 20 tritium net-
work stations were set up in Central Europe, which as a rule have provided tritium
concentrations in monthly composite precipitation samples, and have been
operating continuously since then [2–6] (unpublished data\(^1\) summarized in Ref.[19]). Distinct year-to-year trends, seasonal variations, and a general increase from the Atlantic coast into the continent, are apparent in these data. To quantify the pattern, long-term average tritium concentration ratios have been formed between pairs of stations. The location Valentia (51°N, 10°W), on the west coast of Ireland, has been used as a reference station, because it represents fairly undisturbed marine conditions. In the procedure [19] described previously [7, 8], geometric means are used, and a time average of the concentration ratio for any station \(x\), \(\bar{r}_x\), is obtained as

\[
\bar{r}_x = \frac{c_x}{c_{x\,\text{Val}}} = \prod_{i=1}^{N} \left( \frac{c_{x\,i} - c_0}{c_{x\,\text{Val}\,i} - c_0} \right)^{1/N}
\]

(1)

where the overbar stands for a geometric mean, and

\(c_{x\,i}\) = tritium concentration in composite precipitation sample for month \(i\) at station \(x\)

\(c_{x\,\text{Val}\,i}\) = same for reference station

\(c_0\) = North Atlantic surface water concentration, taken from Ref.[20]

\(N\) = total number of months

The apparent standard deviation of \(r_x\), neglecting \(c_0\) compared with \(c_x\) and \(c_{x\,\text{Val}}\), is given by

\[
\delta r_x = \bar{r}_x \left[ \exp (\delta \log r_x) - 1 \right]
\]

(2)

where

\[
\delta \log r_x = \left[ \frac{1}{(N-1)} \times \sum_{i=1}^{N} \left( \log r_{x\,i} - \bar{r}_x \right)^2 \right]^{1/2}
\]

(2a)

The geometric mean has been used to minimize the effects of highly different individual ratios that are sometimes found. Adding a factor \(\sqrt{1/N}\) to the right-hand side of Eq.(2a), and inserting into Eq.(2), one obtains \(\delta \bar{r}_x\), the standard error of \(\bar{r}_x\).

\(^{1}\) See acknowledgements.
Figure 1 shows the correlation of tritium concentrations between the stations Hof (50°N, 12°E) and Valentia. In view of the distance of about 1500 km between Hof and Valentia (see inset chart in Fig. 2) the correlation is surprisingly good. For all stations used in this work, the standard deviations of the monthly ratios ranged between about ±20% and ±50%, and the standard errors of the long-term mean ratios were typically about ±5% and in no case larger than ±10%. These are therefore typical figures of how well the tritium concentration for a given location, for an individual month or for a long period, can be estimated once the general tritium pattern is known. Results similar to those of the following section, but based on more limited data, have been published previously [7].

The tritium pattern before 1971

Figure 2 gives the tritium concentration ratios of all stations used in this study plotted versus distance from the reference station in a WNW to ESE direction. With the exception of the station Grenoble [19], a well-defined concentration increase from the coast into the continent of up to a factor 3.5 is found. This simple geographical pattern (continental effect [7, 9, 10], is a result of a fairly
FIG. 2. Inland increase of continental tritium concentrations over Western and Central Europe relative to those observed at the reference station Valentia, taken from observations up to 1970. Errors are standard errors of the mean ratios. The station locations are shown in the inset; projections of the station locations on the straight line are used as effective distance from the reference station in the graph. W gives the direction of the mean wind. The stations are:

(1) Valentia (51°N, 10°W); (2) Milford Haven (52°N, 5°W); (3) Wantage (52°N, 1°W);
(4) Oxtfordness (52°N, 2°E); (5) Lisbon (39°N, 7°E); (6) Le Mans (48°N, 0°W); (7) Groningen (53°N, 7°E); (8) Liege (51°N, 5°E); (9) Sindorf (52°N, 7°E); (10) Tastrup (57°N, 12°E);
(11) Huddinge (59°N, 18°E); (12) Limburger Hof (49°N, 8°E); (13) Grenoble (45°N, 6°E);
(14) Thônon-les-Bains (44°N, 7°E); (15) Stuttgart (49°N, 9°E); (16) Hof (50°N, 12°E);
(17) Zugspitze (47°N, 15°E); (18) Garmisch (47°N, 11°E); (19) Petzenkirchen (48°N, 15°E);
(20) Vienna (48°N, 17°E). The curve connecting the data points is a theoretical curve (taken from Ref. [19]) based on a two-layer one-dimensional regional analytical model of the troposphere.

stable tropospheric circulation over Europe with steady winds from westerly directions bearing water vapour from the North Atlantic marine troposphere. Such vapour therefore represents the starting point for any precipitation over Europe (cf. also Ref. [21]). The continental tritium increase is a result of tritium being continuously added from higher atmospheric levels and the continent being quite inefficient to remove this tritium compared with marine conditions.
Evidence that Valentia indeed represents fairly undisturbed open-ocean conditions comes from the fact that at Weathership J, nearly 700 km west of Valentia, the concentrations average only about 20% lower than those at Valentia.

Figure 3 gives the year-to-year trend in the tritium concentrations and the average seasonal variation. The yearly averages have been obtained by normalizing the monthly data from all stations to the same average tritium level using the long-term mean ratios \( \bar{r}_n \), and then linearly averaging all the renormalized monthly data available for the particular year. The year-to-year variation has been relatively minor since 1974. To obtain the seasonal variation, the long-term trend was first removed using a smoothed form of the year-to-year trend in Fig.3. From the corrected data, monthly to yearly mean ratios for each month were formed, which were then averaged over the period 1963–70. This procedure has been carried out for four stations, and the seasonal curve in Fig.3 is the average of the results for these stations — errors are the apparent 1–Sigma scatter in this latter averaging.
FIG. 4. Yearly averages of the ratio of tritium concentrations in precipitation at the stations Hof (50°N, 12°E) and Valentia (51°N, 10°W) between 1962 and 1976. Solid (dashed) line(s) give mean ratio (±2 sigma) between 1962 and 1970. The 1 sigma error is ±40%/√12 for a 12-month average.

There is a tendency for the seasonal variation to be somewhat less marked at the more continental locations, which also means that the continental increase (Fig. 2) is somewhat stronger in winter than in summer. This tendency, amounting to no more than 10%, is neglected in the present context, details being described elsewhere [19].

Figures 2 and 3 give the undisturbed geographical and time pattern of bomb tritium in Western and Central Europe. The geographical pattern apparently has been quite unchanged in time.

The changing tritium pattern from 1971 onward

Evidence for a changing tritium pattern in European precipitation after 1970 is presented in Fig. 4, which shows yearly average tritium concentration ratios for the stations Hof and Valentia for the period 1961–76. It is immediately obvious that after 1970 the inland station Hof averages much higher tritium concentration relative to the coastal station Valentia than in the earlier years, far beyond any reasonable error margins (compare also Fig. 1). If this cannot be explained by a completely changed tropospheric circulation — which would have had to be drastic enough to make news headlines — there cannot be any doubt that a regional source of tritium after 1970 is necessary to explain the findings. Valentia should be very much less affected by any such source because, as pointed out above, precipitation at Valentia represents fairly undisturbed marine conditions,
FIG. 5. Mean monthly concentrations of $^{85}$Kr [28, 29], tritium and $^{90}$Sr (unpublished Heidelberg data) at the stations Freiburg (48°N, 8°E; $^{85}$Kr), Hof (tritium), and Heidelberg (50°N, 9°E; $^{90}$Sr). The backgrounds for $^{85}$Kr (taken from Ref. [30]) and tritium are given as the shaded areas. The $^{85}$Kr background is the base level excluding the $^{85}$Kr peaks; the tritium background is that calculated from simultaneous measurements at Valencia using the concentration ratio of Fig. 2 ($= 2.74$), and making additional allowance for the uncertainty in this ratio by adding a 2-sigma standard deviation ($= 80\%$) amount, in order to give an upper limit, i.e.,

$$c_{\text{shaded}} = (c_{\text{Val}} - c_0) \times 1.74 \times 1.8 \text{ (}c_0 = \text{surface water conc., see Eq.(1))}.$$
and tritium on the scale of the North Atlantic troposphere is still dominated by bomb tritium [1]. The following sections discuss the source of the excess tritium in Europe observed after 1970.

**Phenomenology of the excess tritium**

Apart from the evidence of Fig. 4, the observation is that the monthly tritium concentrations become more scattered after 1970, and that the excess in large part occurs in single pulses. This is particularly so in 1975, for which year Fig. 4 shows the excess to be maximal, about five-fold what is expected for bomb tritium alone. Figure 5 for this year shows monthly tritium values for the station Hof, together with monthly composites of $^{85}$Kr and $^{90}$Sr from other locations in southwest Germany. Tritium during this year obviously exhibits a seasonal pattern quite unlike that of Fig. 3, with two peaks in February and September. Also shown are expected tritium concentrations (shaded area) obtained by converting observations for Valentia on the basis of Fig. 2. The difference between observed and expected concentrations can be taken to be an approximate representation of the excess tritium. For $^{85}$Kr an excess can likewise be defined. It is noted that the tritium excess at least for the first half of the year (see also below) resembles that of $^{85}$Kr, whereas the $^{90}$Sr variation is quite parallel to the expected tritium concentrations, as far as the bomb-produced fractions of both nuclides can be expected at all to vary in parallel [22]. The excess tritium, therefore, at least to a large part, must come from a source that also releases $^{85}$Kr but no $^{90}$Sr.

The February tritium peak was evident at 12 stations situated in an area 46° to 50°N and 6° to 16°E, and the peak in September at five stations between 48° and 50°N and 6° and 12°E but not at stations south of 48°N. This correlation excludes spurious measurements as the cause of the peak. A further conclusion is that the excess tritium follows certain trajectories and in a certain pulse has an extension of no more than a few hundred kilometres in a north-south direction. Weekly tritium data are available for a station in the Netherlands [23]. These show the February tritium peak to have occurred in the week 15–21 February, when also the $^{85}$Kr at Freiburg was maximal. There is evidence that this tritium was released in Western Europe, i.e. west of 6°E. The lowest excess was observed in the southeast at stations in Austria with values two to four times the background; from there it increased towards the northwest and west, and the highest value, about 40 times background(!), was obtained at the station Thonon-Les-Bains near Geneva. That the September tritium peak had no counterpart in $^{85}$Kr may be explained by the $^{85}$Kr station being in an area where also no excess tritium was present at that time (i.e. south of 48°N).
TABLE I. REGIONAL TRITIUM SOURCE STRENGTHS ESTIMATED FROM THE OBSERVED CONCENTRATION EXCESSES [19]

The conversion (see text) is 1100 Ci tritium per TU excess concentration
Uncertainty is at least a factor of 2

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<tr>
<td>Average excess tritium concentration (TU)</td>
<td>14</td>
<td>17</td>
<td>37</td>
<td>49</td>
<td>173</td>
<td>67</td>
</tr>
<tr>
<td>No. of stations when an excess was observed</td>
<td>10</td>
<td>14</td>
<td>22</td>
<td>22</td>
<td>22</td>
<td>22</td>
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<tr>
<td>Required tritium source strength (kCi)</td>
<td>15</td>
<td>19</td>
<td>41</td>
<td>54</td>
<td>190</td>
<td>73</td>
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DISCUSSION

We can exclude nuclear-weapon tests as the source of the excess tritium, on the basis of the regional geographical pattern of bomb tritium given in Fig. 2, as well as of the tritium-to-$^{90}$Sr correlation of Fig. 5, quite apart from the fact that no nuclear weapon tests are reported for 1975 [24]. There remain two potential sources for the excess tritium, i.e. nuclear energy installations and use of tritium in consumer goods. Only the first one of these gives $^{85}$Kr. Order of magnitude estimates of the total releases to be expected for 1975 are as follows: no more than 5 kCi from nuclear reactors, based on a tritium production of 18 kCi/GW(e) annually, less than 10% release from the reactors [18], and a total 3 GW(e) nuclear power production which is the combined figure for France and the Federal Republic of Germany for 1974 [25]; a maximum of about 700 kCi from nuclear fuel reprocessing plants, based on a capacity of about 1000 t uranium yearly (France and the Federal Republic of Germany [13]) and 0.66 kCi released per ton [12]; and about 60 kCi from use of tritium in consumer goods, which is an estimated figure for 1973 ([18], 15% assumed to be released from a total of 400 kCi produced). Both the reprocessing plant figure and that for consumer-goods tritium have considerable uncertainty; the uncertainty of the former is particularly large, because, as was pointed out above, some plants only dispose of a minor part of their tritium into the atmosphere, and because there is fuel reprocessing also for military purposes, on which no information is accessible. The expected releases from reactors certainly are negligible.

From the observed tritium concentration excesses, the required source strengths have been estimated as follows [19]: From plots such as in Fig. 5 (centre graph), the concentration excesses for each year were taken and were averaged over
all stations for which an excess was observed. In order to convert the average excess concentrations into released amounts, it was considered that the regionally released tritium is dispersed into whatever amount of water vapour passes through the area with the general west-east drift. The conversion was made by assuming that the excess tritium effectively is dispersed into one third of the amount of vapour that passes annually through a corridor taken to be 400 km wide, i.e. into about $3.4 \times 10^8$ kg of water [19, 26]. Table I lists the average excess concentrations and the resulting tritium releases.

A similar conversion of excess tritium concentrations into released amounts is obtained by using published atmospheric dispersion data ([27], Fig.6.11, extrapolated), and assuming the source to be located about 500 km west of the southwestern tip of the Federal Republic of Germany (i.e. upstream in the direction of the mean wind, see Fig.2 (inset chart)), where also the $^{85}$Kr station is located. For this area, the observations for February 1975 furthermore give an activity ratio near unity of excess tritium to excess $^{85}$Kr (tritium concentration interpolated for $^{85}$Kr station, 4g/m$^3$ water content of groundlevel air assumed, which yields $\sim 10$ pCi/m$^3$). This ratio is one order of magnitude larger than that expected for fission of nuclear fuel [18]. The total releases for this month are found to be about 25 kCi for both nuclides.

The source strength estimates of Table I are of a magnitude similar to the releases expected from both potential tritium sources. However, both the pulsed nature of the releases and the apparent correlation with $^{85}$Kr (Fig.5) point to nuclear energy installations rather than consumer-goods applications as the major source. On the other hand, the tritium-to-$^{85}$Kr activity ratio for the February 1975 event, being one order of magnitude larger than characteristic of nuclear fission, does not allow definite conclusions at this time. The true case may be discharge from a nuclear fuel reprocessing plant with considerable restraint of $^{85}$Kr, or, by coincidence, a smaller unfractonated release superimposed on a major escape of tritium in process for consumer-goods applications. Whereas the observed tritium-to-$^{85}$Kr activity ratio is characteristic of releases from nuclear reactors [18], the observed amounts could only be provided through releases by a major meltdown of fuel elements, to which again the unfractonated fission ratio would apply. Therefore, we can state that we have identified pulsed releases from one or more reprocessing plants of nuclear fuel, but that we cannot be sure that this has in fact been the major source of the excess tritium.

5. CONCLUSIONS

Tritium concentrations in Western and Central European precipitation exhibit a regular pattern until 1970 (Figs 2–4). Thereafter, a tritium excess is observed for the inland stations. The excess was largest in 1975, when it averaged a factor
of 5. Much of the excess tritium comes in pulses and completely masks the normal seasonal variations (Figs 3 and 5). From the pulsed nature of the excess tritium, from its correlation with $^{85}$Kr and with $^{90}$Sr, and from source strength estimates, it is concluded that the peaks, at least partly, originate from releases from reprocessing plants of nuclear fuel. Additional contributions of tritium from consumer-goods applications, however, cannot be excluded. A more appropriate sampling scheme would be highly desirable in order to resolve more fully the contributions of the various regional tritium sources in Europe.

The erratic geographic and time pattern of tritium in European precipitation observed since 1971, compared with the previous situation, is naturally a considerable drawback for applications of environmental tritium such as in hydrologic studies.

ACKNOWLEDGEMENTS

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REFERENCES

DISCUSSION

L. FARGES: What are the criteria you use in order to draw the line between the Valentia reference station and Central Europe?

W. ROETHER: The line is made to run parallel to the apparent tritium gradient. It deviates from the direction of the mean wind because the distribution of land and sea is such that the average land-sea border exhibits a very similar inclination to the mean wind, and the sea surface is a very effective sink for tritium.
ARTIFICIAL TRITIUM FALL-OUT
AT THE SOUTH POLE

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Abstract

ARTIFICIAL TRITIUM FALL-OUT AT THE SOUTH POLE.

Snow samples were taken from a 5-m-deep pit located near the South Pole station in January 1975, and continuous deuterium, tritium and beta activity profiles have been obtained from them. These three measurements and the stratigraphic level observation allow a precise chronology of the pit from 1950 to 1975 to be deduced, providing a continuous record of artificial tritium fall-out in the southern hemisphere. This has been extended to 1978 using samples from a second pit taken in 1978. Taking advantage of the unusual 1957–58 stable isotope content in the snow, it has been demonstrated that an important part of the isotopic signal in the precipitation is retained in the snow in spite of the low accumulation rate (8.2 g/cm²). The first artificial tritium, from the 1952 Ivy experiments, was detected during 1954. A two years delay between explosions and fall-out is well established, and enables the tritium fall-out to be related to the main nuclear tests from 1952 up to 1960. This delay appears longer for the large 1961–62 devices. For the French southern hemisphere experiments, it is about one year. A sharp tritium decrease is observed after a high 1973 peak, providing a new tritium reference level for future glaciological studies in Antarctica. The beta and tritium peaks occur during the Antarctic summer and the Antarctic winter respectively, showing different injection mechanisms. This winter input and the high tritium values registered at the South Pole indicate a preferential tritium transfer over the polar region. Two mechanisms, stratospheric-tropospheric exchange and direct stratospheric cloud precipitation, could account for this injection.

In January 1975, successive snow samples were taken in a 5-m-deep pit located near the South Pole station (Antarctica, Fig. 1). Continuous profiles of
deuterium content, tritium content and gross beta activity have been measured in this pit covering the period 1950–75, with a threefold purpose:

Determination of the snow core chronology to measure the accumulation rate;
Investigation of the artificial tritium and beta fall-outs;
Relation between deuterium and climatological data.

From the three profiles and the stratigraphic data observed in the field, a chronology of these samples has been determined. A continuous and precisely dated record has been obtained of the artificial tritium fall-out between 1954 and 1975 which appears to be the first well documented for the southern hemisphere.

A second 10-m-deep pit has been sampled in the same area from the beginning of 1978 giving the opportunity to extend the tritium record up to this year.
FIG. 2. Deuterium content versus depth (1975 pit).

FIG. 3. Tritium content at the precipitation date versus depth (1975 pit).
The experimental results necessary to deduce the snow pit chronology are here presented, with special attention to the tritium profile, in particular, to its relation with thermonuclear explosions and to fall-out mechanisms.

1. EXPERIMENTAL RESULTS

The deuterium and tritium measurements have been performed at Saclay and those of gross beta activity at Grenoble. The different isotopic profiles as a function of the samples depth are shown in Figs 2–7.
**FIG. 5.** Beta activity at the counting date (1975) versus depth (1975 pit).

**FIG. 6.** Deuterium content versus depth (1978 pit).
FIG. 7. Tritium content at the precipitation date versus depth (1978 pit).

The deuterium content is expressed in $\delta^{2}D_{\%}$ V.S.M.O.W. (Vienna Standard Mean Ocean Water with a $D/H$ ratio equal to 155.76 ppm) and is measured with a precision of 0.3% in the delta scale (one standard deviation).

The tritium values are given in TU (1 TU is equivalent to $T/H = 10^{-18}$ and $1 \text{ TU} = 3.24 \times 10^{-6} \mu\text{Ci/lt}r$) calculated at the date of precipitation. Down to a depth of 1.39 m the tritium analyses have been performed by direct measurement (liquid scintillation counting) with an experimental error of 12 TU at the time of measurement leading to errors up to 50 TU according to the age of the sample. At greater depths covering the 1951–58 period, the electrolytic enrichment procedure has been used allowing a precision of 0.6 TU and consequently better than 5 TU for the oldest samples. These last results, corresponding to the first artificial tritium fall-outs, are presented separately in Fig. 4.

The gross beta activity results are expressed in disintegrations per hour and per kilogram of water (dis/h per kg) at the counting date (1975) and are determined with a 10% precision.
2. SNOW SAMPLE CHRONOLOGY

The annual pattern of the stable isotopic content in precipitation (D and \(^{18}\)O) shows generally a summer maximum and winter minimum [1]. This is caused by the fractionation effects which occur during the condensation processes between the light (H\(_2^{16}\)O) and the heavy molecules (H\(_2^{18}\)O or HD\(^{16}\)O). This general feature has been observed in the South Pole precipitation by Aldaz and Deutsch [2] during the period November 1964 to October 1965. The South Pole climate is characterized by a sharp transition between a long winter (April to September) and a short summer (December and January) [3] during which the stable isotope peak occurs.

These seasonal variations are very well marked in the 1975 deuterium profile, and the attribution of successive summer maxima, as indicated in Fig. 3, is unmistakable between the surface level (January 1975) and the 1952 summer. In each case we have reported the year relative to the January month of each summer (for instance the number 74 corresponds to December 1973 and January 1974, i.e. the 1973–74 summer).
Summer and winter weather conditions are sufficiently contrasted to permit seasonal stratification to develop in the snow cover [4]. In the 1975 pit, the identification of each stratigraphic layer corresponding to a 'summer layer' has been possible, providing an independent dating of the pit which is found to be in exact agreement with the dating previously deduced from the deuterium measurements up to the 1952 summer, and shows clearly two years of snow accumulation between this level and the bottom of the pit (Fig. 8).

The artificial beta activity values in the Antarctic snow are characterized by two well-defined reference levels, in January 1955 [5] and January 1965 [6] due respectively to the arrival in the Antarctic area of the Castle series and of the 1962 nuclear test radioactive debris. These two reference levels appear markedly at the periods expected from the two previous dating methods. As previously reported [6, 7], a basic feature of the beta activity fall-out in the southern hemisphere is the appearance of a broad summer maximum and winter minimum. This seasonal pattern is displayed in the South Pole beta profile (Fig. 5), and some of these summer beta peaks have been assessed taking into account the preceding chronology. This is not possible between 1964 and 1968 as the large input of radioactive debris following the 1962 tests obliterates the seasonal variations during this period.

**FIG. 9.** Tritium content in the Halley Bay precipitation.
TABLE 1. $\delta^{18}$O EXTREMES AT THE SOUTH POLE DURING THE 1957–58 SUMMER AND THE 1958 WINTER

<table>
<thead>
<tr>
<th>Reference</th>
<th>(16) Pit 5</th>
<th>(16) Pit 3</th>
<th>(16) Pit 4</th>
<th>(16) Pit 1</th>
<th>(16) Pit 2</th>
<th>This work</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\delta^{18}$O maximum</td>
<td>-39.3</td>
<td>-44.7</td>
<td>-41.8</td>
<td>-41.7</td>
<td>-44.1</td>
<td>-42.8</td>
</tr>
<tr>
<td>$\delta^{18}$O minimum</td>
<td>-55.3</td>
<td>-47.0</td>
<td>-55.3</td>
<td>-56.4</td>
<td>-57.3</td>
<td>-55.6</td>
</tr>
<tr>
<td>$\delta^{18}$O amplitude</td>
<td>16.0</td>
<td>2.3</td>
<td>13.5</td>
<td>14.7</td>
<td>13.2</td>
<td>12.8</td>
</tr>
</tbody>
</table>

Finally, the tritium determinations provide a fourth independent method of obtaining chronological information. The mean monthly tritium data relative to the Halley Bay precipitation ($75^\circ$ 30' S) over the period 1966–76 (Fig. 9) exhibits the appearance of a main annual peak usually occurring during August or September in the southern hemisphere precipitation [8, 9]. This annual maximum appears very clearly in the snow pit up to 1954 with the exception of the 1955, 1961 and 1962 peaks which have been determined with respect to the deuterium and the stratigraphic data (Figs 2 and 4). The tritium profile gives an unambiguous chronology by itself at least up to 1963.

Each tritium peak corresponds to a low deuterium content sample formed of snow that fell during the Antarctic winter (April to September). It is difficult to deduce more accurately the period of the maximum tritium fall-out, but these results are in fair agreement with the Halley Bay data which show that tritium injection occurs mainly during August and September (Fig. 9).

To sum up, all these chronological data are reported on Fig. 8 with indications in each case of the annual maximum depth and of each summer layer level. Broken arrows relate to beta or tritium peaks determined with the help of the other data.

The exact coincidence between stratigraphic summer layers and deuterium maxima and the systematic shift between beta and tritium peaks may be observed and will be discussed below (paragraph 5.1). The main point of this figure is to prove very clearly that the 1975 pit chronology deduced from four independent methods is unquestionable. This is important for studying the artificial fall-out chronology and its relation with the tritium production.
The mean annual snow accumulation at the South Pole is equal to 8.2 g/cm² with extreme values ranging from 4.5 to 12.9 cm of water. This value is equivalent to the accumulation rates found in previous works [10–12].

The dating of the 1978 samples is not as straightforward as that for 1975. Figures 6 and 7 show the tritium and deuterium profiles and the stratigraphic levels are indicated on the horizontal scale. A discrepancy appears between the deuterium and stratigraphic data at the 0.41 m level, the deuterium maximum being ambiguous. However, the Halley Bay record (Fig. 9) as well as the tritium measurements in the 1975 pit (Fig. 2) show that the 1973 tritium peak is more important than the 1974 one, giving an indication in favour of the chronology established from the stratigraphic observations.

This dating, reported on Figs 6 and 7, has been chosen. However, an attempt will be made to confirm it by making deuterium tritium and beta measurements on deeper samples.

3. RELATION BETWEEN THE ISOTOPIC DISTRIBUTION IN THE PRECIPITATION AND A PROFILE MEASURED IN A SNOWPIT

Prior to a detailed investigation of the mechanism of arrival of tritium at the South Pole through the analysis of snow samples taken in a snowpit, it is necessary to establish how long an isotopic signal in the precipitation is retained in the samples of the pit.

Johnsen [13] and Dansgaard and co-workers [14] have developed a theory which foresees a smoothing of the stable isotope seasonal variations by diffusion in the vapour phase during firmification. This process would affect in the same way the tritium profile. In low accumulation areas (less than 20 g/cm² yearly), the obliteration of seasonal isotopic variations would occur over a few decades.

How fast this smoothing effect would occur at South Pole station has been investigated, taking advantage of the unusual isotopic signal corresponding to the 1958 year, which has been determined and reported at different times by various workers [15–17] and now measured in the 1975 pit. In all the previous works, the δ¹⁸O content has been determined and for comparison we have transformed our results in δ¹⁸O by the relation \( \delta D = 8 \delta ^{18}O + 10 \) [18]. Table I summarizes the different determinations of this 1958 isotopic signal.

With the exception of pit 3, it may be seen that a slight smoothing appears, but it is, for the 1975 pit, at most equal to 25% of the total amplitude measured just a few months after the time of precipitation, 20 years ago. From this it may be concluded that, at least for this interval of time, an important part of the isotopic signal in the precipitation is kept in the snow at the South Pole station.
4. TRITIUM FALL-OUT AND NUCLEAR EXPLOSIONS

Figures 2 and 7 show a well-dated distribution of successive artificial tritium fall-out at the South Pole between 1953 and 1977. A direct investigation of the mechanism of transfer of tritium in the atmosphere thus becomes possible by comparing the information shown on these figures with the calendar of the nuclear tests over the last 25 years.

A sharp maximum of tritium in the snow deposited between 1954 and 1962 is observed during the year 1960. As the maximum of the nuclide production over the period 1952–60 occurred in 1958 [19, 20], it may be concluded that the arrival of tritium at the South Pole is delayed by two years after its production located in the northern hemisphere (low latitude U.S. experiments, U.S.S.R. Arctic tests). Taylor [7] had previously indicated that the first tritium fall-out after an equatorial test series would be observed in Antarctica in the second spring season after injection. This is in fair agreement with our observations.

Figure 4 shows on an enlarged scale the tritium distribution, the deposition chronology of the snow samples in the deepest part of the 1975 pit and the calendar of the four main experiments between the first nuclear test in 1952 and 1956 [21]. The direct relation indicated on the figure between each nuclear test and the individual isotopic maximum has been established assuming a two years delay for the transport of artificial tritium in the atmosphere.

It may be seen for the first time that artificial tritium from the first Ivy explosions can be detected in the Antarctic snow deposited in 1954. An upper limit for the natural tritium concentration at the South Pole equal to 34 T.U. may then be derived.

When assessing the results shown on Figs 2 and 7, we observe a sharp decrease in the tritium fall-out after the 1960 peak corresponding to the moratorium observed for the nuclear tests in 1959 and 1960. Afterwards the very important U.S. and U.S.S.R. tests carried out between September 1961 and December 1962 provide a regular increase in the tritium concentration in the snow between 1963 and 1966.

The 1966 peak is the most important one, in the snow deposited at the South Pole as well as in the Kaitoke precipitation, even though no significant nuclear tests were carried out during the 1963–66 period. This indicates a different mechanism for the transfer of the tritium from the stratosphere to the troposphere possibly due to the higher altitude of the explosions for the 1961–62 tests in the stratosphere [7]. This gives rise to an apparent transit of the tritium up to four years between its stratospheric injection and its deposition.

The following important tritium injections have been produced by the Chinese tests carried out in the northern hemisphere (40° N) in 1967 giving the 1969 maximum.
The French tests in the southern hemisphere, from July to September 1968, were likely to have contributed also to the 1969 peak as no interhemispheric transport is involved in that case.

Two sets of data show the distribution of tritium in the snow deposited between 1970 and 1974 at two nearby places at the South Pole. The very high values, up to 1800 TU, measured in the 1975 pit do not appear in the 1978 one. A local variability could possibly be responsible for these differences. This has been proposed by Epstein and co-workers [15] to explain similar discrepancies between the stable isotopes distribution in two pits at the South Pole.

In any case, an attempt must be made to explain the very contrasted distribution of tritium in the 1975 pit which shows very high peaks in 1971 (1350 TU) and 1973 (1800 TU) in comparison with those observed in 1970 (450 TU), 1972 (390 TU) and 1974 (180 TU).

The 1971 peak can be ascribed to the simultaneous arrival of tritium produced by the 1969 Chinese (40° N) and the 1970 French tests (22° S), the low 1972 fall-out showing the one year delay observed for the French tests producing tritium. However, the available data concerning nuclear tests do not allow us to explain the appearance of the very large 1973 peak (equivalent to the 1966 one). Nevertheless, for the three cases in which the distribution of tritium in the precipitation is known (Halley Bay and the pits 1975 and 1978), a sharp decrease in the tritium concentration occurs after this high 1973 maximum. The low values measured up to 1978 could be possibly connected with less important French nuclear explosions in 1972, 1973 and 1974, followed subsequently by the cessation of atmospheric tests. In the same way, the Chinese explosions carried out in the Northern hemisphere after 1974 do not seem to introduce a large tritium fall-out in the southern hemisphere.

This 1973 tritium peak will therefore be considered as a new tritium reference level to be used in future glaciological studies in the South Pole area and possibly over the whole Antarctic continent.

5. FALL-OUT MECHANISMS

5.1. Beta and tritium peaks shift

The beta and tritium maxima occur successively at the South Pole during the Antarctic summer and the Antarctic winter (Fig. 8). A similar pattern for T and 89Sr fall-out in the southern hemisphere has previously been reported [7]. According to Taylor [7], the particulate products are concentrated after the nuclear tests in the lower stratosphere, then they mix southward in this stratospheric region and enter the troposphere at mid latitudes. On the other
hand, the tritium formed at upper levels is brought down from higher stratospheric altitudes within the circumpolar vortex, and then is passed into the troposphere over the polar regions during the Antarctic winter when no temperature inversion exists at the south polar tropopause.

In addition to a different seasonal injection, the South Pole results show a delay of approximately one year and a half between the bulk of the tritium and beta activity fall-outs. This value is based upon the shift observed between the 1955 and 1965 beta peaks and the 1956 and 1966 tritium maxima corresponding respectively to the Castle tests and to the major 1961—62 experiments [4].

An alternative mechanism is tentatively proposed to explain a preferential tritium fall-out in the Polar region during the Antarctic winter. According to Stanford [22] the average winter temperatures in the lower stratosphere are low enough to allow the direct growth of stratospheric clouds and their further precipitation in the higher troposphere. In this process, a significant percentage of tritium-rich stratospheric water could be removed from the stratosphere and account for large tritium injections during the Antarctic winter. The climatological data [3] show that 1973, when the very high tritium fall-out is not clearly related to the nuclear explosion calendar [4], is the coldest year (−84.5° C) at the 100 mb stratospheric level over the July-September period. This suggests that the stratospheric cloud formation mechanism could have played a particular role during this year.
5.2. Geographical variation of tritium fall-out in Antarctica

The previous study of the artificial tritium distribution in East Antarctica [23] has been extended to include results relative to the South Pole and Dome C stations (Fig. 1). Figure 10 shows the mean tritium content and the total amount of deposited tritium during the period from August 1956 – January 1972 versus the distance of each station to the coast. The altitude increases from D41 (z = 975 m) to Dome C (z = 3240 m) and at the South Pole z = 2800 m (Fig. 1).

A regular increase of the tritium content in the snow with the distance to the coast is observed as the maritime air masses initially depleted in tritium pick up high tritiated moisture in the upper part of the troposphere when moving from the coast towards the inner regions of Antarctica [23]. A significant altitude effect is ruled out as the tritium concentrations measured at the South Pole and D 100 are in a ratio of 2.4 notwithstanding that these two stations are at the same altitude (2800 m). Thus the mechanism of tritium transport does not appear similar to what has been observed in Greenland where the increase of tritium deposition has been attributed solely to an altitude effect [24].

When examining the variation in the total amount of tritium deposited it is interesting to note the specially high value measured at the South Pole station which may reflect a preferential local injection from the stratosphere.

This is in agreement with the previous observations deduced from the comparison between beta and tritium fall-out suggesting a specific tritium injection over the polar region.

ACKNOWLEDGEMENTS

We are very grateful to D. Mazaudier who carried out tritium measurements, to M. Lelu and G. Mondet for the deuterium analyses and to R. Chiron for his technical assistance. This work was supported in the field by Terres Australes et Antarctiques Françaises, Expéditions polaires françaises and the U.S. National Science Foundation (Office of Polar Programs).

REFERENCES

[22] STANDFORD, J.L., Possible sink for stratospheric water vapor at the winter Antarctic Pole, J. Atmos. Sci. 30, 1436.
L. FARGES: How do you carry out sampling in the snow column?

L. MERLIVAT: Samples were taken continuously from the pit walls, and then placed in separate vials. Core sampling was not possible because the material was friable and insufficiently dense.

F.A. PRANTL: I much enjoyed your presentation, particularly because I believe there is still so much to be learned from this type of work. You reported a time-lag of six months between the occurrence of the tritium and the total beta activity peaks in the pits. You attributed this to a difference in the atmospheric transport rates for tritium and for total beta activities associated with particles.

In similar work on Alpine glaciers we also found such a phenomenon. After considerable thought and investigation we concluded that the reason for this time-lag was that, during the summer, when the snow surface remains uncovered for several months, dry deposition of particles leads to a peak in total beta activities, whereas the tritium is deposited mainly during the winter with the fall of snow. Could this effect play a role in your case also?

L. MERLIVAT: That may be the case, but I believe that another mechanism may come into play here. You see, the time-lag seems to be about 18 months rather than six, as was indicated by the interval between the maximum of beta activity in 1965 and of tritium in 1966.
DISTRIBUTION AND REACTIONS OF TRITIATED HYDROGEN AND METHANE*

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Washington,
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Abstract

DISTRIBUTION AND REACTIONS OF TRITIATED HYDROGEN AND METHANE.

About 99% of the world's tritium has been converted to HTO and most is in the oceans. Of the sizable fraction in the atmosphere, most is in the stratosphere, again as HTO. However, in that portion of the atmosphere which is in dynamic contact with the earth's surface, the troposphere, most of the tritium is as HT with about 2% as CH₃T. The specific activity is high; the T/H ratio is greater than 10⁶ and 10³ that of seawater for hydrogen and methane respectively. In the case of hydrogen, this is two or three orders of magnitude higher than before the nuclear era. It is shown that the direct oxidation reactions to form water or the exchange reactions with water are negligible. The remaining mechanisms for conversion of HT and CH₃T are biological and photochemical. Both routes are considered to be important. For the former, the contributions from specific processes and their distribution over the earth are unknown. For the latter, the steps are known but the rate constants for many of the reactions in the complex photochemical sequence are not well known. The result is that we cannot accurately fix by direct calculation the lifetime of HT or CH₃T in the atmosphere. It is at least several years. In view of possible future accidental releases of tritium which may be large compared with the present world HT inventory, it seems desirable to extend our investigations of these 'inert' species.

INTRODUCTION

The natural inventory of tritium is about 1.1 × 10⁸ Ci (11 kg) of which about 90% is in the ocean as HTO. Man's activities (largely thermal weapons testing) have increased this to about 3 × 10⁹ Ci [1]. Although most of this has been transferred to the oceans, the environmental tritium is still an order of magnitude higher than before the nuclear era. Approximately 99% exists as HTO. The remaining 1% is mostly HT with a significant fraction as CH₃T.

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† Operated by Battelle Memorial Institute for the U.S. Department of Energy.
<table>
<thead>
<tr>
<th>Hydrogen</th>
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<th>Fixed H$_2$O (kg)</th>
</tr>
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<tbody>
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<td>H$_2$O</td>
<td>in H$_2$O/kg</td>
<td>in H$_2$O/kg</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oceans</td>
<td>1.3E 21</td>
<td>3.6E 19</td>
</tr>
<tr>
<td>Ice caps</td>
<td>(2.5 - 2.9) E 19</td>
<td>2.5E 16</td>
</tr>
<tr>
<td>Soil moisture</td>
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<td>Lower groundwater</td>
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<td>Rivers</td>
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<tr>
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</tr>
<tr>
<td>Stratosphere</td>
<td>3.1E 12</td>
<td>5E 10</td>
</tr>
</tbody>
</table>

a Organic compounds excluded, water data from Franka [2].
b Top 100 m of the ocean.
Future accidental releases of tritium will most likely be in the elemental form with some accompanying CH$_3$T. Since these releases can potentially be extremely large compared with the present inventory of HT, it is important to examine the present distribution of tritium and the reactions which convert these species to HTO.

The purpose of this study is to review the distribution of these species, and consider the rate of transformation to the more biologically active form, HTO.

DISTRIBUTION OF HYDROGEN

Table 1 shows the approximate distribution of free water, hydrogen and methane. The annual turnover of water is about 3.5 × 10$^{17}$ kg of which about 10% is river flow [2].

Fixed sources of hydrogen include anaerobic organic decay, photodissociation of H$_2$O and CH$_4$, and solar protons. It is removed by photo-oxidation, bacterial action, and planetary escape. The latter is relatively small, and Hunten [3] estimates the escape half-life at 10$^4$ a. The atmospheric concentration is normally placed at 0.5 ppmv (volume parts per million). Recent measurements have refined this value considerably to a slightly higher value [4--7]. Industrial emissions produce local variations and may also be increasing the total amount.

The amount of H$_2$ dissolved in the global water is uncertain, and the value listed in Table 1 is calculated from the equilibrium solubility. However, Schmidt's [4] surface seawater data showed amounts in excess of this value suggesting H$_2$ production in the water. Hydrogen is also produced by soil bacteria but, as will be mentioned later, most of it is probably consumed in other bacterial reactions.

The other hydrogen-containing molecule in significant quantity in the atmosphere is methane. It is also produced by bacterial action [8--12]. Scholz [13] found that its concentration varied from 1 to 1.4 ppmv in surface air to ≤ 0.05 at the stratopause. The most recent data of Ehhalt [14] indicates an average troposphere concentration of 1.35 ppmv with the southern hemisphere having a slightly lower concentration than the northern. The amount decreases with increasing height in the stratosphere resulting from oxidation to water.

TRITIUM DISTRIBUTION

Most of the weapons test tritium was deposited in the stratosphere as HTO. It leaks to the troposphere at a slow and variable rate. Extensive monitoring
<table>
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<th></th>
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<th></th>
<th>CH₄T</th>
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<tbody>
<tr>
<td></td>
<td>Ci</td>
<td>Conc, TU</td>
<td>Ci</td>
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<tr>
<td>Ocean, top 100 m</td>
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<td>10 – 20</td>
<td>(1 – 10) E3</td>
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<tr>
<td>Troposphere (3.8E18 kg air)</td>
<td>(2 – 8) E6</td>
<td>—</td>
<td>(8 – 18) E6</td>
<td>(3 – 7) E6</td>
<td>(6 – 20) E5</td>
<td>~ 5E4</td>
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<tr>
<td>Stratosphere (1.3E18 kg air)</td>
<td>(0.6 – 5) E8</td>
<td>(2 – 8) E7</td>
<td>(1 – 4) E6</td>
<td>—</td>
<td>&lt; 1E5</td>
<td>~ 5E4</td>
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</table>
indicates that the HTO content of water is highly variable, from around 1 TU\(^1\) in vapour above the ocean to 15 – 20 TU in the surface ocean to \(> 10^6\) in the stratosphere. The atmosphere has been extensively sampled by many workers [13 – 22]. In the stratosphere the HTO concentration varies with altitude and with latitude. In the troposphere the variation is even greater due to the rapid water turnover rate. The data of Östlund and Ehnhult are used for the estimates made in Table II. The analysis made by Schell and co-workers [16] of the global variations with time and location suggests that a reasonably complete description of HTO behaviour exists.

The concentrations for HT and for CH\(_3\)T are less certain because the monitoring data are much fewer. Östlund's [22] mean value of 44.6 HT molecules/mg air is used in the table. It should be noted that the tritium content of atmospheric hydrogen is greater than \(10^6\) TU, and that of methane greater than \(10^9\) TU. The tritium concentration in both compounds is much greater than before the nuclear era.

**REACTIONS OF ATMOSPHERIC TRITIUM**

**T\(_2\)-H\(_2\) exchange**

The reactions of hydrogen molecules are slow. The exceptions involve atoms or free radicals and thus catalysts or a radiation source are required. At high concentrations the tritium beta radiation can initiate the reaction, and Dorfman and Mattraw [23] found that at 0.4 mol fraction T\(_2\), half times of 4 – 14 h were observed for the reaction. At low concentrations, typical of tritium releases, the rate is extremely slow, but the normal excess of H\(_2\) would eventually convert all the T\(_2\) to HT.

**T\(_2\) \(\rightarrow\) HeT\(^+\) + beta reaction**

The natural decay of T\(_2\) will produce the \(^{3}\)HeT\(^+\) ion at a rate of about 11% annually. Wilzbach [24] first employed the recoil energy of the ion to label organic compounds. In the decay of T\(_2\) and HT, Wexler [25] has shown that the THe\(^+\) and HHe\(^+\) species, respectively, predominate. Subsequent steps produce nononuclear species with the charge preferring the helium nucleus. Cacace [26] published a comprehensive review of the decay products and reactions of tritiated molecules including hydrocarbons. Careful studies of the reaction with methane were made showing that CH\(_3\)T is the principal product.

\(^1\) The tritium unit, TU, is defined as a H/T ratio of \(10^{18}\) 1 kg tritium \(10^4\) kCi. 1μCi/litr air (STP) = \(17.1 \times 10^{10}\) atoms T/mg air.
Reactions of THe\(^{+}\) with H\(_2\) have not been reported. It seems reasonable to expect that reactions such as

\[
\text{THe}^{+} + \text{H}_2 \rightarrow \text{TH} + \text{H} + \text{He}^{+}
\]  

(1)

should lead to appreciable HT formation because of the lower zero point energy of HT compared with H\(_2\).

Information on the reaction with liquid water was obtained by Stöcklin and Cacace [27] in experiments designed to study the tritiation of thymidine and deoxyribose in aqueous solution. It was found that T\(_2\) was more effective than HT by a factor of about 12 at concentrations so low that radiation-induced mechanisms are not important.

No information on gas-phase reactions between THe\(^{+}\) and H\(_2\)O is available. It is clear, however, that at concentrations below those where radiation is no longer the dominant factor, the reactions of THe\(^{+}\) can still contribute significantly to formation of other tritiated molecules. If we define the lifetime of a molecule as the reciprocal of its first-order rate constant, \(k_1\), then \(k_1 = 2k_d\) where \(k_d\) is the radioactive decay constant. The lifetime or residence time, \(\tau\), of the T\(_2\) entity is thus \(1/k_1\) or 8.57a.

**T\(_2\)(HT) + H\(_2\)O exchange**

The water-hydrogen exchange reaction has been utilized for isotopic enrichment of deuterium. Equilibrium favours the heavy isotope in the water phase. For the gaseous reaction

\[
\text{HT} + \text{H}_2\text{O} \rightleftharpoons \text{HTO} + \text{H}_2, \quad K_{298} = 6.25, \quad K_{398} = 3.40
\]  

(2)

In the absence of catalysts, the reaction does not proceed [28]. However, a variety of materials, many metals and metal oxides in particular, permit more rapid equilibration especially at the higher temperatures. The catalytic process has been reviewed by Murphy [29] and Levins [30] and is still being studied for the separation of both deuterium and tritium [31–34].

Radiation is an efficient catalyst, and the early work of Yang and Gevantsman [35] appears to define the mechanism adequately. Using tritium diluted with helium, they found a rate of formation of tritiated water,

\[
\frac{dC}{dt} = 1.7 \times 10^{-3} (\text{Co})^2, \quad \text{Ci} \cdot \text{ltr}^{-1} \cdot \text{h}^{-1}
\]  

(3)

where Co is the initial T\(_2\) concentration in Ci/ltr over the range 0.05 to 0.7 Ci/ltr. The second order tritium dependence is a result of the first power radiation.
dependence. The interpretation was supported by X-ray irradiation tests which showed that the exchange rate was proportional to energy absorption.

Further information on the mechanism for the radiation-induced exchange has been provided by the work of Baxendale and Gilbert [36], by Firestone and co-workers [37,38], and by Bibler and Firestone [39] who studied the deuterium-water exchange at much higher hydrogen (or deuterium) concentrations. They found that at higher temperatures, above about 200°C, a chain mechanism involving hydrogen atoms was involved. Radiation-induced reactions are, in fact, similar to the atmospheric photochemical reactions discussed in a later section.

The hydrogen-water reaction is important because of the great excess of atmospheric moisture over hydrogen. Thus, at equilibrium, essentially all the tritium will exist as HTO. However, it is seen that in the absence of radiation or very effective catalysts the reaction rate is negligible, and for concentrations of hydrogen or tritium normally encountered in the atmosphere this would require many years.

**Oxidation of T2**

Like the previous reactions, the reaction between hydrogen and oxygen is negligible at ordinary temperatures unless catalysed. The same catalysts, as employed above, are generally effective, in particular the noble metals.

Eakins and Hutchinson [40] carried out a study of metal surfaces on the oxidation rate. Elemental tritium was mixed with wet or dry air, and the effects of mild steel, brass, platinum and aluminium surfaces noted. The rate constants for the experimental conditions of a surface-to-volume ratio corresponding to a typical laboratory (wall area/volume) varied from about $6 \times 10^{-4}$ ltr Ci$^{-1}$ h$^{-1}$ for brass to $2.4 \times 10^{-2}$ for platinum. The former value was the same as for the control with no catalyst. In humid air the rate constants were from 3 to 80 times larger.

The radiation-induced reaction has been studied by Dorfman and Hemmer [41], by Casaletto and co-workers [42], and by Belovodskii and co-workers [43]. Dorfman and Hemmer studied the reaction between T$_2$ and O$_2$ by direct pressure measurement, and found first-order kinetics with $k = 1.19 \times 10^{-4}$ min$^{-1}$ over a concentration range of 94 to 324 Ci/ltr. Casaletto and co-workers examined the reaction at lower concentrations. Below 1 Ci/ltr, they observed second-order kinetics with the rate constants $k = 1.2 \times 10^{-3}$ ltr Ci$^{-1}$ h$^{-1}$ in O$_2$ and $0.62 \times 10^{-3}$ ltr Ci$^{-1}$ h$^{-1}$ in dry air. The presence of water vapour in O$_2$ increased the rate by a factor of 3. The latter rate constant is seen to be in general agreement with that of Eakins and Hutchinson [40].

Belovodskii and co-workers [43] repeated some of the experiments with the system tritium-dry air-water vapour, and using a wider concentration range, $10^3$ Ci/ltr to 600 Ci/ltr. They fitted the data with a 5/3 concentration dependence.
They also observed no moisture effect, and concluded that the oxidation mechanism rather than exchange was dominant. The data presented earlier from exchange and oxidation studies lead to the opposite conclusion.

REACTIONS OF METHANE

At least 80% of the methane is of biological origin. Koyama [10, 11] carried out extensive studies of methane production in agricultural areas. Its production, utilization as a growth substrate and eventual oxidation in the atmosphere has been discussed by Schlegel [8], Levy [12], Junge [44], Ehhalt [9], Nicolet [45] and others. Ehhalt in 1974 [9] fixed the rate of production of CH₄ at $5.4 \times 10^{11}$ kg/a, of which about 80% was of biogenic origin. The surprisingly high T/H ratio in atmospheric methane raises questions both as to its origin and to its fate.

The studies of Caace [26] indicated the efficacy of the H⁺ mechanism for producing CH₃T. Earlier work of Kandel [46] on the radiation-induced reaction showed that the direct combination produced a maximum CH₃T concentration in about 12 h with peaks in the CH₂T₂ and CHT₃ concentrations at 25 and 50 h respectively. However, these reactions are effective only at high tritium concentrations.

The water-CH₃T exchange reaction was considered for purification of gas released from underground explosions [47]. No observable reaction was observed in the absence of catalysts. Such a reaction would not, of course, explain the enrichment over the TU level in water. Likewise, production of CH₂T by exchange of HT and CH₄ can be ruled out. Not only is such an exchange unlikely to occur at a measurable rate, but there appears to be no correlation between the rise in T content in H₂ and in CH₄ [15].

It appears likely that tritiated methane is a byproduct of any laboratory or industrial work involving tritium as suggested by Haines and Musgrave [48]. The situation is in many ways reminiscent of the methyl iodide problem in the nuclear industry — wherever radioactive iodine is involved, methyl iodide appears. In almost any real situation there is sufficient organic matter present (grease, paint, plastic, etc.) that CH₄I (or CH₃T) can be formed.

The principal destruction mechanism is photochemical oxidation in the atmosphere and is discussed later.

BIOCHEMICAL REACTIONS

As noted earlier, hydrogen is both produced and consumed by biogenic processes, and methane is produced in large quantities. Schlegel's review [8]
indicates the paths that involve hydrogen, methane and other key trace gases. Under aerobic conditions hydrogen and methane are quickly oxidized by microorganisms. However, under anaerobic conditions, methane is the principal product. As was indicated earlier, such processes should (assuming no highly specific isotopic selectivity by bacteria) produce CH$_4$T with the same T/H ratio as the water environment. However, it is conceivable that some atmospheric HT could be utilized thus producing a methane of higher T content.

One of the interesting questions is the isotopic selectivity of the biogenic processes involving hydrogen and methane. It is known that cells generally differentiate against the heavier hydrogen isotopes both in the production of hydrogen and in the incorporation of water into cell tissue.

Tritium studies in higher forms of life were carried out by Smith and Marshall [49] who indicated that low concentrations of an enzyme hydrogenase could cause oxidation of tritium. Smith and co-workers [50] found that intestinal bacteria from rats were effective in oxidation of tritium in the animal body.

Schmidt [4] concluded that biochemical production in surface ocean water and photochemistry are the major natural sources of hydrogen, and that utilization by soil bacteria provided the major sink. His calculations, based on field experiments on the uptake of H$_2$, gave an extrapolated loss rate of $1.1 \times 10^{11}$ kg/a. The correct figure may be lower still, but if it is of this magnitude, then biological activity in the soil will prove to be the predominant loss mechanism for H$_2$ and thus for HT.

PHOTOCHEMICAL REACTIONS

These are probably the most complex of the reactions considered. They involve free radicals in chain reactions and tie hydrogen and tritium to the very important carbon, nitrogen and sulphur cycles. A comprehensive survey cannot be made here, but several of the important reactions will be mentioned.

Table III lists some of the primary photochemical steps of importance. The radical reactions that follow primarily involve H, O, C and N species. Table IV lists about a third of the significant chemical reactions. The reaction rate constants are continually being revised, and the uncertainty factor listed is not an evaluation of the data but rather an indication of the magnitude of recent changes. Data are taken from Heichlen [51], McEwan and Phillips [5], Chameides [53], Davis [54], Luther and Deuever [55], and Crutzen [56].

Concentrations of a few of the reaction species can be measured. The remaining are calculated by setting up a model of the atmosphere involving those reactions assumed to be significant. Even for the one dimensional case (vertical) this is not an easy task, and the concentrations derived are only in fair agreement.
TABLE III. PHOTOCHEMICAL PROCESSES

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Minimum energy (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1 ( \text{O}_2 + h\nu \rightarrow \text{O}(^3\text{P}) + \text{O}(^1\text{D}) )</td>
<td>175.0 [59]</td>
</tr>
<tr>
<td>( \rightarrow 20(^3\text{P}) )</td>
<td>242.4 [59]</td>
</tr>
<tr>
<td>P2 ( \text{O}_3 \rightarrow \text{O}_2 + \text{O}(^1\text{D}) )</td>
<td>310 [60]</td>
</tr>
<tr>
<td>( \rightarrow \text{O}_2 + \text{O}(^3\text{P}) )</td>
<td>1180 [60]</td>
</tr>
<tr>
<td>P3 ( \text{H}_2\text{O} \rightarrow \text{OH} + \text{H} )</td>
<td>239 [59], 242 [52]</td>
</tr>
<tr>
<td>( \rightarrow \text{H}_2 + \text{O}(^1\text{D}) )</td>
<td>147 [61], 176 [60]</td>
</tr>
<tr>
<td>( \rightarrow \text{H}_2 + \text{O}(^3\text{P}) )</td>
<td>242 [60]</td>
</tr>
<tr>
<td>P4 ( \text{H}_2\text{CO} \rightarrow \text{H}_2 + \text{CO} )</td>
<td>360 [57], 350 [56]</td>
</tr>
<tr>
<td>( \rightarrow \text{H} + \text{CHO} )</td>
<td>360 [57], 350 [56]</td>
</tr>
<tr>
<td>P5 ( \text{HO}_2 \rightarrow \text{OH} + \text{O} )</td>
<td>270 [57]</td>
</tr>
<tr>
<td>P6 ( \text{H}_2\text{O}_2 \rightarrow 2\text{OH} )</td>
<td>565 [59], 577 [60]</td>
</tr>
<tr>
<td>P7 ( \text{CH}_4 \rightarrow \text{CH}_2 + \text{H}_2 )</td>
<td>160 [57]</td>
</tr>
<tr>
<td>P8 ( \text{CH}_2\text{OH} \rightarrow \text{CH}_2\text{O} + \text{H}_2 )</td>
<td>(~200) [56]</td>
</tr>
</tbody>
</table>

Besides variations caused by horizontal mixing, temperature and pressure, the uncertain rate constants lead to large differences in computed concentrations. Table V lists the approximate concentrations of the minor species. Values actually used in the various models of the atmosphere vary by a factor of 2 or 3 and often by an order of magnitude.

The path of natural tritium starts with photochemical oxidation of the T atoms produced in the stratosphere to HTO. It is interesting to note that even before atmospheric bomb testing the tritium content of hydrogen was greater than that of water. Hartkeck's [58] early paper explained this on the basis of competitive and repeated dissociation of TO₂ radicals, formed, for example, in reactions 5 and 11.

The oxidation of methane begins largely with reaction 18. This is followed by rapid oxidation of CH₃ which, through a series of steps involving oxy and peroxy radicals and formaldehyde, produces CO, CO₂ and H₂O. The cycle also produces H₂ by P-4 and P-8 and indirectly through 22 and 9. Thus, the methane which is produced at ground level is continually oxidized reaching an extremely low level in the upper stratosphere. Hydrogen, on the other hand, is both produced and removed by photochemical steps. Midway through the stratosphere H₂ is the dominant hydrogen species. Although most of the detailed calculations have concerned the stratosphere and above, the present problem requires that we
<table>
<thead>
<tr>
<th>Reaction</th>
<th>$k^a$</th>
<th>[Ref]</th>
<th>$k_{300K}^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. $\text{H}_2\text{O} + \text{O} (^1\text{D}) \rightarrow 2\text{H}_2\text{O}$</td>
<td>3.5E-10</td>
<td>[51]</td>
<td>2.3E-10 (0.7)</td>
</tr>
<tr>
<td>2. $\text{HO}_2 + \text{O} (^3\text{P}) \rightarrow \text{O}_2 + \text{H}$</td>
<td>4.2E-11</td>
<td>[51]</td>
<td>4.2E-11 (0.1)</td>
</tr>
<tr>
<td>3. $\text{HO} + \text{HO} \rightarrow \text{H}_2\text{O} + \text{O}$</td>
<td>1.0E-10 (-560/T)</td>
<td>[55]</td>
<td>1.6E-12 (0.5)</td>
</tr>
<tr>
<td>4. $\text{H} + \text{O}_3 \rightarrow \text{O}_3 + \text{H}$</td>
<td>1.23E-10 (-562/T)</td>
<td>[55]</td>
<td>1.9E-11 (0.7)</td>
</tr>
<tr>
<td>5. $\text{H} + \text{O}_2 \rightarrow \text{H}_2\text{O}$</td>
<td>2.1E-32 (-290/T)</td>
<td>[51]</td>
<td>5.5E-32 (3)</td>
</tr>
<tr>
<td>6. $\text{O} + \text{O}_2 \rightarrow \text{O}_3$</td>
<td>1.1E-34 (510/T)</td>
<td>[53]</td>
<td>6.0E-34 (0.4)</td>
</tr>
<tr>
<td>7. $\text{H} + \text{H} \rightarrow \text{H}_2$</td>
<td>2.6E-32</td>
<td>[59]</td>
<td>2.6E-32 (50)</td>
</tr>
<tr>
<td>8. $\text{H}_2 + \text{O} (^1\text{D}) \rightarrow \text{H} + \text{OH}$</td>
<td>2.9E-10</td>
<td>[51]</td>
<td>2.9E-10 (3)</td>
</tr>
<tr>
<td>8a. $\text{H}_2 + \text{O} (^3\text{P}) \rightarrow \text{H} + \text{OH}$</td>
<td>3.0E-14 (-4480/T)</td>
<td>[62]</td>
<td>9.8E-21 (3)</td>
</tr>
<tr>
<td>9. $\text{H} + \text{H}_2 \rightarrow \text{H}_2 + \text{H}$</td>
<td>6.8E-12 (-2020/T)</td>
<td>[53]</td>
<td>8.1E-15 (2)</td>
</tr>
<tr>
<td>10. $\text{HO} + \text{O}_3 \rightarrow \text{HO}_2 + \text{O}_2$</td>
<td>1.6E-12 (-1000/T)</td>
<td>[51]</td>
<td>5.7E-14 (0.2)</td>
</tr>
<tr>
<td>11. $\text{HO}_2 + \text{HO}_2 \rightarrow \text{H}_2\text{O}_2 + \text{O}_2$</td>
<td>1.7E-11 (-500/T)</td>
<td>[55]</td>
<td>3.3E-12 (0.5)</td>
</tr>
<tr>
<td>12. $\text{HO}_2 + \text{HO} \rightarrow \text{H}_2\text{O} + \text{O}_2$</td>
<td>1.7E-11 (-500/T)</td>
<td>[54]</td>
<td>3.7E-12 (5)</td>
</tr>
<tr>
<td>13. $\text{H}_2\text{O}_2 + \text{HO} \rightarrow \text{H}_2\text{O} + \text{H}_2\text{O}$</td>
<td>1.0E-11 (-750/T)</td>
<td>[55]</td>
<td>8.2E-13 (0.9)</td>
</tr>
<tr>
<td>14. $\text{HO}_2 + \text{O}_3 \rightarrow \text{HO} + \text{2O}_2$</td>
<td>1.0E-13 (-1250/T)</td>
<td>[51]</td>
<td>1.6E-15 (1.4)</td>
</tr>
<tr>
<td>15. $\text{HO}_2 + \text{O} (^3\text{P}) \rightarrow \text{O}_2 + \text{HO}$</td>
<td>8.0E-11 (-500/T)</td>
<td>[54]</td>
<td>1.6E-11 (2)</td>
</tr>
<tr>
<td>16. $\text{HO}_2 + \text{H} \rightarrow \text{H}_2 + \text{O}_2$</td>
<td>4.2E-11 (-350/T)</td>
<td>[51]</td>
<td>1.3E-12 (0.6)</td>
</tr>
<tr>
<td>17. $\text{CH} + \text{HO} \rightarrow \text{CH}_2 + \text{H}_2\text{O}$</td>
<td>3.83E-12 (-1850/T)</td>
<td>[53]</td>
<td>8.6E-15 (0.2)</td>
</tr>
<tr>
<td>18. $\text{CH}_2 + \text{O} (^1\text{D}) \rightarrow \text{CH}_2 + \text{HO}$</td>
<td>3.6E-10</td>
<td>[53]</td>
<td>3.6E-10 (3)</td>
</tr>
<tr>
<td>19. $\text{CH}_3 + \text{O} (^3\text{P}) \rightarrow \text{CH}_3 + \text{O}_2$</td>
<td>2.6E-31</td>
<td>[51]</td>
<td>2.6E-13 (0.2)</td>
</tr>
<tr>
<td>20. $\text{CH}_3 + \text{O} \rightarrow \text{CH}_3\text{O}_2$</td>
<td>1.1E-11 (-1000/T)</td>
<td>[55]</td>
<td>4.0E-13 (2)</td>
</tr>
<tr>
<td>21. $\text{HO}_2 + \text{NO} \rightarrow \text{NO}_2 + \text{H}$</td>
<td>2.05E-13 (-70/T)</td>
<td>[54]</td>
<td>1.6E-13 (0.3)</td>
</tr>
<tr>
<td>22. $\text{CH} + \text{O}_2 \rightarrow \text{CO} + \text{H}$</td>
<td>1.0E-13</td>
<td>[51]</td>
<td>1.0E-13 (0.7)</td>
</tr>
<tr>
<td>23. $\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$</td>
<td>1.7E-12 (-1350/T)</td>
<td>[52]</td>
<td>1.89E-14 (0.2)</td>
</tr>
</tbody>
</table>

*a* cm$^3$ particle$^{-1}$ s$^{-1}$, cm$^6$ particle$^{-2}$ s$^{-1}$ for 2 or 3 particle reaction, respectively. Numbers in parentheses indicate exponential term in the rate equation.

*b* Numbers in parentheses indicate approximate range of values (factor) recently reported.
<table>
<thead>
<tr>
<th>Altitude (km)</th>
<th>$O_3$</th>
<th>$O(^{4}D)$</th>
<th>HO</th>
<th>HO$_2$</th>
<th>H$_2$O$_2$</th>
<th>CO</th>
<th>CH$_3$O</th>
<th>CH$_3$O$_2$</th>
<th>H$_2$O</th>
<th>H</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>(5 – 10) E 11</td>
<td>2E 2</td>
<td>3E 6</td>
<td>8E 8</td>
<td>1E 11</td>
<td>3E 12</td>
<td>2E 10</td>
<td>4E 8</td>
<td>5E 17</td>
<td>–</td>
</tr>
<tr>
<td>10</td>
<td>(3 – 10) E 11</td>
<td>5E 2</td>
<td>2E 6</td>
<td>2E 8</td>
<td>3E 9</td>
<td>7E 11</td>
<td>4E 8</td>
<td>2E 7</td>
<td>3E 15</td>
<td>–</td>
</tr>
<tr>
<td>20</td>
<td>(3 – 50) E 11</td>
<td>1</td>
<td>1E 6</td>
<td>2E 7</td>
<td>1E 9</td>
<td>2E 11</td>
<td>5E 6</td>
<td>8E 12</td>
<td>E 1</td>
<td>4</td>
</tr>
<tr>
<td>40</td>
<td>(4 – 30) E 11</td>
<td>1E 2</td>
<td>1E 7</td>
<td>2E 7</td>
<td>4E 7</td>
<td>8E 9</td>
<td>4E 6</td>
<td>5E 11</td>
<td>E 4</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>~ 2E 6</td>
<td>1E 4</td>
<td>1E 7</td>
<td>6</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>5E 7</td>
</tr>
</tbody>
</table>

* TABLE V. MINOR ATMOSPHERIC SPECIES APPROXIMATE DAYLIGHT CONCENTRATIONS$^a$

*Molecules cm$^{-3}$*

$^a$ Data from Crutzen [56], Heichlen [51], Levy [12], Nicolet [45], and Shimaski [57]. The actual concentrations vary greatly depending on the time of day, the latitude and the season. The values listed are illustrative of the magnitudes involved.
also consider tropospheric reactions. The tropospheric chemistry is basically the same. Differences include greater concentrations of inert molecules, added complications from reactive pollutants, and reduced photochemical reactions because of diminished u.v.

Levy's [12, 63] model would oxidize about \(1.4 \times 10^{12}\) kg CH\(_4\)/a and produce H\(_2\) at about \(10^{11}\) kg/a making ground-level photochemistry of methane a major source of H\(_2\). It would also place the process as the major destruction mechanism for methane. McConnell and co-workers [64] and Warneck [65] have suggested that the OH concentration used by Levy was too high, and that the rates should be an order of magnitude lower. For comparison, the tropospheric CH\(_4\) removal rate deduced by Ehnhalt [14] was \(5.6 \times 10^{11}\) kg/a.

Schmidt's H\(_2\) production values [4] can be listed here: ocean \(4.0 \times 10^9\), photochemistry \((4.6 - 9.2) \times 10^9\), and anthropogenic \(1.3 \times 10^{10}\) kg/a.

The removal of H\(_2\) (and HT) may be expected by the same mechanisms as for CH\(_4\). Comparing reactions 8 and 10, it is seen that the OH reaction, 10, clearly dominates in the troposphere because of the greater OH concentrations compared with O(1D). Secondly, it has a high activation energy, and the rate increases by an order of magnitude in the warmer temperature of the lower troposphere. A calculation similar to that for methane gives roughly \((3 - 30) \times 10^9\) kg H\(_2\)/a removed.

**Residence times**

From the atmospheric inventory and rate data on either a source or sink, a residence time can be computed. The value for H\(_2\) and hence HT, based on photochemical removal reactions, is about 5 – 50 a, and based on soil bacterial reactions about 6 years. Junge estimated it at \(6.5 \pm 2\) a [66]. Mason [5], using recent data from the northern and southern hemispheres, and an estimate of the hemispheric air exchange rate, placed the value at 6 – 10 a.

The residence time for CH\(_3\)T has about the same uncertainty. The problem is probably simpler than for HT, but there are fewer data. Nicolet [45] lists 3 a. The calculations of Levy [12] gave 2 a whereas Ehnhalt [9] fixed it at 4 – 7 a. More recent calculations of Ehnhalt [14] place the limits at 3.5 and 11 a.

**SUMMARY AND CONCLUSIONS**

Over 50% of the tritium in the troposphere is as HT with a few per cent as CH\(_3\)T. The total HT is of the order of \(9 \times 10^6\) Ci. Although the total hydrogen content of the atmosphere may be increasing, the relative tritium content has gone up much faster. The data available for Begemann’s [67] estimate in 1963 showed the values for hydrogen two to three orders of magnitude lower, and those
for methane about a factor 10 lower than the present levels. The uncertainties
listed in Table II are probably extreme, and new data from Östlund [18] indicate
that considerable refinement will soon be possible.

It is important to note that $9 \times 10^4$ Ci ($\sim 900$ g T) is a relatively small
amount and represents, for example, only a fifth of the inventory of a 1000 MW
fusion reactor. In a fusion power economy the behaviour of accidental releases
of $^3$H may be a significant question.

Little data exist to test the conversion rates of tritium released to the
atmosphere. Of three well-monitored accidental releases of $^3$H [69 – 70], only
the last produced a concentration, 0.03 Ci/ltr, barely high enough to permit
an estimate of the conversion to HTO. The calculated amount formed during
the release period is well below the normal concentration of HTO in released tritium.

At very high concentrations of $^3$H or HT (> 10 Ci/ltr) the beta radiation
energy can promote both exchange with hydrogen or water vapour and oxidation
to water. However, in any real release event, rapid dilution occurs. The two
removal of conversion processes remaining are bacterial action and photo-
chemical oxidation for HT and photochemical oxidation alone for CH$_3$H. Both
appear important, but the contribution from each cannot be accurately fixed.
For the biogenic processes the uncertainties are the specific reactions and their
variations over the surface of the earth. For the photochemical processes the
uncertainties are largely in the rate constants for several key radical reactions.

The tropospheric residence times of HT and CH$_3$H are similar and are
probably of the order of 4 – 12 years. No data exist for $^3$H. The reactivity of
the decay species, T$^3$H, will limit its lifetime to about 9 years, i.e. half of the
tritium decay lifetime. The long lifetime of HT affords a measure of biological
protection. However, it suggests that the HT and CH$_3$H may continue to increase
in the atmosphere.

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DISCUSSION

M.H. CHEW: As stated in your paper, the world tritium inventory is about
$3 \times 10^6$ Ci, and you say also that future accidental releases may well be very
large compared with the present inventory. Can you give us any examples of
such releases?

L.L. BURGER: I was making my comparison with the tritium in the
troposphere, which is only $10^7$ Ci, not $10^6$. The accidental release I postulated
was $5 \times 10^7$ Ci yearly. This may seem very large, but if and when a fusion
power economy comes into being, tritium production and storage will be
greater than at present by whole orders of magnitude. We should note that the inventory in a single fusion reactor may be as large as $2.4 \times 10^8$ Ci. The design characteristics of fusion reactors suggest very low normal releases ($< 10$ Ci/d), but accidental releases are another matter. The largest single documented release in elemental form has been about $5 \times 10^5$ Ci, and the total release annually at present is probably greater than $10^6$ Ci. With these numbers in mind future releases of $5 \times 10^7$ Ci yearly, while not expected, do not seem inconceivable.

T. SHIOKAWA: With reference to the decay of $T_2$ and HT, about 20 years ago Wexler and myself estimated the THe$^+$ ion only by the mass spectrometric technique. Recently, however, we have applied a computer treatment based on molecular orbital theory and have found that the lifetime of THe$^+$ is theoretically very long.

May I suggest a correction to Eq.(1) in your paper: it should read

THe$^+ + H_2 \rightarrow TH + H^+ + He$, because the ionization potential of helium is much higher than that of hydrogen. This fact is most important for a correct understanding of this reaction.

L.L. BURGER: I wrote the equation for the species formed from the decay of $T_2$ showing the charge on the helium nucleus instead of on the hydrogen because, as I recall, Wexler's measurements indicated that that was the preferred distribution — in spite of the fact that ionization energies predicted the opposite. However, the point I wanted to make was that HT should form from hydrogen. I was not so much concerned with the distribution of charges. Your calculations on the stability of the THe$^+$ ion are of considerable interest. I was not aware that this had been done.

V. KASCHE: It has recently been found that nitrogen-fixing bacteria may simultaneously oxidize hydrogen or HT. Have you considered whether this process in the soil may influence the TH transport characteristics in the biosphere?

L.L. BURGER: I have not considered the individual contributions of different bacteria. I did refer to Schlegel's work, in which he identified various bacterial reductions involving hydrogen. These included the reduction of nitrate, sulphate and carbon dioxide. I was not aware of hydrogen utilization in nitrogen fixation, but it should indeed be a significant factor.

T.R. THOMAS: The world's environmental inventory of $^3$H being about $3 \times 10^8$ Ci, as we have heard, would the release of, say, $10^6$ Ci of $^3$H annually from a 2000 t/a LWR reprocessing plant have a significant impact in terms of health effects, bearing in mind the postulations based on the existing inventory? I ask this question because it has been proposed to electrolyse the waste water from reprocessing plants and release $^3$H as HT.

L.L. BURGER: Tritium released from a reprocessing plant is, of course, 99% HTO. Your question is whether this could be released if it were converted to HT. I do not feel qualified to say what the environmental effects would be, but looked at from the point of view of numbers alone it would seem to be a reasonable thing to do. It certainly does not make much sense in such a plant to attempt conversion of small residues of HT to HTO.
TRITIUM POLLUTION IN THE SWISS LUMINOUS COMPOUND INDUSTRY

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Abstract

TRITIUM POLLUTION IN THE SWISS LUMINOUS COMPOUND INDUSTRY.

The Swiss luminous compound industry is an important consumer of tritium. About 350 kCi go into production of tritium gas-filled light sources and 40 kCi into production of tritium luminous compound annually. To illustrate the pollution problem, a factory is mentioned that handles 200 kCi annually and a chain of luminizers, processing 20 kCi over the same period as tritium luminous compound. This material is manufactured by coating phosphors with tritiated polystyrene having a specific activity up to 200 Ci/g. Because of the high specific activity, the radiation damage produces an average activity release of 5.2% annually, which is one of the main reasons for public and occupational exposure. The processing of large quantities of tritium gas requires special equipment, such as units made entirely of stainless steel for purification and hydrogenation, oxidation systems for highly contaminated air, glove boxes, ventilation and monitoring systems. Nevertheless, contamination of air, surfaces, water and workers cannot be avoided. Only in a few cases were MPC-values for tritium content in urine of workers exceeded. From these results, biological half-lives between 5–15 days were estimated. Regular medical examinations showed no significant influence in blood picture parameters, except in one single case with a tritium concentration in urine of 2.8 mCi/litre. Entirely different problems arise in most luminizing factories where luminous paint is processed as an open radioactive source.

INTRODUCTION

Tritium luminous compounds have been produced in Switzerland since 1962. After years of technical developments, a most effective form of tritium luminous compound, a polymer of tritiated polystyrene, was established, obtained by ordinary hydrogenation and subsequent polymerization. Special sensitive phosphors of the zinc sulphide type are coated with a thin layer of about 0.01 μm of tritiated polystyrene having a specific activity of about 200 Ci/g. The proper running reaction to obtain the monostyrene base product, the radiation resistance of the polymer, its stability and favourable solubility properties, when combined with different comonomers, were the factors influencing the choice of this compound for use in luminous materials.

In the early 1960s tritium was still in strong competition with other isotopes regarding its suitability for luminous material, mainly radium-226 and promethium-147. Hunzinger [1] has published 1972 figures on the use of various isotopes in Swiss
TABLE I. PRODUCTION AND USE OF TRITIUM LUMINOUS COMPOUND IN SWITZERLAND (kCi)

<table>
<thead>
<tr>
<th>Year</th>
<th>1962</th>
<th>63</th>
<th>64</th>
<th>65</th>
<th>66</th>
<th>67</th>
<th>68</th>
<th>69</th>
<th>70</th>
<th>71</th>
<th>72</th>
<th>73</th>
<th>74</th>
<th>75</th>
<th>76</th>
<th>77</th>
</tr>
</thead>
<tbody>
<tr>
<td>Production</td>
<td>3</td>
<td>18</td>
<td>18</td>
<td>22</td>
<td>24</td>
<td>41</td>
<td>40</td>
<td>78</td>
<td>72</td>
<td>54</td>
<td>47</td>
<td>42</td>
<td>33</td>
<td>38</td>
<td>33</td>
<td></td>
</tr>
<tr>
<td>Processed</td>
<td>2</td>
<td>16</td>
<td>10</td>
<td>13</td>
<td>18</td>
<td>32</td>
<td>34</td>
<td>44</td>
<td>45</td>
<td>34</td>
<td>30</td>
<td>29</td>
<td>30</td>
<td>18</td>
<td>15</td>
<td>16</td>
</tr>
</tbody>
</table>

watches, and demonstrated that in 1971 practically only tritium was used. The quantity of tritium processed during the last 16 years by the Swiss luminous compound industry is fairly substantial, as shown in Table I.

Since 1976, tritium luminous compounds have been in competition with another kind of illumination system for watches, the so-called Beta- or Microlights, tritium gas-filled glass tubes, coated internally with phosphor. Special LCD watches are commonly illuminated with these tubes.

In Switzerland there are two producers of tritium luminous compounds, about 50 luminizing factories, many watch factories completing watches with luminous dials and hands. Altogether about 200 persons have regular contact with tritium activities in the form of luminous material. The characteristics, quality and control methods for the tritium luminous compound as well as the total activity content of the finished products (luminous watches and instruments) are strictly limited by different national and international laws and recommendations [2–4].

The radiation damage from tritiated polystyrene, followed by the release of tritium from luminous compound in the form of tritium gas and various organic radicals, with possible oxidation to tritium water, are one of the main causes of public and occupational exposure. A tritium release of 4.5–8.5% is calculated with an average value of 5.2% annually, depending on the specific activity and quality of the tritiated polystyrene.

During the last ten years data have been collected on pollution measurements in laboratories and the surroundings of a tritium luminous compound manufacturer, and at the same time an analysis has been made of the data of occupational exposure in the Swiss luminizing industry.

PRODUCTION OF TRITIUM LUMINOUS COMPOUND AND CORRELATED PROBLEMS

The radioactive base product for tritium luminous compound, tritiated monostyrene, is obtained by catalytic hydrogenation of phenylacetylene. This reaction runs very well if the tritium gas available is free from impurities, particularly from helium-3 which blocks the surface of the catalyst. Therefore, the incoming raw
material in gaseous form is purified by circulation over pyrophoric uranium pots. The non-absorbable residues of the gas are then pumped off to be burned in a copper-oxide furnace. The tritiated water is absorbed on a molecular sieve column and the gas mixture emitted is almost free from tritium contamination. By oxidation of tritium-contaminated air the environmental pollution can be reduced by a factor of up to 1000.

After purification and absorption of tritium gas, the uranium pots are connected to the hydrogenation unit. This vacuum unit, also built entirely of stainless steel, has a capacity of 2500 Ci. Some 1500 Ci are worked up in one lot. The end of the hydrogenation reaction is determined by calculation of the applied quantity of phenylacetylene, starting pressure, temperature and volume of the unit. The residual tritium is pumped back and absorbed on the pyrophoric uranium pots by means of a Toepfer pump. The active monostyrene obtained is distilled under high vacuum on to inactive monostyrene and comonomer. The purity of the styrene is controlled by gas chromatography.

Average impurity values of 1 to 2% ethylbenzene and phenylacetylene are found. During the catalytic hydrogenation of phenylacetylene, ethylbenzene occurs before all the phenylacetylene is hydrogenated to styrene. This initial analysis already provides preliminary information about the expected potential radiation hazard characteristics. The higher the impurity values, the worse the degree of polymerization and the stability of the polymer. On the other hand, ethylbenzene cannot be built into the polymer chains and will remain a slightly volatile component. Therefore, the polymer after thermal polymerization and
dissolution is precipitated in methyl alcohol. By this step, ethylbenzene, phenylacetylene and readily volatile oligomers of styrene, which are the most hazardous components, are eliminated from the polymer. The precipitate is then again dissolved in toluene for activation of the phosphors. The coating process of specially treated zinc sulphide grains consists of moistening the inorganic crystals with tritiated copolymer solution, and drying in a specially designed stirring machine. The latter is connected to a cooling trap to retain the solvents. To maintain a fine particle size the powder is sieved before the final inspection. The regular quality control includes checking of the brightness of powder material as well as of the applied compound. Luminosity measurements are made on a photometer with special correlation filters, adapted to human eye sensitivity. As shown in Fig.1, the brightness efficiency of applied paint, e.g. powder material mixed with suitable adhesives, is in the range of 20 to 25%. The choice of the adhesive is also an important radiation protection factor. Therefore only lacquers recommended by the luminous compound manufacturer should be used, since these will not attack the active polystyrene coating and create unnecessary danger of contamination with dissolved tritiated material.

In addition, the recommended adhesives are fully tested for good flexibility, adhesiveness, corrosion and water resistance to avoid chipping of paint. Immersion or soak tests in distilled water, and the percentage determination of extracted activity, are one of the most important parameters concerning radiation protection. The extracted activity from luminous compound depends widely on the specific activity and the adhesives used. The average values vary between 0.05 and 0.10%, compared with a tolerance of 5% according to Radiation Protection Standards for
Radioluminous Timepieces [2]. The determination of tritium release to the air from applied luminous material, caused by radiation damage, is made with an ionization chamber connected to the sample by a closed circuit. Our findings were similar to those of other authors and to official measurements made by the Swiss Laboratory for Watch Making Research (LSRH) — activity releases of 4.5–8.5% annually, depending on the specific activity involved, with an average value of 5.2%.

Because of the radiation damage and radioactive decay of tritium, a decrease in luminosity is obviously inevitable. The influence of specific activity on the brightness decrease is shown in Fig.2. Apart from radiation parameters, heavy damage of zinc sulphide crystals may be caused by the u.v. of sun-rays and air humidity. Such reduction of zinc sulphide to elementary zinc also influences the decrease in luminosity. This effect, however, may be reduced by special treatment of the phosphor.

After completion of quality control, the concentrated grades of the luminous pigment are blended down to any desired activity or brightness level (Fig.1) and other colours can be produced by addition of colouring agents. Uncoloured yellowish or white luminous compounds are normally suggested, because of their economic luminosity efficiency.

LABORATORY EQUIPMENT, WORKING METHODS, CONTAMINATION LEVEL AND COUNTER MEASUREMENTS, OCCUPATIONAL EXPOSURE AND ENVIRONMENTAL POLLUTION IN A LUMINOUS COMPOUND FACTORY

From 1962, when regular production of tritium luminous compound began, there has been a steady increase in tritium gas consumption, as shown in Table II. Apart from the production of luminous compound, purification and transfer on pyrophoric uranium pots, tritium labelling of organic substances and research work are also carried out by the company. With the increase in tritium consumption, all the contamination problems had to be solved. Since 1970 the whole tritium process takes place in a separate building specially designed for this purpose.

All the rooms are connected to an efficient ventilation system with an air exchange 18 to 30 times hourly, regulated by air monitors. Every production step is carried out in a separate exhaust glove box, connected to the ventilation system, so that operators are never in direct contact with radioactive material. The main problem is the diffusion of tritium gas through rubber gloves and the plexiglass windows of the boxes.

To eliminate the risk of serious air pollution caused by pumping out the vacuum units, an additional gas circulation system was installed. A Toepler pump injects or circulates gas residues through pyrophoric uranium pots. By this method
### TABLE II. AMOUNTS OF TRITIUM PROCESSED SINCE 1962

<table>
<thead>
<tr>
<th>Year</th>
<th>Amount (kCi)</th>
<th>Year</th>
<th>Amount (kCi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1962</td>
<td>14</td>
<td>1970</td>
<td>75</td>
</tr>
<tr>
<td>1963</td>
<td>14</td>
<td>1971</td>
<td>95</td>
</tr>
<tr>
<td>1964</td>
<td>12</td>
<td>1972</td>
<td>150</td>
</tr>
<tr>
<td>1965</td>
<td>20</td>
<td>1973</td>
<td>145</td>
</tr>
<tr>
<td>1966</td>
<td>23</td>
<td>1974</td>
<td>165</td>
</tr>
<tr>
<td>1967</td>
<td>27</td>
<td>1975</td>
<td>80</td>
</tr>
<tr>
<td>1968</td>
<td>28</td>
<td>1976</td>
<td>93</td>
</tr>
<tr>
<td>1969</td>
<td>65</td>
<td>1977</td>
<td>183</td>
</tr>
</tbody>
</table>

### TABLE III. TRITIUM CONTENT IN (A) URINE OF OPERATORS; (B) CANALIZATION WATER; AND (C) METEOR SAMPLES

(A) Tritium content in urine of operators

<table>
<thead>
<tr>
<th>Occupation</th>
<th>$^3$H-content in urine ($\mu$Ci/litre)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Working in high active area</td>
<td>10–25</td>
</tr>
<tr>
<td>Mechanic</td>
<td>6–25</td>
</tr>
<tr>
<td>Dispatch</td>
<td>3–7</td>
</tr>
</tbody>
</table>

(B) Average tritium content in canalization water ($\mu$Ci/litre)

| Leaving factory area | 0.3 |
| Leaving municipal purification plant | 0.004 |

(C) Average tritium content of water and snow samples ($\mu$Ci/litre)

| Water and snow samples 50 m from exhaust roof of tritium building | 0.1 |
| Water and snow samples 200 m from exhaust roof of tritium building | 0.02 |
| Water and snow samples 2000 m from exhaust roof of tritium building | 0.001 |
### TABLE IV. INDIVIDUAL BIOLOGICAL HALF-LIVES FOR TRITIUM-EXPOSED OPERATORS

<table>
<thead>
<tr>
<th>Worker</th>
<th>Type of contamination</th>
<th>Urine (µCi/litre)</th>
<th>Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Vacuum-pump oil</td>
<td>50</td>
<td>7.6</td>
</tr>
<tr>
<td>B</td>
<td>Release from luminous compound</td>
<td>20</td>
<td>13</td>
</tr>
<tr>
<td>B</td>
<td>Release from luminous compound</td>
<td>30</td>
<td>8</td>
</tr>
<tr>
<td>C</td>
<td>Organic vapour</td>
<td>35</td>
<td>7</td>
</tr>
<tr>
<td>C</td>
<td>Release from luminous compound</td>
<td>20</td>
<td>9.3</td>
</tr>
<tr>
<td>D</td>
<td>Release from luminous compound and $^3$H$_2$-gas</td>
<td>20</td>
<td>12</td>
</tr>
<tr>
<td>D</td>
<td>Release from luminous compound and $^3$H$_2$-gas</td>
<td>22</td>
<td>12.5</td>
</tr>
<tr>
<td>E</td>
<td>Release from luminous compound and $^3$H$_2$-gas</td>
<td>5</td>
<td>15</td>
</tr>
</tbody>
</table>

much more tritium can be reabsorbed in a shorter time to pyros. The unabsorbable gas residues will be burned to water after pumping through a copper oxide oven. The tritium water is almost quantitatively absorbed on a molecular sieve column.

In view of the considerable amounts of tritium worked up in these production laboratories, internal contamination of persons can hardly be avoided. Weekly urine measurements are carried out, with additional examinations, if excessive contamination is likely to have occurred. For persons involved in the normal production process for a daily period of some four hours on average, the values vary between 7 and 20 µCi/litre urine. To keep the level as low as possible, the following precautionary steps are undertaken: clothes and shoes must be changed, woollen gloves are worn inside rubber gloves, weekly decontamination with detergent and water of all laboratories is carried out together with regular wipe test controls.

Surface contamination levels are in the range of $10^{-3}$ µCi/cm$^2$. Only regular cleaning maintains the values. The waste water collected is checked for its tritium content, solid particles are filtered off and the water is diluted to 0.3 µCi/litre before transfer to public canalization. There are about 20 litres of contaminated water weekly.

In addition to our own measurements, the official health protection authorities check operators, canalization and rain-water, and also measure activity of rain and snow in the surroundings. Some of these results are collected in Table III.

By analysis of the results of tritium concentration in urine, we have estimated individual effective half-lives for body burden tritium. Operators exceeding the maximal permissible level are no longer exposed to tritium activity in order to
<table>
<thead>
<tr>
<th>Examination</th>
<th>Number of cases</th>
<th>Average value</th>
<th>Range</th>
<th>Normal value for non-exposed persons</th>
<th>Person with reversible tritium panmyelopathy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Haemoglobin</td>
<td>165</td>
<td>93.4%</td>
<td>76–113%</td>
<td>80–95%</td>
<td>47%</td>
</tr>
<tr>
<td>Erythrocyte</td>
<td>163</td>
<td>4.55 Mio/mm³</td>
<td>3.7–6.0 Mio/mm³</td>
<td>3.5–4.5 Mio/mm³</td>
<td>2 Mio/mm³</td>
</tr>
<tr>
<td>Trombocyte</td>
<td>158</td>
<td>199 000/mm³</td>
<td>120–390 000/mm³</td>
<td>200–350 000/mm³</td>
<td>33 000/mm³</td>
</tr>
<tr>
<td>Leukocyte</td>
<td>186</td>
<td>4770/mm³</td>
<td>2400–10 000/mm³</td>
<td>4–10 000/mm³</td>
<td>1600/mm³</td>
</tr>
<tr>
<td>Staff cell</td>
<td>165</td>
<td>9.2%</td>
<td>1–30%</td>
<td>3–5%</td>
<td>21%</td>
</tr>
<tr>
<td>Segmented cell</td>
<td>165</td>
<td>49%</td>
<td>26–73%</td>
<td>50–70%</td>
<td>29%</td>
</tr>
<tr>
<td>Eosinophil cell</td>
<td>165</td>
<td>3.15%</td>
<td>0.5–11%</td>
<td>2–4%</td>
<td>–</td>
</tr>
<tr>
<td>Monocyte</td>
<td>165</td>
<td>6.5%</td>
<td>1–14%</td>
<td>2–8%</td>
<td>12%</td>
</tr>
<tr>
<td>Lymphocyte</td>
<td>165</td>
<td>32%</td>
<td>10–50%</td>
<td>25–40%</td>
<td>38%</td>
</tr>
</tbody>
</table>
maintain the maximum permissible concentration at one third. Some individual half-lives are estimated during a holiday period or after ceasing contact with tritium. The results are listed in Table IV. We found that effective half-lives are independent of chemical composition of tritium intake, but depend above all on the level of contamination. For excreted tritium concentrations in urine above 30 μCi/litre, half-lives between 7–9 days were found. For concentrations up to MPC, the period increases to 10–14 days and it exceeds 14 days for low concentrations up to 5 μCi/litre. Increased consumption of liquid reduces the individual half-lives. In a particular case an effective half-life of 5 days was observed with an intake of about 10 litres of liquid daily.

MEDICAL EXAMINATIONS

Blood pictures were specially investigated in routine medical examinations of workers involved in luminous compound production. Out of more than 150 blood analyses we have tried to find correlation between year dose, contemporary body burden and blood picture parameters. There is absolutely no correlation between tritium concentration in urine, ranging from 2 to 62 μCi/litre, and blood picture parameters. Also accumulated doses from internal tritium contamination, with corresponding yearly doses of 500–6000 mrem, have no influence on blood picture changes. In a particular case of reversible tritium panneryopathy, with a completely destroyed blood picture as described by Flammer [5], a body burden of 112 mCi and tritium concentration in urine of 2820 μCi/litre were determined. Results of blood picture statistics are listed in Table V. For comparison, also normal values of non-exposed persons and values of the panneryopathic person with obvious change in blood picture are shown in Table V. According to our experience, routine blood picture controls do not give reliable information about the degree of radiation hazard of workers being exposed to tritium or being contaminated within the range of MPL.

WASTE DISPOSAL

About 4% of the processed activity for luminous compound production becomes active waste, such as molecular sieves of oxidation units, methylalcohol from precipitation of tritiated polystyrene, fixed on silicous earth, equipment waste, rubber gloves, working material and luminous compound waste. These wastes are canned and are then placed in gas-tight cylinders, which in turn are welded and placed in bigger barrels. The empty space between cylinder and
TABLE VI. INFLUENCE OF AUTOMATION AND INSTALLATION STANDARDS IN LUMINIZING PLANTS ON BODY BURDEN OF OPERATORS

<table>
<thead>
<tr>
<th>Plant</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of luminizers</td>
<td>2</td>
<td>30</td>
<td>4</td>
</tr>
<tr>
<td>Processed activity yearly (Ci)</td>
<td>1000</td>
<td>5500</td>
<td>1000</td>
</tr>
<tr>
<td>Average $^3$H-content in urine</td>
<td>8.2</td>
<td>3.6</td>
<td>12.9</td>
</tr>
<tr>
<td>Body burden $\frac{\text{man} \cdot \text{rem}}{\text{Ci processed}}$</td>
<td>$3.3 \times 10^{-3}$</td>
<td>$3.9 \times 10^{-3}$</td>
<td>$1 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

Plant (A): Very high level in automation of luminizing.
Plant (B): Very high level of ventilation and organization.
Plant (C): Average ventilation and mainly manual luminizing.

barrel is filled with concrete. These barrels are collected once a year by the Federal Health Protection Organization for final disposal. Low active waste, such as cleaning papers and solvents, are also collected; papers can be burnt under control and solvents are worked up.

LUMINIZING FACTORIES: THEIR WORKING METHODS AND CONTAMINATION LEVELS

There are some 50 luminizing plants in Switzerland, processing about 20,000 Ci of tritium yearly. As already explained, the radiation damage from tritiated polystyrene produces an annual release of 5%. Since the luminizers keep only minimal quantities of material in stock, the average release is assumed to be less than 3 Ci for a medium plant. The ordering quantities are small and range from 1 to 30 Ci. This material is stored in exhausted glove boxes, where it is also mixed in small portions with lacquer to be processed by different methods on exhausted tables. The painting of dials and hands with manual or semi-automatic methods cannot be reasonably executed in glove boxes. A big problem is the use of gloves for manual painting of very small dials and hands. The direct contact of hands with the active material, contaminated surfaces and instruments results in additional internal contamination.

Producers of tritium luminous compound, and also some users of such material, have developed very effective luminizing methods, such as pneumatic pens and semi-automatic machines for dial and handpainting, in order to achieve high production figures and reduce painting cost. These semi-automatic instruments have
considerable importance from the point of view of radiation protection: reduction of contact time with open radioactivity, working with non-dispersable material (mixture of luminous pigment with lacquer) and decrease in internal body and surface contamination.

For more than five years we have been collecting results of urine measurements from different luminizing plants, as well as figures of total amounts of tritium activity processed, specific activity of used compound and data concerning internal contamination of workers. As shown in Table VI, there are big differences between the plants, depending on the availability and the standard of processing equipment, ventilation installation, organization and working discipline. Working conditions have generally greater influence on internal contamination than total amount processed and specific activity of the luminous compound used [6].

The processing of luminous material causes unavoidable high surface contamination. One mCi of tritium, being the equivalent of 4 mg medium quality luminous paint, is sufficient to contaminate a surface of 100 m² up to the MPL of $10^{-3}$ μCi/cm². Even with the exempted activity amount of 100 μCi, a floor of 10 m² can be contaminated up to $10^{-3}$ μCi/cm². It is therefore not surprising that surface contamination levels between 0.01 and 1 μCi/cm², with an average value of 0.7 μCi/cm², can occur during processing hundreds of curies. Only by thorough daily decontamination would it be possible to remain within the limits. A comparison of tritium, 90Sr and 147Pm, shows that a distinction should be made for permissible surface contamination levels for tritium, as it is in practice for maximum permissible values of total body uptake, exempted activity limit and MPC for air and water (Table VII).

AIR AND WATER POLLUTION

Because of fairly expensive investment, standardization problems and memory effects, tritium air monitoring systems are still not installed in luminizing factories. On the other hand, internal contamination, arising through skin by contact with contaminated surfaces and instruments or even oral uptake, seems to be as important as incorporation of contaminated air. There is a general tendency to improve ventilation systems and working conditions rather than install costly monitoring systems, since regular urine measurements provide direct and reliable information on radiation hazards.

Waste water is strictly controlled by the authorities. A filtration unit, for elimination of possible radioactive compound particles in waste water, is connected to a continuous working sampling system. After passing through two separate sedimentation chambers, the waste water is pumped through a filter and a small percentage is then branched off for control purposes. This system ensures a reduction in waste water contamination by a factor of 5 to 10.
<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$^3$H</th>
<th>$^{147}$Pm</th>
<th>$^{90}$Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td>MP body content ($\mu$Ci)</td>
<td>1200</td>
<td>31</td>
<td>2</td>
</tr>
<tr>
<td>MPC laboratory air (Ci/m$^3$)</td>
<td>$2 \times 10^{-3}$</td>
<td>$6 \times 10^{-8}$</td>
<td>$10^{-9}$</td>
</tr>
<tr>
<td>MPC drinking water for exposed persons (Ci/m$^3$)</td>
<td>0.1</td>
<td>$6 \times 10^{-3}$</td>
<td>$10^{-6}$</td>
</tr>
<tr>
<td>MPC waste water ($\mu$Ci/m$^3$)</td>
<td>330</td>
<td>20</td>
<td>$3.3 \times 10^{-3}$</td>
</tr>
<tr>
<td>Exempted activity limit ($\mu$Ci)</td>
<td>100</td>
<td>$10^{-1}$</td>
<td>$10^{-3}$</td>
</tr>
<tr>
<td>MPL surface contamination in controlled area ($\mu$Ci/cm$^2$)</td>
<td>$10^{-3}$</td>
<td>$10^{-3}$</td>
<td>$10^{-3}$</td>
</tr>
</tbody>
</table>
Depending on the contamination level in the working areas, the tritium concentration in waste water ranges from 1 to 10 μCi/litre. These values seem to be very high but the amounts of contaminated water occurring are very low for both the manufacturer and also the luminizers. About 5 to 10 litres of waste water occur per person and day plus 10 litres of such water resulting from decontamination for every 20 m² of area. During processing of luminous compound, some 5% of material accumulates as active waste in the form of unprocessed material, paint being removed from instruments, or resulting from production rejects and from cleaning and decontamination of equipment. Up to now, each luminizing factory has had to collect solid and liquid waste and deliver it once a year to the Federal Institute of Reactor Research for further disposal.

CONCLUSIONS

Production and processing of kilocurie quantities of tritium luminous compound can be carried out without excessive body burden for persons and environmental pollution. Modern processing equipment, suitable facilities, working discipline, regular decontamination and controls are the basic necessity to achieve this aim.

Although surface contamination levels often exceed the existing MPL, internal body burden always remains within the limits. This demonstrates that there seems to be a discrepancy between body content and surface contamination values. The rules distinguish between different beta isotopes for all maximum permissible values with the exception of MPL values for surface contamination. Revision should therefore be considered.

REFERENCES


DISCHARGES OF TRITIUM TO THE ENVIRONMENT FROM UNRESTRICTED USE OF CONSUMER PRODUCTS CONTAINING THIS RADIONUCLIDE

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Abstract

DISCHARGES OF TRITIUM TO THE ENVIRONMENT FROM UNRESTRICTED USE OF CONSUMER PRODUCTS CONTAINING THIS RADIONUCLIDE.

Not only nuclear installations but also consumer products containing tritium are an important source of man-made tritium discharge to the environment. In the Federal Republic of Germany about the same tritium activity is annually added to consumer products as is released each year from all nuclear installations. The total tritium activity distributed may rise considerably if devices with Gaseous Tritium Light Sources (GTLS) are permitted for large-scale unrestricted use and consequently also for large-scale uncontrolled disposal. The tritium added to consumer products and, at least partly, finally discharged to the environment is converted to HTO and participates in the normal water cycle of the earth. Therefore it would be very desirable to know how much tritium is used worldwide for such purposes, and it is proposed that the competent national authorities should report to an international organization the amount of tritium in consumer products permitted for unrestricted use and disposal. Finally a review of the normal waste management of tritium in the Federal Republic of Germany is given and doses that could result from incineration and pyrolysis of waste contaminated with tritium are assessed.

1. SURVEY OF CONSUMER PRODUCTS CONTAINING TRITIUM

Consumer products containing tritium may be classified as follows:

(a) Radioluminous products containing tritium in paint or plastic.
(b) Radioluminous products containing tritium in sealed tubes.
(c) Electronic, electrical and other devices.

Table I gives a list of currently available consumer products. It is derived from an UNSCEAR Report [1] and gives a survey of the worldwide situation. There are many devices in the list that are not regarded as consumer products by the Federal German authorities, e.g. gas chromatographs, or are not permitted
### TABLE I. CURRENTLY AVAILABLE CONSUMER PRODUCTS CONTAINING TRITIUM

<table>
<thead>
<tr>
<th>1. Radioluminous products containing tritium in paint or plastic</th>
<th>Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Timepieces</td>
<td>1 – 25 mCi</td>
</tr>
<tr>
<td>Compasses</td>
<td>5 – 50 mCi</td>
</tr>
<tr>
<td>Instrument dials and markers</td>
<td>25 mCi</td>
</tr>
<tr>
<td>Automobile lock illuminators</td>
<td>2 – 15 mCi</td>
</tr>
<tr>
<td>Automobile shift quadrants</td>
<td>25 mCi</td>
</tr>
<tr>
<td>Bell pushes</td>
<td>0.3 mCi</td>
</tr>
<tr>
<td>Rims for underwater watches</td>
<td>0.3 mCi</td>
</tr>
<tr>
<td>Spirit levels</td>
<td>5 – 25 mCi</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>2. Radioluminous products containing tritium in sealed tubes</th>
<th>Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Timepieces</td>
<td>0.2 – 0.4 Ci</td>
</tr>
<tr>
<td>Ordinary compasses</td>
<td>0.2 – 0.4 Ci</td>
</tr>
<tr>
<td>Marine compasses</td>
<td>0.2 – 2 Ci</td>
</tr>
<tr>
<td>Marine navigational instruments</td>
<td>0.25 Ci</td>
</tr>
<tr>
<td>Markers</td>
<td>4 Ci</td>
</tr>
<tr>
<td>Instruments, signs and indicators</td>
<td>2 Ci</td>
</tr>
<tr>
<td>Exit signs for commercial buildings</td>
<td>15 Ci</td>
</tr>
<tr>
<td>Large signs</td>
<td>30 Ci</td>
</tr>
<tr>
<td>Small exit signs</td>
<td>2 Ci</td>
</tr>
<tr>
<td>Step markers</td>
<td>2 Ci</td>
</tr>
<tr>
<td>Mooring buoys and lights</td>
<td>2 Ci</td>
</tr>
<tr>
<td>Public telephone dials</td>
<td>0.5 Ci</td>
</tr>
<tr>
<td>Light switch markers</td>
<td>0.2 Ci</td>
</tr>
<tr>
<td>Bell pushes</td>
<td>10 mCi</td>
</tr>
<tr>
<td>Miniature light sources</td>
<td>20 mCi</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>3. Electronic and other devices</th>
<th>Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electronic tubes</td>
<td>$1 \times 10^4$ μCi</td>
</tr>
<tr>
<td>Cold-cathode tubes</td>
<td>90 μCi</td>
</tr>
<tr>
<td>Glow lamps</td>
<td>10 μCi</td>
</tr>
<tr>
<td>Antistatic devices contained in precision balances</td>
<td>1 mCi</td>
</tr>
<tr>
<td>Gas chromatographs</td>
<td>250 mCi</td>
</tr>
</tbody>
</table>
for use as products exempted from licensing and/or reporting (exempt products) in the Federal Republic of Germany. Also the activities given in Table I appear to be rather high for some products. For instance, nearly all wrist-watches with tritium-painted dials produced today in the Federal Republic of Germany have a tritium activity of 0.1 mCi per watch or even less. The reason for this is that the radioluminous paint is used very sparingly today, and only the pointers and some markings on the dials of the watches are painted.

In general, there seems to be a general tendency to replace the tritium in radioluminous paint by tritium contained in sealed tubes, the so-called 'Gaseous Tritium Light Sources' (GTLS). The light is then emitted by phosphor coated on the inner surfaces of the glass containers that are in general made of borosilicate glass with a wall thickness between 0.1 and 2 mm. Not more than 2% of the tritium gas in the container should be in the form of tritiated water. In comparison with tritiated luminous paint a GTLS has the advantage that it is a sealed source and therefore the risk of incorporation can be neglected as long the glass tube is gas tight. The use of GTLS has the disadvantage that fairly large tritium activities are needed. As in addition many new applications for the illumination of consumer products are possible with the use of GTLS, the total tritium activity that is distributed without any restriction may rise considerably with time.

2. **TOTAL ACTIVITY**

There seem to be no general data available on the total activity of tritium that is added to consumer products in all parts of the world. Therefore only figures valid for the Federal Republic of Germany are given. Here the authorities are reluctant to give exemptions for the unrestricted use and waste disposal of consumer products containing tritium. Only electronic tubes and apparatus containing scales or dials with luminous paint (e.g. timepieces) are exempt products in the Federal Republic of Germany. The whole tritium activity added yearly to these products is some 1000 Ci. Radioluminous products with GTLS are not allowed for unrestricted use. They are produced mainly for export and also used in military applications. About 14000 Ci of tritium contained in GTLS are imported annually into the Federal Republic of Germany and 10000 Ci are exported.

These figures could rise considerably if exemptions for devices with GTLS were given on a large scale. As an example, the amount of tritium in wrist-watches will be assessed, because liquid crystal display watches with GTLS can be used and disposed of unrestrictedly in many countries today. The watches are normally equipped with 2 GTLS each containing an activity of 100 mCi. In the Federal Republic of Germany the consumption of wrist-watches has been nearly
TABLE II. WASTE DISPOSAL FACILITIES IN THE FEDERAL REPUBLIC OF GERMANY IN 1975

<table>
<thead>
<tr>
<th>Type of waste disposal</th>
<th>Number of plants</th>
<th>Percentage of waste (%)</th>
<th>Number of persons serviced ($\times 10^4$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direct deposition in tips</td>
<td>4415</td>
<td>76</td>
<td>47</td>
</tr>
<tr>
<td>Incineration</td>
<td>31 (48$^a$)</td>
<td>21</td>
<td>13</td>
</tr>
<tr>
<td>Compost production</td>
<td>24</td>
<td>3</td>
<td>2</td>
</tr>
</tbody>
</table>

$^a$ Including plants incinerating residue from compost production.

constant in the last few years, amounting to 12 million watches yearly. If it is assumed that this figure will hold also for the future, and that each fourth watch sold will be equipped with GTLS, 600 000 Ci of tritium would thus be distributed yearly among the population of the Federal Republic of Germany. If it is further assumed that the useful lifetime of a wrist-watch is five years, 450 000 Ci of tritium are disposed of annually with the normal waste. In Section 3 it will be shown that by the waste disposal methods used in the Federal Republic of Germany about 60% of the activity enclosed in the watches, i.e. 270 000 Ci tritium annually, would be released to the environment in the Federal Republic of Germany, if wrist-watches with GTLS came into general use in this country. This value can be compared with 6000 Ci of tritium released in 1976 from all nuclear installations of the Federal Republic of Germany.

3. DISCHARGES TO THE ENVIRONMENT

In principle a discharge of tritium to the environment is possible at every stage during the lifetime of the consumer product; that means during production, distribution, normal use, possible mis-use, repair and maintenance, and normal waste disposal.

Since production of consumer products is permitted in all countries only under the strict control of the competent national authorities, no significant discharges of tritium have to be taken into account for this stage of the lifetime.
TABLE III. DEVELOPMENT OF WASTE INCINERATION IN THE FEDERAL REPUBLIC OF GERMANY

<table>
<thead>
<tr>
<th>Year</th>
<th>Number of plants</th>
<th>Incinerated waste (X 1000 t/a)</th>
<th>Number of persons serviced (X 1000)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1960</td>
<td>1</td>
<td>80</td>
<td>0.2</td>
</tr>
<tr>
<td>1965</td>
<td>7</td>
<td>718</td>
<td>245</td>
</tr>
<tr>
<td>1970</td>
<td>24</td>
<td>2829</td>
<td>859</td>
</tr>
<tr>
<td>1975</td>
<td>31</td>
<td>4517</td>
<td>1337</td>
</tr>
<tr>
<td>1977</td>
<td>39</td>
<td>5297</td>
<td>1515</td>
</tr>
</tbody>
</table>

TABLE IV. RELATION BETWEEN POPULATION DENSITY AND WASTE INCINERATION IN THE FEDERAL REPUBLIC OF GERMANY

<table>
<thead>
<tr>
<th>Community classification (X 1000 inhabitants)</th>
<th>Percentage of the population living in the different classes</th>
<th>Percentage of waste incinerated in the different classes</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1975</td>
<td>1980 (estimated)</td>
</tr>
<tr>
<td>Under 10</td>
<td>27.5</td>
<td>0</td>
</tr>
<tr>
<td>10 – 50</td>
<td>28.4</td>
<td>0.5</td>
</tr>
<tr>
<td>50 – 100</td>
<td>9.3</td>
<td>10.6</td>
</tr>
<tr>
<td>100 – 500</td>
<td>17.1</td>
<td>65.9</td>
</tr>
<tr>
<td>More than 500</td>
<td>17.7</td>
<td>65.9</td>
</tr>
</tbody>
</table>

The discharge during the other stages depends on the form in which the tritium is contained in the consumer product. Whereas products with radioluminous paint or plastic will lose a certain part of their activity during the lifetime, for radioluminous products with GTLS it may be assumed that the device provides adequate protection during distribution, normal use, maintenance and in many cases also during repair. Taking also into account that the portion of GTLS destroyed by misuse is so small that it may be neglected, it may be considered that normal waste disposal is the critical stage, and this is now considered in detail.
In the Federal Republic of Germany today almost all domestic refuse is regularly collected and disposed of. Tables II–IV present a survey of the situation. Today 25% of domestic refuse is incinerated in 44 plants, three of them working up the waste of more than 1 million people and six the waste of less than 0.1 million. As indicated by Table IV, nearly all waste incineration plants are situated in densely populated conurbations; the population density of these areas may be assumed to be 20/ha [2–4].

When assessing the part of tritium that is discharged to the environment by the various waste disposal methods, only devices with GTLS are considered, as because of their high activities these devices will contribute overwhelmingly to the total tritium content of the refuse. For instance, the tritium activity of 450 000 Ci, derived in Section 2 as an upper limit for wrist-watches with GTLS, would lead to a mean contamination of 24 mCi/t for the West German domestic refuse. It can be assumed that the shielding given by the device to the GTLS is sufficient to protect it during waste transport. Whether the device will also resist the strong mechanical forces of the packing and milling machines used at many disposal plants seems to be doubtful. Concerning this point further investigations are needed. This also applies to the doses that workers on these machines could receive. It is very likely that on tips working with waste packing and levelling a high percentage of the GTLS are broken and the tritium set free. Assessment of doses is difficult in this case because it is not known what part of the tritium becomes airborne and how fast it is oxidized. In compost production it is supposed that all tritium is set free because of the milling needed in this kind of waste disposal. At temperatures of about 1000°C used in waste incineration plants the whole tritium becomes free also. It will be oxidized and emitted over the stack of the plant together with the other airborne releases from the incineration.

4. DOSE ASSESSMENTS

In principle doses resulting from possible tritium discharges must be assessed for each stage of the lifetime of a consumer product. Because the production and, as far as direct handling with the radioactive material is involved, also repair and maintenance is carried out by supervised occupational radiation workers, only the other stages named in Section 3 must be considered. Possible doses resulting from distribution, normal use and credible mis-use strongly depend on the type of consumer product, its intended use and details of the construction and design, particularly relating to containment and shielding of the radioactive material. Therefore dose assessments concerning these stages must be done for each product separately when the exemption is given. Only the doses due to waste disposal are therefore investigated here.
4.1. Global dose assessment

An assessment of the population dose from the disposal of consumer products containing tritium was made by Wrixon and Webb [5]. It was assumed that the tritium will rapidly mix with the atmosphere, will be slowly converted to tritiated water and finally mix with the circulating water of the northern hemisphere to a depth of 75 m resulting in an average tritium concentration of $2 \times 10^{-15}$ Ci/g at equilibrium if $10^6$ Ci tritium are released annually. With the assumption that the water in the body fluids of the population will finally have the same tritium concentration, the collective dose for the population of the United Kingdom (taken as $5 \times 10^7$) at equilibrium is assessed as 9.4 man-rem/a for an annual release of $10^6$ Ci. With the quality factor 1.7 the corresponding value for the Federal Republic of Germany (population of $6.2 \times 10^7$) would be $2 \times 10^{-5}$ man-rem/a for a tritium release of 1 Ci/a.

The global assessment model described above does not take into consideration the doses that the tritium may deliver to persons and populations before mixing with the surface waters of one hemisphere. It is shown below that this leads to an important underestimation of doses, at least for certain modes of waste disposal.

4.2. Incineration of waste

As described in Section 3, the oxidized tritium is emitted by the stack of the waste incineration plant. The tritium concentration in the air around the plant depends not only on the amount and the height of the release, but also on the climatological and topographical features of the site of the plant. It is not intended here to calculate exactly doses around each of the 44 incineration plants in the Federal Republic of Germany, but to present a simple method applicable to the assessment of doses at any site in the Federal Republic of Germany. This method is based on the annual average gaseous dispersion factor $\chi$ used to estimate doses from reactor effluents, when the exact weather conditions concerning atmospheric stability and windspeed are not known and precise values for the doses are not needed. Also the influence of wash-out and fall-out is not taken into consideration in these $\chi$-values, which are given in Fig. 1 for emission heights of 20, 50, 100 and 150 m. Thus only the distribution of the wind directions must be known for a certain site. The $\chi$-values given in Fig. 1 apply to a 100% wind frequency in a 30° sector. If an even wind distribution is assumed, $\chi$-values therefore must be lowered by a factor of 12. The tritium concentration of the ground level air is $3.2 \times 10^{-9}$ Qx Ci/m$^3$ if Q is the tritium activity in curies emitted yearly. A very simple formula for the upper limit of the whole body dose of a person living in this air is derived from the assumption that the ratio of tritiated to normal water in the body fluids of that person is the same as in the surrounding air. If a chronic tritium concentration of 1 Ci per gram of body water delivers a
dose rate of $10^8$ rad yearly [5], a quality factor of 1.7 is taken and $h$ is the mean value of the absolute humidity of the ground level air in g/m$^3$, the yearly dose is given by

$$D_t = 5.4 \, Q \, \chi \, h^{-1} \, [\text{rem/\text{a}}] \quad (1)$$

The right ordinate of Fig. 1 shows the doses given by Eq. (1) for a mean humidity of 6.6 g/m$^3$, a value derived from the weather conditions of the Federal Republic of Germany, an activity release of 1 Ci/a and by the assumption that the wind is evenly distributed over all directions.

It is quite clear that Eq. (1) leads to an over-estimation of the whole body dose. For a single person Eq. (1) would give the correct dose only if that person would (a) remain during the whole year at the same place; (b) consume only food growing at this place or coming from animals living at this place; (c) obtain all drinking water from the condensed moisture of this place.
In particular the last assumption is a large over-estimation. It is therefore interesting to use the established formulas [6] to calculate the percentages that the different pathways for tritium intake contribute to the dose given in Fig. 1. With consumption rates of 7300 m$^3$ air, 240 kg botanical products, 110 kg milk and milk products and 75 kg meat annually the following percentages are calculated:

(1a) 4.8\% for inhalation
(1b) 4.8\% for intake over the skin (this value is assumed to be equal to the calculated inhalation value [7])
(2a) 13.3\% for consumption of contaminated botanical products
(2b) 3.4\% for consumption of contaminated milk and milk products
(2c) 2.7\% for consumption of contaminated meat.

If it is assumed that about half of the yearly food consumption of individual members of the population may in reality be produced in the neighbourhood of their living places, the doses given in Fig. 1 are over-estimated by a factor of 5. But we must bear in mind that these doses are derived from some general assumptions that are not always on the safe side. For instance, the assumption that the wind directions are evenly distributed may under-estimate for certain directions in some regions the doses by a factor of 2–3. Therefore it seems to be prudent to use the values given in Fig. 1 when the doses of critical groups of the population are assessed.

The assessment of the population dose is made with 10\% of the dose values given in Fig. 1. Assuming a population density of 20 persons per hectare for distances up to 10 km from the incineration plant, and an emission height of 100 m, leads to a population dose of $3 \times 10^{-3}$ man·rem/a for a tritium release of 1 Ci/a. If also larger distances are taken into account, integration over the extrapolated 100 m graph up to 50 km gives an additional population dose of $1 \times 10^{-3}$ man·rem/Ci if the population density in this area is assumed to be 2 persons per hectare.

4.3. Pyrolytic decomposition of waste

In 27 of the 31 waste incineration plants in Table II the heat evolved is used for heating purposes or for steam and electricity generation. The electricity yield of the refuse incinerated in 1973 was equivalent to 0.2\% of the total electricity produced in the Federal Republic of Germany. An even more effective thermal utilization of waste can be done by pyrolysis, the thermal decomposition of the refuse without oxygen. Today the production of oil and gas from waste by pyrolytic processes is carried out in the Federal Republic only on a laboratory scale and in some pilot plants. A pyrolytic process that is especially suitable for
<table>
<thead>
<tr>
<th>Type of dose</th>
<th>Global model</th>
<th>Local and regional models</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Initially</td>
<td>At equilibrium</td>
</tr>
<tr>
<td>Population doses per annual activity release (X man-rem/Ci)</td>
<td>$1 \times 10^{-6}$</td>
<td>$2 \times 10^{-5}$</td>
</tr>
<tr>
<td>Expected population doses if GTLS with 450,000 Ci are disposed of annually (X man-rem)</td>
<td>0.3</td>
<td>5</td>
</tr>
<tr>
<td>Expected doses of critical groups if GTLS with 450,000 Ci are disposed of annually (X mrem)</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>
the conversion of domestic refuse into gas produces about 1 m$^3$ of gas with a specific energy of 6300 kJ/m$^3$ from 1 kg of waste [8].

If devices with GTLS were in the waste it must be assumed that the T$_2$ is set free during the pyrolytic process (temperatures up to 1100°C), mixes with the H$_2$-content of the pyrolytic gas (about 20% of the gas volume), remains in that gas during the cleaning procedure and is oxidized if the gas is burned at its final location. Although it is not known whether commercial pyrolysis of domestic refuse will be used in the Federal Republic of Germany and if so, what will be done with the resulting gas, an estimation of possible doses is given to demonstrate that future techniques of waste re-use may give rise to even higher doses than those calculated in Section 4.2.

If it is assumed that the gas is burned only in equipment that is vented by stack, the estimation method derived in Section 4.2 can be used, the only difference being that the stack height will be lower. By using the dose values given by the 20 m graph of Fig. 1, a population dose of $13 \times 10^{-3}$ man·rem/a for a tritium release of 1 Ci/a is assessed. The assumption made above is only fulfilled if the gas is burned directly at the pyrolytic plant or used for special applications; it is not fulfilled if the gas is fed into the public gas supply. Although in the Federal Republic of Germany unvented room heating and warm water supply with gas is not allowed, unvented cooking with gas leads to relative high doses in this case.

The total gas consumption in the Federal Republic of Germany amounted to $65 \times 10^8$ m$^3$ in 1976. Of this volume 13.7% was consumed in $6.6 \times 10^6$ households equipped with gas, resulting in a mean consumption of 1350 m$^3$ per household. Most of this gas is used for room heating and warm water supply, only 200 m$^3$ per year and household being used for cooking. With the assumption that each household equipped with gas is also cooking with gas, that all cooking is done unvented in a room of 20 m$^3$ with a ventilation rate of 2 hourly and that one person is present in this room during cooking, a population dose of 0.14 man·rem/a for a tritium release of 1 Ci/a is assessed. The calculation is carried out with an inhalation rate of 0.83 m$^3$/h and an inhalation dose factor of 170 rem/Ci. It is further assumed that the tritium intake over the skin is equal to the inhaled activity [7]. If it is assumed that the whole gas supply of a town comes from waste pyrolysis, the doses for single persons may be rather high. Taking into account the mean waste contamination of 24 µCi/kg assumed in Section 3 as an upper limit for wrist-watches with GTLS, a person cooking in a 20 m$^3$ room with a ventilation rate of 2/h would receive a dose of 34 mrem/a.

5. SUMMARY AND CONCLUSIONS

Because the West German authorities have hitherto been very restrictive in permitting exemptions of consumer products containing radioactive substances,
the total tritium activity used annually for the production of consumer products and consequently the tritium activity released annually from these products does not exceed the annual contribution from the total nuclear power production in the Federal Republic of Germany. This latter figure is also still fairly low because reprocessing is performed at present on a small scale only. In the near future in the Federal Republic of Germany the major annual tritium releases from nuclear power production will originate from fuel reprocessing plants. About $2 \times 10^3 \text{ Ci/GW(e)}$ will still be released even if 90% retention is assumed [9]. But a liberal exemption policy concerning devices with GTLS would easily lead to annual tritium releases in excess of the contributions from reprocessing plants. If 20 GW(e) were produced annually this would lead to a release of $4 \times 10^4 \text{ Ci}$ tritium annually. If, on the other hand, the use of wrist-watches with GTLS were unrestricted, this would cause annual tritium releases which could approach levels larger by about one order of magnitude. As devices with GTLS are permitted for unrestricted use and waste disposal in many countries today, consumer products may contribute an important part to man-made peaceful discharges of tritium to the environment. In accordance with an OECD/NEA guide [10] it is therefore proposed that international organizations should be kept informed by competent national authorities of the amount of tritium in consumer products.

Assessment of the doses resulting from tritium releases is relatively easy in the case of nuclear power plants, but comparatively difficult in the case of releases from consumer products. This is primarily because in the latter case the identification of the time and the points of the release as well as the actual activities released at each time and point is extremely difficult: the release may occur at any time and any point between manufacture and disposal of the product. In this review only doses from two different modes of waste disposal are considered. The results in Table V show that the global model under-estimates the population dose of the Federal Republic of Germany if waste disposal methods used in this country are taken into account.

REFERENCES


LARGE-SCALE DISTRIBUTION OF TRITIUM IN A COMMERCIAL PRODUCT

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*Presented by H.E. Book*

Abstract

**LARGE-SCALE DISTRIBUTION OF TRITIUM IN A COMMERCIAL PRODUCT.**

Tritium enters the environment from various sources including nuclear reactor operations, weapons testing, natural production, and from the manufacture, use and ultimate disposal of commercial products containing tritium. A recent commercial application of tritium in the United States of America involves the backlighting of liquid crystal displays (LCD) in digital electronic watches. These watches are distributed through normal commercial channels to the general public. One million curies (MCi) of tritium were distributed in 1977 in this product. This is a significant quantity of tritium compared with power reactor-produced tritium (3 MCi yearly) or with naturally produced tritium (6 MCi yearly). This is the single largest commercial application involving tritium to date. The final disposition of tritium from large quantities of this product, after its useful life, must be estimated by considering the means of disposal and the possibility of dispersal of tritium concurrent with disposal. The most likely method of final disposition of this product will be disposal in solid refuse; this includes burial in land fills and incineration. Burial in land fills will probably contain the tritium for its effective lifetime, whereas incineration will release all the tritium gas (as the oxide) to the atmosphere. The use and disposal of this product will be studied as part of an environmental study that is at present being prepared for the U.S. Nuclear Regulatory Commission.

1. INTRODUCTION

The total inventory of tritium in the environment and the means by which this inventory is increasing is of special interest and importance to the world population because any released tritium is rapidly assimilated into the earth's ecology. Tritium enters the environment from various primary sources including nuclear reactor operations, weapons testing and natural production. The natural production of tritium occurs predominantly through the fast-neutron irradiation of nitrogen by cosmic rays and through the direct ejection of tritium by stars [1]. There is yet another pathway for tritium to enter the environment. Tritium that has been manufactured for commercial uses may be released during the manufacture, use and ultimate disposal of commercial products containing tritium.
Examples of commercial products containing tritium that are distributed in the United States of America are electron tubes, tritium-painted timepieces, exit signs and other self-luminous products.

A recent commercial application of tritium in a product available to the general public in the United States of America involves the backlighting of liquid crystal displays (LCD) in digital electronic watches. Sealed glass tubes containing tritium as the activator and a phosphor as the luminescent material are used to backlight the entire LCD area for night-time viewing of the digital output. These watches, in final assembled form, are distributed through normal commercial channels to the general public. It is difficult to estimate the number of watches that will be distributed in the future because of the recent introduction of watches with LCDs, the low prices already experienced for these watches, competition with LCD watches not containing tritium and consumer acceptance of this product. However, one manufacturer has estimated distribution of 3 million watches in 1979. Since each watch contains a maximum of 200 mCi of tritium, 0.6 MCI will be made available for consumer use from this one manufacturer alone. This product application does represent a significant quantity of tritium when compared with power-reactor-produced tritium (3 MCI yearly) or with naturally produced tritium (6 MCI yearly), and qualifies it as the single largest commercial application involving tritium to date.

2. BACKGROUND

One of the oldest commercial applications of radioactivity in products available to the general public is the use of radioluminescent materials (principally for use in timepieces).

The original method of timepiece lighting involved painting a mixture of radium and phosphor on the surfaces of the timepiece desired for lighting. Even though this method is still being used to some extent, the use of radium is declining rapidly, having been replaced largely by painted mixtures of a tritium compound and phosphor or a promethium-147 compound and phosphor. These are the methods most used today for mechanical watches.

Consumer products, such as watches containing radioactive material, can only be distributed in the United States of America under regulations that provide for exempt distribution of the product. This means that a manufacturer may transfer possession of or distribute the product to persons exempt from licensing and regulatory requirements. However, the manufacturer must have a specific licence that authorizes this transfer or distribution. The U.S. Nuclear Regulatory Commission issues specific licences to persons to manufacture products containing

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1 This does not refer to countries with centrally planned economies.
radioactive material and to distribute these products to persons who are not licensed. These products must be manufactured in accordance with standards established by the U.S. Nuclear Regulatory Commission. After such a product is distributed, there is no further regulatory control over the use and disposal of the product.

The use of tritium-activated lighting for timepieces for the general public began in the United States of America in 1960, when regulations were established allowing the distribution of timepieces, each containing up to 25 mCi of a painted mixture of tritium, to persons exempt from licensing requirements. In 1969, a class exemption from licensing requirements was added to the regulations and provided for the distribution of self-luminous products containing tritium, krypton-85 or promethium-147, and for the use of such products by persons not licensed to possess radioactive material. In 1975, the first specific licence in the United States of America was granted, under the class exemption for self-luminous products, for the distribution of digital electronic watches, containing approximately 200 mCi of tritium each, to persons exempt from licensing. The Federal government retains jurisdiction over any specific licences granted which authorize distribution under this class exemption for self-luminous products.

3. LICENCE REQUIREMENTS

Strict product qualification and analysis is required to ensure that the product meets the safety criteria in the regulations before a specific licence to distribute under the class exemption is granted. The product must be designed and manufactured so that wear and abuse are unlikely to cause a potential dose to persons in excess of specific guidelines during normal handling and use of the product. An application for a licence must include a description of the product and its use; a description of the by-product material including quantity, chemical form, physical form and solubility; details of construction and design; safety features under normal and severe conditions of handling; quantities involved in distribution and storage; method of labelling and marking; procedures and results of prototype testing; estimated dose commitments using a strict set of criteria; detailed quality control procedures; and other specific information. The analysis, required before acceptance of this product application for distribution to exempt persons, considers breakage and accident incidents for single units in use, distribution and storage, and multiple or bulk units in manufacture, distribution, storage and disposal. The safety criteria in the regulations limit the average dose, or dose commitment, to members of the group expected to receive the highest dose from normal use and disposal of a single exempt unit to 1 millirrem yearly, and limit the dose, or dose commitment, received by persons engaged in marketing, distributing, and servicing of exempt products, as a result of exposure
to the quantities of exempt units likely to accumulate in one location, to 10 milli-
rem yearly. In addition, prototype test criteria required by the U.S. Nuclear
Regulatory Commission include a detectable tritium leakage limit of 0.1 μCi/d
per watch, after all other tests (vibration, temperature, thermal shock, pressure,
impact and immersion) have been performed. The analysis and evaluation of the
use of tritium for backlighting LCD digital watches satisfied all the safety criteria
in the regulations, and about 12 licences have been granted since 1975 to manu-
facture and distribute these watches, under the class exemption for self-luminous
products.

4. PRODUCT CHARACTERISTICS

Digital electronic watches that use hybrid integrated circuits are a fairly
recent development, with the first watches of this type coming on the market
about 1973. These watches can utilize either light-emitting diodes (LED) or a
liquid crystal display (LCD) for the digital readout. Tritium backlighting is used
for the LCD digital watch only, i.e. the method used provides for backlighting
the entire area behind the opaque numbers of the display. The self-luminous
module contains sealed glass tubes which contain tritium gas and a phosphor
coating on the internal surface of the tube. The tritium tubes are normally sealed
automatically by laser and have very low leakage rates of tritium gas — usually
less than 0.01 μCi/d for each unit [3]. This would represent a tritium leakage of
less than 3.6 Ci yearly from 1 million watches. Automatic laser sealing produces
extremely uniform and controlled sealed areas due to the precise manipulation
of a focused laser beam.

In the construction of the watch, the tritium tubes are shock-mounted in
a metal pan and the LCD with a diffuser is placed over the pan. The LCD-source-
pan assembly, along with electrical connectors and electronics, is assembled into
a single package called a watch module. The module is then mounted in a watch
case. The watch is extremely shock-resistant with respect to damage to the
tritium tubes and can withstand severe handling, such as a drop on to a steel
plate from heights of 2 m. Access to the light sources is only by disassembly
of the watch which requires some destruction of the components that were used
to assemble it. Any repair and maintenance which involves removal of the module
must be performed under a specific licence, granted by the U.S. Nuclear Regulatory
Commission, which authorizes redistribution. Dose estimates provided by the
manufacturers show whole-body dose commitments of about 2.5 μrem yearly to
persons using the watch and 1.0 μrem yearly to persons engaged in the distribution
and marketing of the product. A recent report [2] estimates doses of 3 to 20 μrem
yearly to persons using the watch and 0.4 to 100 μrem yearly to individuals
engaged in distribution and marketing.
Customer acceptance, low price and product uniqueness have all contributed to the early distribution of large numbers of tritium backlit digital watches. Quantities of tritium, contained in backlit digital watches distributed in the United States of America, are as follows:

*Fiscal year ending:*

- 30 June 1977 – 0.013 MCi tritium
- 30 June 1978 – 1.00  MCi tritium
- 30 June 1979 – 4.00  MCi tritium (estimated)
For purposes of comparison, there are approximately 0.3 MCi tritium distributed in all other commercial products each year in the United States of America, and approximately 0.7 MCi tritium produced by power reactors in the United States of America each year (see Fig. 1).

5. ULTIMATE DISPOSAL

The analysis required before licensing this product for distribution to exempt persons, evaluates the possibility of breakage and accident for single units in use, distribution and storage, and for multiple or bulk units in manufacture, distribution, storage and disposal. However, the long-term disposition, that is, the final or ultimate disposition of tritium from large quantities of this product after its useful life, must be estimated by considering the means of disposal and the possibility of dispersal of tritium concurrent with disposal.

The most likely method of the final or ultimate disposition of this product will be disposal in solid refuse; this includes burial in land fills and incineration. Burial in land fill, in all probability, will contain the tritium in the product for its effective lifetime because of the lack of stress. However, incineration will release all the tritium gas (immediately converted to the oxide) to the atmosphere. After some time (the digital electronic watch may have an average lifetime of six to ten years), the yearly production rate of the product will approximate to the yearly disposal rate. After the product's disposal rate approaches some equilibrium value in the future, it is possible by considering the factors of production rate, lifetime, radioactive decay and disposal rate, to estimate that an approximate quantity of 2 MCI of tritium contained in this product may be disposed of yearly as solid refuse. This would only occur in about 10 years time and only if present trends in acceptance and use of this product continue. The fraction of this disposal quantity of tritium that will ultimately be released to the atmosphere will depend on a number of unknown factors, the principal one being the fraction of solid refuse disposed of by incineration. The quantity of tritium released to the environment a future date could be up to 1 MCI yearly and, thus, would represent a significant addition of tritium to the environment when compared with the power-reactor-produced tritium of 3 MCI yearly or to naturally produced tritium of 6 MCI yearly.

The U.S. Nuclear Regulatory Commission, as part of a continuing programme of licensing and regulatory review, has undertaken a study to evaluate the environmental effects of the use and disposal of a number of commercial products containing radioactive materials; this will include digital electronic watches containing tritium. The results are expected within two years and will serve as the basis for a general environmental impact statement to be prepared by the U.S. Nuclear Regulatory Commission. Tritium does not bioaccumulate in the
environment, in food chains or in man [4]. However, tritium does accumulate physically in the environment, and, therefore the environmental impact statement will investigate both the effects of the use of this product and the effects of the fraction of tritium released to the environment during disposal of this product.

6. SUMMARY

A large-scale commercial application of tritium is the use of sealed glass tubes containing tritium and a phosphor to backlight liquid crystal displays in digital electronic watches. Projections indicate that large quantities of this product will be disposed of or discarded sometime in the future, after the product's useful life is over. The total quantity of tritium contained in the discarded items, after considering radioactive decay, may be about 2 M Ci yearly approximately ten years in the future. The fraction of this disposal quantity of tritium that will be released to the environment (for example, by incineration) will depend on a number of unknown factors. The quantity of tritium that is actually released to the environment in the future could be up to 1 M Ci yearly.

REFERENCES

DISCUSSION

on the previous three papers

S. PORTER: Mr. Krejčí, can you describe the chemical form of the $^3$H released in the manufacture and use of luminous compounds in Switzerland, and have you any information, obtained from examination of exposed workers, concerning the long biological half-life $^3$H compartments (> 15 d)?

K. KREJČÍ: The $^3$H is in the form of short-chain organic radicals of the styrene type, CH$_3$T, HTO. Above all HTO and HT are involved. Regarding your second question, I am afraid I cannot tell you anything about biological half-lives for $^3$H contents below 0.5 μCi/litre in urine.

P.J.C. DINNER: I have a question on the paper presented by Mr. Book. Has the United States Nuclear Regulatory Commission examined the question of the social cost of exposure to tritium timepiece releases of the order of 1 MCl/a, the figure you have mentioned? It would be interesting to know how the social benefits of beta-lights are to be evaluated against the cost of the radiation exposure consequently suffered by the United States and world populations.

H.E. BOOK: The quantities of tritium that may be released to the environment from digital electronic watches are only estimates at present, and have been derived by considering current trends in the acceptance and use of this product. The Nuclear Regulatory Commission is preparing an environmental impact statement that will evaluate the benefits and radiological impact of this product, as well as a number of other consumer products containing radioactive materials.

W. ROETHER: Mr. Krejčí, can you give any figures on the European total or world total production and release of tritium associated with the manufacture and use of consumer goods?

K. KREJČÍ: One can say that in 1978 about 2 million Ci will be processed in the world-wide production of timepieces, the figure for Europe alone being about half that amount. As far as releases from consumer goods are concerned, the world-wide total is probably something under 3000 Ci/a in the case of luminous compounds and 50 Ci/a in that of beta-lights.

T.R. THOMAS: Mr. Book, if the U.S. Nuclear Regulatory Commission continues to allow the use of $4 \times 10^6$ Ci/a of $^3$H in LCD watches and expects a release to the environment during waste incineration of up to $1 \times 10^6$ Ci/a, will light-water reactor reprocessing plants also be permitted to release their inventories to the environment?

H.E. BOOK: The question of tritium emissions from reprocessing plants goes beyond the scope of this paper. However, the U.S. Environmental Protection Agency establishes generally applicable environmental standards, contained in Title 40, Code of Federal Regulations, Part 190. These environmental standards require the consideration of all inputs to the environment for any particular
substance. An environmental standard for tritium has not been established to
date.

R.V. OSBORNE: I am interested in the inverse correlation that Mr. Krejčí
observed between the half-lives of tritium and the concentrations in urine. If
the chemical composition of the tritium compounds taken in is not responsible,
what explanation has he?

K. KREJČÍ: Higher liquid intake at higher urine levels may have a certain
influence with respect to the shorter half-lives, but probably other factors also
influence the excretion rate.

R. JALBERT: Were the surface contamination levels mentioned in your
paper estimated or measured — i.e. were they removable contamination values?

K. KREJČÍ: Surface contamination is determined by wipe or smear tests,
and it is assumed that 25% of the $^3$H activity is removed.

R. JALBERT: What contamination levels are typically found in rooms or
areas adjacent to the controlled areas maintained at the maximum surface
contamination values of $10^{-3}$ μCi/cm$^2$?

K. KREJČÍ: The controlled area is a separate building. All other buildings
in the immediate vicinity show surface contamination levels in the range of
$10^{-4} - 10^{-5}$ μCi/cm$^2$, depending on the type of work being done in these premises.
EVALUATION OF FUTURE DISCHARGES

Session 2, Part 2
PRODUCTION AND EMISSION OF TRITIUM FROM NUCLEAR FACILITIES, AND THE RESULTING PROBLEMS

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Abstract

PRODUCTION AND EMISSION OF TRITIUM FROM NUCLEAR FACILITIES, AND THE RESULTING PROBLEMS.

The production rate of $^3$H by various nuclear reactions is calculated for the various parts of nuclear power reactors. The $^3$H-production per MW(e) and year for boiling-water reactors, pressurized-water reactors, MAGNOX, AGR, high-temperature reactors, heavy-water reactors and fast breeders is: 20 Ci, 19 Ci, 26 Ci, 18 Ci, 15 Ci, 170 and 32 Ci respectively. If the total $^3$H of a large reprocessing plant is released through a 100-m high stack, the maximum radiation exposure exceeds the permissible total body dose of 30 mrem/a in the Federal Republic of Germany. By increasing the height of the stack, only the maximum radiation exposure can be reduced, not the collective dose. The collective dose over the first pass exposure is approximately 0.017 man·rem/Ci and after global distribution $9 \times 10^{-5}$ man·rem/Ci. From the radiation protection point of view, between 80 and 90% of $^3$H should be retained in reprocessing plants. At the moment it is advisable to mix the tritium with ocean water. From this mixed tritium the radiation exposure of the total body will be lower than 0.1 mrem yearly in the next century. For the disposal of tritium in the ocean an international agreement is required.

1. INTRODUCTION

Tritium produced in nuclear power plants is only partly emitted there. In the case of the reprocessing of nuclear fuel, the tritium is mainly emitted from reprocessing plants. The emission rates will increase with the construction of reprocessing plants of larger capacities.

2. PRODUCTION OF TRITIUM IN NUCLEAR POWER PLANTS

In nuclear power plants tritium is produced by fission processes as well as neutron capture and decay processes in the fissile material, breed material, coolant, moderator, reflector, and control- and structure materials. On the basis of
FIG. 1. $^3$H-production per 1 MW(th) in a boiling-water reactor (BWR) in the first three years of operation (without $^3$H production in absorber rods).

FIG. 2. $^3$H-production per 1 MW(th) in a pressurized-water reactor (PWR) in the first three years of operation.
Fig. 3. $^3$H-production per 1 MW(th) in a heavy-water reactor (HWR) in the first three years of operation (without $^3$H-production in absorber rods).

Fig. 4. $^3$H-production per 1 MW(th) in a MAGNOX reactor in the first three years of operation (without $^3$H-production in absorber rods).
FIG. 5. $^3$H-production per 1 MW(th) in an advanced gas-cooled reactor (AGR) in the first three years of operation (without $^3$H-production in absorber rods).

FIG. 6. $^3$H-production per 1 MW(th) in a high-temperature reactor (HTR) in the first three years of operation (without $^3$H-production in absorber rods).
detailed spectral calculations for the single reactor cells as well as the energy-dependent cross-sections for the different neutron reactions, the tritium production rate in the various reactor types has been calculated. The basic data are summarized in Ref. [1] except for some negligible modifications. Figures 1—7 show the tritium production per 1 MW(th) in the first three years of operation. The annual change of the fuel elements has been taken into consideration. Tritium production is mainly caused by ternary fission except in heavy-water reactors. It appears that most of the tritium is produced in heavy-water reactors and fast breeders.

Table I shows the tritium activity per MW(e) and per year produced in the various reactor types.

3. EMISSION OF TRITIUM FROM NUCLEAR POWER PLANTS AND REPROCESSING PLANTS

As there are no special retention facilities for tritium in light-water reactors, the tritium that is produced in the coolant and the part that reaches the coolant from the fuel elements and the absorber in the case of boiling-water reactors is
TABLE I. **3H-PRODUCTION IN VARIOUS TYPES OF REACTORS**

<table>
<thead>
<tr>
<th>Significance</th>
<th>3H-production (Ci/MW(e) yearly)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>BWR</td>
</tr>
<tr>
<td>Efficiency</td>
<td>0.33</td>
</tr>
<tr>
<td>Fission</td>
<td>18</td>
</tr>
<tr>
<td>6Li in fuel</td>
<td>0.25</td>
</tr>
<tr>
<td>9Be in fuel</td>
<td>4 E5</td>
</tr>
<tr>
<td>6Li in graphite</td>
<td>0.5</td>
</tr>
<tr>
<td>9Be in graphite</td>
<td>1 E4</td>
</tr>
<tr>
<td>3H in water</td>
<td>8 E3</td>
</tr>
<tr>
<td>4He in water</td>
<td>4 E3</td>
</tr>
<tr>
<td>6Li in water</td>
<td>0.8</td>
</tr>
<tr>
<td>9Be in water</td>
<td></td>
</tr>
<tr>
<td>6Li in sodium</td>
<td>1</td>
</tr>
<tr>
<td>9Be in sodium</td>
<td>2</td>
</tr>
<tr>
<td>6Li in sodium</td>
<td>0.01</td>
</tr>
<tr>
<td>9Be in sodium</td>
<td>0.01</td>
</tr>
<tr>
<td>Absorber rod</td>
<td>2</td>
</tr>
</tbody>
</table>

*a* Without 3H which remains in the reactor, e.g. reflector.

*b* No boron in absorber rods.

released into the atmosphere and the surface water. Measurements have shown that about 0.1% of the tritium produced diffuses from the fuel elements with zirconium canning and 1% from the fuel elements with steel canning. The diffusion rate from the steel-canned absorber rods of boiling-water reactors is smaller than 1%. Therefore one can expect, according to the data in Table I, emission rates of 200 Ci/a for nuclear power plants with boiling-water reactors of 1000 MW(e) and 900 Ci/a (in the form of HTO) for nuclear power plants with pressurized-water reactors. The greater tritium emission rate in pressurized-water reactors results from the formation of 3H due to the boron added to the water, which was assumed to be 500 ppm. No 3H is produced in the absorber rods of pressurized-water reactors as the absorber material is a mixture of Ag, In and Cd. A separation of this emission into atmosphere and surface water is very difficult. One can expect emission rates as given in Table II, which are confirmed by measurements [2, 3].

The emission rate of high-temperature reactors is mainly influenced by the absorption of 3H in the graphite [4]. 3H produced by 3He is partly fixed by recoil processes in the graphite of the fuel elements. Only a very small part of the tritium formed by fission is released from the coated particles. The emission rate is essentially dependent upon the lithium impurities which were assumed to be as
TABLE II. $^3$H-EMISSION RATES FROM NUCLEAR POWER REACTORS UNDER NORMAL OPERATION AND REPROCESSING PLANTS WITHOUT $^3$H-RETENTION

<table>
<thead>
<tr>
<th>Nuclear facility</th>
<th>Emission rate (Ci/a)</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Atmosphere</td>
<td>Surface water</td>
</tr>
<tr>
<td>Reactor (1000 MW(e))</td>
<td></td>
<td></td>
</tr>
<tr>
<td>BWR</td>
<td>30</td>
<td>150</td>
</tr>
<tr>
<td>PWR</td>
<td>20</td>
<td>900</td>
</tr>
<tr>
<td>HTR</td>
<td>10</td>
<td>900</td>
</tr>
<tr>
<td>FBR</td>
<td>a</td>
<td>200</td>
</tr>
<tr>
<td>Reprocessing plant (40000 MW(e) full load)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>BWR and PWR b</td>
<td>$7 \times 10^5$</td>
<td>1000</td>
</tr>
<tr>
<td>HTR</td>
<td>$6 \times 10^5$</td>
<td>1000</td>
</tr>
<tr>
<td>FBR</td>
<td>$6 \times 10^5$</td>
<td>1000</td>
</tr>
</tbody>
</table>

a $^3$H-leakage from fuel elements in sodium 50% which is stored.
b Without $^3$H-retention facility, lowest amount emitted in surface water.

low as 0.02 ppm [1]. HTO produced by steam generator leakages (approximately 10 g/h) in the primary circuit is retained in the gas purification plants and can be ultimately disposed of under control. A large part of the gaseous $^3$H in the primary cooling circuit diffuses, however, through the heat exchanger into the secondary circuit and from there reaches the environment with the waste water. On the basis of data given in Table I and measurements on the AVR reactor [5], one can expect the emission rates given in Table II for large HTRS in the future. For process-heat reactors in the future, the $^3$H-transport into the process gas that has been estimated in Ref. [6] to be 160 Ci annually has to be taken into account.

For fast breeders it is assumed that 50% of the tritium produced in the fuel elements diffuses into sodium. The emission rate is essentially dependent upon the efficiency of the $^3$H traps. Presupposing a very high efficiency, only the decontamination facilities for repairing components that are washed with water can be taken into consideration. The quantity of $^3$H diffused into the steam circuit via the intermediate circuit is very difficult to estimate. From Refs [7, 8], one can expect the emission rates given in Table II.

As heavy-water reactors, MAGNOX reactors, and AGRs are of little importance, these reactors will not be taken into further consideration. The emission rate of heavy-water reactors is mainly dependent upon the retention of the heavy water.

During the process of dissolving the fuel in reprocessing plants, the tritium contained in the fuel is completely released. That part which has diffused into the canning remains there. Actually, no $^3$H-retention facilities are planned for
operating reprocessing plants. Only for the planned first large reprocessing plant in the Federal Republic of Germany, a $^3$H-retention of about 80% is projected [9]. If no $^3$H-retention and no controlled ultimate disposal of $^3$H is assumed, the emission rates given in Table II result for reprocessing plants with a capacity of 40 000 MW(e) (1500 t LWR fuel elements annually). The $^3$H-emission into waste water as given is probably the smallest achievable at a justifiable expense.

For cost reasons only the following possibilities to retain $^3$H exist:

(a) Enrichment of tritium by recycling the process waters and ultimate storage of quantities of the order of 1000 m$^3$ annually [9].

(b) Enrichment of tritium by recycling and further concentration by rectification [10].

(c) Collecting of the tritium before dissolving the fuel by voloxidation [11].

The emission rate from reprocessing plants is thus dependent on the development of the retention facilities.

4. RADIATION EXPOSURE IN THE VICINITY OF A LARGE REPROCESSING PLANT

In order to show the problems that are caused by the total emission of the tritium inventory of a large reprocessing plant, Fig.8 shows the average concentration of tritium in the ground level air in the main wind direction, that is in an eastern direction for a site near Hanover, in the north east of the Federal Republic of Germany. The first large reprocessing plant in the Federal Republic of Germany is planned in this area [9]. The average atmospheric humidity per year is 7.5 g H$_2$O/m$^3$ air. The calculation of the atmospheric humidity at ground level is based on the Gaussian plume model limiting the height of the mixture. The three-dimensional weather statistics originate from the German Weather Service. New data for the diffusion parameters have been used for the 100-m high emitters of the Nuclear Research Center Jülich [12], which are valuable for a similar ground profile. The figure shows that the maximum tritium concentrations in the atmospheric humidity are much higher than those measured at the time of the maximum of the atomic bomb fall-out in central Europe [13].

The surface water concentration would be about 300 pCi/litre from a release of 1000 Ci/a and a mixture volume of 100 m$^3$/s; the Elbe has an average water flow of 700 m$^3$/s.

To show the importance of this tritium concentration for the population in the vicinity of a large reprocessing plant, Table III gives the maximum possible radiation exposure of an adult according to the methods of calculation used in the Federal Republic of Germany [14, 15], taking into account extreme consumption habits. For the calculation of the air pathway the model of Anspaugh et al. [16]
FIG. 8. Tritium concentration of the air humidity in the main wind direction (east) depending on the distance from a reprocessing plant near Hanover, $E = 7 \times 10^5$ Ci annually.

TABLE III. MAXIMUM RADIATION EXPOSURE OF AN ADULT FROM GASEOUS AND LIQUID RELEASES IN THE NEIGHBOURHOOD OF NUCLEAR FACILITIES NEAR HANOVER

<table>
<thead>
<tr>
<th>Nuclear facility</th>
<th>Maximum radiation exposure of total body (mrem/a)</th>
<th>Atmosphere</th>
<th>Surface water</th>
<th>$\bar{y} = 100$ m$^2$/s</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$H = 100$ m</td>
<td>$H = 200$ m</td>
<td>$3H$ only</td>
<td>total</td>
</tr>
<tr>
<td>Reactor (1000 MWel)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BWR</td>
<td>2 E3</td>
<td>0.1</td>
<td>3 E4</td>
<td>0.02</td>
</tr>
<tr>
<td>PWR</td>
<td>1 E3</td>
<td>0.1</td>
<td>2 E4</td>
<td>0.02</td>
</tr>
<tr>
<td>HTR</td>
<td>6 E4</td>
<td>0.04</td>
<td>1 E4</td>
<td>6 E3</td>
</tr>
<tr>
<td>FBR</td>
<td>6 E3</td>
<td>0.2</td>
<td>1 E3</td>
<td>0.04</td>
</tr>
<tr>
<td>Reprocessing plant (40000 MWel)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BWR, PWR</td>
<td>41</td>
<td>65</td>
<td>7</td>
<td>11</td>
</tr>
<tr>
<td>HTR</td>
<td>35</td>
<td>100</td>
<td>6</td>
<td>17</td>
</tr>
<tr>
<td>FBR</td>
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<td>47</td>
<td>6</td>
<td>8</td>
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</tbody>
</table>

* Retention factor for aerosols 1 E8; iodine 100; $^3H$, $^{14}C$ and $^{85}Kr$, 1.
FIG. 9. Maximum radiation exposure of an adult in the main wind direction (east) depending on the distance from a reprocessing plant near Hanover, $E_{100\%} = 7 \times 10^5$ Ci annually. Emission height 200 m.

FIG. 10. Maximum radiation exposure of an adult in the main wind direction (east) depending on the distance from a reprocessing plant near Hanover, $E_{100\%} = 7 \times 10^5$ Ci annually. Emission height 200 m.
is used but without the factor 0.5 for the influence of non-tritiated water. The $^3$H-total-body dose factor that has to be used is according to Ref. [17] 230 rem/Ci for inhalation and 130 rem/Ci for ingestion [14]; this factor includes a quality factor of 1.7. Table III shows that with an emission height of 100 m and an emission rate as shown in Table II, maximum doses of 40 mrem/a are to be expected. The radiation protection ordinance [18] of the Federal Republic of Germany demands that the additional radiation exposure from emissions of nuclear power plants should be as low as possible, and should not exceed 30 mrem/a for the total body as well as for the air pathway and the water pathway. Therefore the contaminated waste air has to be released via a 200-m stack, to remain within the limits of the radiation protection ordinance in the vicinity of reprocessing plants.
FIG. 13. Maximum radiation exposure (mrem/a) of an adult in the neighbourhood of a reprocessing plant near Hanover, $E = 7 \times 10^5$ Ci annually, height = 200 m.

The calculated dose via the air pathway is shown in Figs 9, 10, 12 and 13, and the depletion of the ground level air concentration in the main wind direction in Fig. 11. In order to calculate the wash-out near the emitter, which is not considered in Ref. [16], a wash-out constant during a rainfall of $3 \times 10^{-8}$ a/(mm-s) [19] was assumed. With the assumption that the tritium concentration in the plants adapts very quickly during the rain period (5%) or the rainless period to the rain or air concentration (according to Ref. [16] the half-life in plants is approximately 1 d) and assuming that the harvest of the plants is distributed equally among the rain periods and the dry periods, the effective $^3$H-concentration in the plants is, according to the formula for wash-out [14], with an emission rate of $7 \times 10^5$ Ci/a

$$C_{eff}^i = 2 \times 10^{-3} \frac{\text{Ci}}{1} \times \frac{P_i}{u \cdot x}$$

where $P_i$ = precipitation frequency with wind direction i
$u$ = average wind velocity in m/s
$x$ = distance from emitter in m

For dose calculations it was assumed that during one year ground products are daily ingested with 0.5 litre water contents; according to Ref. [17] the total water uptake via food is 1 litre/d. Figures 9 and 10 show the dose from wash-out in the main wind direction.
5. RADIATION EXPOSURE FROM GLOBALLY DISTRIBUTED TRITIUM

Almost all the tritium released finally reaches the oceans. Figure 14 shows the usual model for estimating the dose from globally distributed tritium, which is more realistic for air-borne emissions such as nuclear weapon fall-out than local emissions via river water. Selecting the mixing times shown in Fig. 14 [20], one can expect doses in the future from globally distributed $^3$H as shown in Fig. 15. In order to be on the safe side in the calculations, a very high increase in nuclear power plants [21] was assumed in the northern as well as the southern hemisphere. The specific activity model was used for dose calculations. The contribution of globally distributed $^3$H to the total body dose is negligible compared with the contribution of the first pass exposure in the vicinity of one emitter.

6. COLLECTIVE DOSE FROM TRITIUM EMISSIONS

It was shown in Table III that it is possible to lower the maximum radiation exposure by increasing the height of the stack. The collective dose, however, remains almost the same.

In order to calculate the collective dose from the first pass exposure, the calculation was divided into three: (1) radiation exposure from tritium dispersion in the atmosphere including the depletion of the radioactive plume; (2) radiation exposure from drinking precipitation; (3) radiation exposure from precipitation of precipitated water. In Section 4 the calculation for the first pass exposure was described. Mean values have been assumed for consumption habits. The calculation gives a collective dose of 3000 man · rem based on an annual emission of 7 E5 Ci until the total tritium has precipitated. Approximately 6000 man · rem can be
FIG. 15. Expected radiation exposure in the northern hemisphere from globally distributed tritium from nuclear power plants and reprocessing plants.

TABLE IV. APPROXIMATE CONTRIBUTION TO THE COLLECTIVE DOSE OF VARIOUS COUNTRIES FROM THE FIRST PASS EXPOSURE FOR AN EMITTENT NEAR HANOVER

\( (S_{\text{first pass}} (\text{H}) \approx 0.017 \text{ man-rem/Ci}) \)

<table>
<thead>
<tr>
<th>Country</th>
<th>Contribution</th>
<th>Country</th>
<th>Contribution</th>
</tr>
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<tbody>
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<td>Luxemburg</td>
<td>0.1</td>
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<tr>
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<tr>
<td>Germany, Federal Republic of</td>
<td>30</td>
<td>United Kingdom</td>
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</tbody>
</table>
calculated via drinking water with an average rainfall of 700 mm yearly in Europe. In order to estimate the contribution of evaporated water the calculation was based on the water cycle in the Federal Republic of Germany [22]. According to this, about 50% of the rainfall evaporates on the ground; the other 50% comes from the sea. After the first rainfall about half the tritium is dispersed into the sea. Because of the non-tritiated seawater a dilution factor of two results. Continuing the calculation like a chain, the contribution of evaporated tritium is \(0.5 \times (3000 + 6000)\) man·rem. A part of the evaporated tritium, however, is precipitated into the oceans so that a total of 3000 man·rem can be expected. With an emission rate of \(7 \times 10^5\) Ci the collective dose from the first pass exposure is then 12000 man·rem or approximately 0.017 man·rem/Ci tritium. Table IV shows the distribution among the different countries. It is interesting that the same collective doses can be stated if a constant wind distribution is assumed.

Because of small tritium emission rates into rivers (Table II) the collective dose for this emission is not discussed here.
The global contribution has been calculated according to the model in Fig. 14 (see Section 5). The release takes place in the northern hemisphere. Figure 16 shows the results in dependence on the integration time up to the final decay. Compared with the contribution from the first pass exposure, the global contribution is negligibly small — approximately smaller by a factor of 20.

According to the new ICRP concept [23], the collective dose is the most essential value for radiation protection, if the maximum individual dose is smaller than the maximum permissible values. Therefore, in order to reduce the collective dose, the emission rates of reprocessing plants must be reduced and the tritium retained should be distributed to the oceans for the present, if injection into deep earth cavities is not feasible. This should be continued until methods are developed for further reduction of the collective dose.

7. CONCLUSIONS

If the total tritium of a large reprocessing plant is released via a 100-m-high stack, the maximum radiation exposure exceeds the permissible total body dose of 30 mrem yearly in the Federal Republic of Germany. By increasing the height of the stack, the maximum radiation exposure only can be reduced, the collective dose, however, remains practically constant. Assuming costs of detriment at 200 DM/man-rem = 100 $/man-rem, the emission from a large reprocessing plant gives a cost of detriment of DM 2.4 E6 ≈ $ 1.2 E6. According to the unfinished cost-benefit analysis, up to 80% of the tritium can be retained in reprocessing plants for these costs. So far we have concentrated on $^3$H, $^{85}$Kr and $^{139}$I. But large reprocessing plants release more than 500 Ci $^{14}$C per year [24], the collective dose from this, mainly due to global distribution, being more than 50 times higher than that due to $^3$H (Fig. 16). For radiation protection reasons we should concentrate more on studying $^{14}$C.

ACKNOWLEDGEMENT

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REFERENCES

DISCUSSION

P. BOVARD: You refer to your intention to retain the tritium so as to reduce the collective dose. What method do you propose to use for this retention process, and as from what capacity of plant would you recommend that it be applied?

Also, have you carried out any comparative studies of the costs of higher stacks and of processing, and at what height of stack does it become cheaper to process the tritium for retention?

H. BONKA: In all our studies we consider large reprocessing plants with a capacity of 1500 t LWR fuel. The cheapest way to lower the gaseous HTO emissions is to cool the air. If the air contains tritium in the form of HT or T₂ (especially in the case of the emissions from high-activity waste storage), it is necessary to oxidize the tritium. The best method of handling the tritiated waste water is to recycle the process water.

In reply to your second question, as shown in my paper it is virtually impossible to lower the collective dose by increasing the height of the stack. Such an increase only lowers individual doses in the immediate vicinity. At a distance of about 10 km the individual dose is practically independent of the height of the point of emission. Considerations of stack costs are therefore largely irrelevant.

W. ROETHER: You have given figures for releases of tritium from reprocessing plants in the form of liquid versus gaseous effluents. On what evidence are your figures based and how reliable are they?

H. BONKA: In the Federal Republic of Germany the first big reprocessing plant will be located near a large salt pile for purposes of deposition of the radioactive waste. The location, Gorleben, in the northeastern part of the country is far away from the open sea with good water-mixing conditions. It is impossible to discharge all the °H into the River Elbe without exceeding the dose limits established by the Radiation Protection Ordinance of the FRG. As I have just said, by increasing the height of the stack it is possible to lower individual doses. The Radiation Protection Ordinance lays down no limits for collective doses. Therefore all the gaseous °H, in the form of HTO, HT and so on, will be emitted from a stack. Only liquids with very low °H concentrations will be discharged to surface waters.
For a location by the open sea I consider that at present the best procedure is to discharge the \(^{3}\text{H}\) in a controlled manner into the sea, as I propose in my paper when discussing the lowering of the collective dose. The emission rates stated are approximate values for the first reprocessing plant in the Federal Republic of Germany.

J.A. HETHERINGTON: You estimate that the annual detrimental cost of discharging the tritium waste from your envisaged power programme will be about \$1.2 \text{E6, and you claim that cost/benefit considerations suggest that it will be economically practicable to retain 80\% of the activity arising. Can you say what means you will use to avoid the need to discharge? How will 80\% of the tritium be retained at reprocessing, for example?}

H. BONKA: At reprocessing plants we have two different sources of \(^{3}\text{H}\) release: the reprocessing plant itself and the high-activity waste store. To reduce the gaseous releases the cheapest method is to cool the air and retain the \(^{3}\text{H}\) gaseous emissions in aqueous form. However, the greatest releases emanate from storage of the high-activity waste. Here we have to oxidize \text{HT} and \text{T}_2 before we retain it by cooling. At the moment I am myself not certain whether it is preferable continuously to blow fresh air through the storage facility, which means an open cycle, or to have a closed cycle arrangement. The tritiated water can be injected into deep geological strata or discharged, with good mixing, into the sea; as I have said, this is at present the procedure to be preferred.

E.L. COMPERE: Can you compare the effects of \(^{85}\text{Kr}\) and tritium releases from reprocessing plants? In the United States of America we do not reprocess commercially at present and have as yet no definitive restriction on tritium release. We do, however, have a limit on \(^{85}\text{Kr}\) release.

H. BONKA: The \(^{85}\text{Kr}\) emission rate from a reprocessing plant with a capacity of 1500 t LWR fuel yearly is, in the absence of a retention facility, nearly \(2 \text{E7 Ci annually. The contribution to the collective dose associated with the first pass exposure is negligible compared with the contribution from globally distributed }^{85}\text{Kr}.\)

According to Fig.16 of my paper, the collective dose to the skin from beta rays is approximately \(1 \text{E6 man}\cdot\text{rem} \) and to the total body from gamma rays \(1.3 \text{E4 man}\cdot\text{rem}.\) The equivalent total body collective dose is, applying a weighting factor for the skin of 0.01, in accordance with the latest ICRP recommendation, \(10,000 \text{man}\cdot\text{rem} + 13,000 \text{man}\cdot\text{rem} = 2.3 \text{E4 man}\cdot\text{rem}.\) According to my paper the equivalent total body collective dose from the overall \(^{3}\text{H}\) emission is \(1.24 \text{E4 man}\cdot\text{rem}.\) That means that the total \(^{85}\text{Kr}\) emission has double the effect of the total \(^{3}\text{H}\) emission, assuming a linear relationship between dose and detriment.
AN ANALYSIS OF TRITIUM RELEASES TO THE ATMOSPHERE BY A CONTROLLED THERMONUCLEAR REACTOR

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Abstract

AN ANALYSIS OF TRITIUM RELEASES TO THE ATMOSPHERE BY A CONTROLLED THERMONUCLEAR REACTOR.

Atmospheric removal of routinely and accidentally released tritium from a controlled thermonuclear reactor (CTR) has been investigated. The assumed form of the tritium was HTO, or tritiated water vapour. Assuming a CTR operation in midwestern United States of America, surface water and ground-level air concentration values of tritium were computed for three space (or time) scales: local (within 50 km of a reactor site), regional (up to 1000 km from the site), and global. The local and regional analyses use a Gaussian plume model with a factor incorporating scavenging of tritium by precipitation. Results show that within 50 km of the site atmospheric concentrations of tritium will be the limiting factor for routine releases. On the regional scale, surface water concentrations tend to become the limiting factor. An analysis for an accidental release of tritium shows surface water concentrations in the vicinity of the reactor site to be quite high after rainfall. The global analysis utilizes a linear set of differential equations describing the atmospheric transport of tritiated water vapour. Solutions are obtained using the assumption that the amount of precipitable water vapour and surface water in a 10 deg latitude belt remain constant over an extended period of time. The solutions are studied as a function of various input parameters. In particular the variation of surface water depth, which is dependent on the distribution of water and land in each latitude belt, is factored into the equations. The atmospheric and surface water concentrations of tritium obtained are latitude-average values.

INTRODUCTION

A Controlled Thermonuclear Reactor (CTR) is a potential source of tritium to the environment. Tritium can be released from a CTR either in its elemental form, such as HT, or as tritiated water vapor, HTO. Jacobs [1] cites many references which show that the predominant form of tritium in the atmosphere is in tritiated water vapor, and that tritium released to the atmosphere in its elemental form is quickly converted to tritiated water vapor. Therefore in this analysis it is

* Operated by Battelle Memorial Institute for the U.S. Department of Energy.
assumed that all tritium released from a CTR into the environment is in the form of tritiated water vapor, HTO.

Once emitted tritium takes the form of tritiated water vapor its properties are essentially the same as those of atmospheric water vapor. Thus tritium becomes a component of the hydrologic cycle, and behaves essentially the same as water vapor in the hydrologic cycle. The removal of tritium from the atmosphere occurs primarily through the precipitation process, although near the point of release some tritiated water vapor can be diffused to the ground and taken up directly by vegetation. Once deposited, the tritium can infiltrate into the ground water, remain as part of the surface runoff and storage, be assimilated into plants, or transpired and evaporated back into the atmosphere.

Tritium concentration in the hydrosphere can be analyzed in three different space (or time) scales [2]. The first scale studies ground-level and surface water tritium concentrations within 50 km of a proposed CTR under normal operating conditions. The second scale studies regional ground-level atmospheric concentrations (up to 1000 km) at any given time during normal operation of a CTR. The third scale examines the expected long-term global concentrations in the air and surface water supply. In addition, an analysis of surface water concentration near the CTR after an accidental release of tritium during a rainfall episode is presented.

ROUTINE RELEASES

The technique for computing average annual ground-level concentrations of tritium within 50 km of a CTR power plant involves evaluation of the Gaussian plume formulation:

$$\frac{x_a}{Q} = \frac{1}{\pi \sigma_y \sigma_z u} \exp \left( \frac{-y^2}{2 \sigma_y^2} \right) \exp \left[ -k_{eq} \frac{x}{u} \right]$$ (1)

where:

- $x_a$ = atmospheric tritium concentration ($ci/m^3$)
- $Q$ = source term to the atmosphere ($ci/yr$)
- $\sigma_y = 0.5t$ = horizontal standard deviation of the plume (meters)
- $\sigma_z = [2K_v(x/u)^2]^{1/2}$ = vertical standard deviation of the plume (meters)
- $t$ = plume travel time (sec)
- $K_v$ = vertical diffusion coefficient ($m^2/sec$)
- $u$ = mean wind speed affecting the plume (m/sec)
- $k_{eq}$ = equilibrium scavenging coefficient (1/sec)
- $x$ = downwind distance from the source (meters)
- $y$ = crosswind distance (meters)

The second exponential term in equation (1) represents the depletion of the plume due to scavenging of tritium by precipitation. Under equilibrium conditions, where the in-cloud and
below-cloud tritium concentrations are equal, and where the
elevation of the release is at ground-level, the equilibrium
scavenging coefficient, $k_{eq}$, can be written [3]:

$$k_{eq} = \left(\frac{2}{\pi}\right)^{1/2} \frac{\rho_w J}{\rho_a \sigma_z}$$

where:

- $\sigma_z' = $ Smith Singer dispersion coefficient
  - $0.63x^{0.86}$, $x \leq 4.4$ km
  - $859$ m, $x > 4.4$ km
- $\rho_a = $ absolute humidity of $H_2O$ (gm/cm$^3$)
- $\rho_w = $ density of water (gm/cm$^3$)
- $J = $ rainfall rate (m/sec)

The results of the ground-level atmospheric tritium con-
centration estimates are shown in Figure 1. The concentrations
in this figure have been normalized to a one curie per year
release. To determine the concentrations for any source rate
the normalized concentrations in the figure must be multiplied
by that source rate. The climatology of wind trajectories for
a site in the midwestern United States was used in this analysis.
FIG. 2. Normalized surface water concentration (X/WQ) as a function of distance (x).

FIG. 3. Ground-level air concentration values of tritium for the eastern United States of America.
The average annual surface water tritium concentrations, $X_g$, are determined from:

$$X_g = \frac{1 - \exp(-k_{eq}(X/\bar{u}))}{AW}$$  \hspace{1cm} (3)

where:

$W =$ average annual depth of the surface or ground water mixing layer, (m/yr)

$A =$ area of precipitation (m$^2$)

Since this analysis is intended to provide conservative estimates of tritium concentrations that would show the maximum expected impacts for normal operation and accidental releases, the value of $W$ was assumed to be determined from the mean annual net rainfall:

$AW = P - E$

where:

$P =$ amount of precipitation (m$^3$/yr)

$E =$ amount of evaporation (m$^3$/yr)

Thus, this method assumes no ground water reservoir into which tritium would be mixed after tritiated rainfall percolates through the ground.

Figure 2 shows the variation of surface water concentration with distance from a CTR. Again the results shown are normalized. An annual rainfall of about one meter per year and
evaporation of about one-half meter per year were used for the hydrologic parameters. This figure shows that the smaller dispersion within 4.4 km of the CTR acts to increase surface water concentrations by as much as two orders of magnitude above what would have been estimated from a constant dispersion.

The regional scale deals with plume trajectories from a CTR over a travel time of several days, or distances up to 1000 km. Figure 3 shows the results of the regional ground-level air concentration estimates. A climatology of air trajectories over the eastern United States was used in this analysis [4]. Figure 2 shows the surface water tritium concentrations out to a distance of 1000 km. These estimates of surface water concentrations were made using concentric areal rings around the CTR site. Therefore, the concentrations in Figure 2 are limited to distances up to 1000 km.

**ACCIDENTAL RELEASE**

The analysis procedure involves the same basic approach that was used to estimate surface water concentrations during routine operations. This approach, assuming equilibrium conditions, will produce conservative results for distances near the emission source. The relation for determining surface water tritium concentrations, $x_g$, is given by:

$$\frac{x_g}{Q_t} = 1 - \exp \left[ -k \frac{\sigma}{\sigma \text{eq}} \right]$$

(5)
where:

\[ Q_t = \text{the total amount of tritium released in the accident (gm or Ci)} \]

\[ k_{eq} = \text{is defined by equation (2)} \]

\[ W = Jt \ (\text{no mixing with surface or groundwater systems}) \]

\[ J = \text{the rainfall rate during the accidental release} \]

\[ t = \text{length of the storm period} \]

The effect of rainfall rate on surface water concentrations at various distances from the site is shown in Figure 4. Since no ground water mixing is assumed a large rainfall rate actually represents greater dilution, with highest concentrations occurring for light rainsfalls. However, many other factors enter into these computations, including the manner in which the scavenging coefficient is computed and the depth of surface water available for mixing with rainwater.

Investigation of environmental conditions that might exist during a CTR accident could result in even greater impacts. These investigations require a detailed statistical evaluation of climatological data, as well as a complete analysis of potential CTR accident scenarios.

GLOBAL TRANSPORT

The balance equation for tritium in the atmosphere can be developed by examining Figure 5, which shows the pertinent variables involved. The subscript \( i \) denotes any of the 10° latitude belts where \( i = 1 \) represents the 80°-90° N latitude belt, \( i = 2 \) represents the 70°-80° N latitude belt, etc. The equation is written:

\[
\frac{d}{dt} \left[ AW_a x_a \right]_{(i)} = \left[ Q_a + EA x_a - FA x_a - AW_a x_a / 18 \right]_{(i)}
+ V_{in}(i) x_a(i-1/2) \text{sgn} V_{in}(i) / 1/2
- V_{out}(i) x_a(i+1/2) \text{sgn} V_{out}(i) / 1/2
\]

(6)

where:

\[ P = \text{latitude-averaged precipitation (m of H}_2\text{O/yr)} \]

\[ E = \text{latitude-averaged evaporation (m of H}_2\text{O/yr)} \]

\[ W_a = \text{latitude-averaged atmospheric water vapor (Precipitable water)} \ (\text{m of H}_2\text{O)} \]

\[ A = \text{area of latitude belt (m}^2) \]

\[ V_{in} = \text{water vapor exchange between (i) and (i-1)} \]

\[ V_{out} = \text{water vapor exchange between (i) and (i+1)} \]

\[ \text{latitude belt (m}^3 \text{ of H}_2\text{O/yr)} \]
\[ t = \text{time (yr)} \]
\[ Q_a = \text{source of tritium to atmosphere, distributed throughout the latitude belt (gm of HTO/yr)} \]
\[ \chi_a = \text{concentration of tritium in atmospheric water vapor (gm of HTO/m}^3\text{ of H}_2\text{O)} \]

The subscripts of \( \chi_a \) in the last two terms are written so that the concentration of the appropriate latitude belt, which depends on the direction of flow, is selected. The term with the factor of 18 in the denominator represents the radioactive decay with 18 years being the mean life of tritium [5]. The units of \( \chi_a \) were chosen so that the magnitudes of the tritium concentrations in the atmosphere and surface water would be more nearly the same order of magnitude. This facilitates the solution of the set of equations obtained.

Similarly, the balance equation for tritium in the surface water can be developed and is written:

\[
\frac{d}{dt} \left[ AW_s \chi_g (i) \right] = \left[ Q_s + PA \chi_a - E \chi_g - AW_s \chi_g / 18 \right] J(i) \\
+ S_{\text{in}}(i) \chi_g (i-1/2) - S_{\text{out}}(i) \chi_g (i+1/2)
\]

where:

\[ W_s = \text{latitude-averaged surface water (m of H}_2\text{O)} \]
\[ S_{\text{in}} = \text{surface water exchange between (i) and (i-1) latitude belt (m}^3\text{ of H}_2\text{O/yr)} \]
\[ S_{\text{out}} = \text{surface water exchange between (i) and (i+1) latitude belt (m}^3\text{ of H}_2\text{O/yr)} \]
\[ Q_s = \text{source of tritium to surface water (gm of HTO/yr)} \]
\[ \chi_g = \text{concentration of tritium in surface water (gm of HTO/m}^3\text{ of H}_2\text{O)} \]

With the assumption that \( W_a, W_s \) and \( A \) are independent of time \( t \) and using the necessary boundary conditions between adjacent latitude belts, equations (6) and (7) can be written in the more compact form [5]:

\[
\frac{d}{dt} [X_i] = \sum_{j=1}^{36} (a_{ij} X_j) + b_i
\]

where:

\[ X_i = \text{tritium concentration (gm of HTO/m}^3\text{ of H}_2\text{O)} \]
\[ b_i = \text{source terms of tritium ((gm of HTO/m}^3\text{ of H}_2\text{O)/yr)} \]
\[ a_{ij} = \text{coefficients determined by constant parameters (1/yr)} \]
The subscript of $X$ has the following meaning; for $i$ from 1 to 18, concentrations in the precipitable water; for $i$ from 19 to 36, concentrations in the surface water. The coefficients $a_{ij}$ compose a non-symmetric matrix consisting of the tridiagonal terms plus a few terms farther out.

If the $b_i$ are assumed to be constant, the complete solution of the set of differential equations has the form:

$$X_i = \sum_{j=1}^{36} C_j e^{\lambda_j t} + K_i \quad (9)$$

where the $C_j$ are determined by initial conditions and $K_i$ is a constant.

Climatological data [7] and physical data [8] were obtained for each 10° latitude belt. Since the amount of water and land varies between latitude belts, three different cases of the surface water parameter were investigated.
FIG. 7. HTO concentration in atmospheric water vapor for three cases of surface water depth with release into atmosphere (—) or surface water (---). Concentration and release in latitude belt 40°–50°N.

1) uniform depth of 0.5 meter, simulating all land areas.

2) uniform depth of 75.0 meter, simulating all ocean areas.

3) non-uniform depth, where the depth for each latitude belt is determined by a linear weighting of the actual water and land areas.

The roots, λ_j, of the characteristic equation are inversely related to a turnover time. Taking a median value of those roots pertaining to the precipitable water vapor a turnover time of 7.7 days is obtained, which is close to the global value of 8 days [7]. For the non-uniform surface water case the range of turnover times is 7.6 to 17.9 years, which is comparable to turnover times for the North Pacific of 9 to 15 years [9].

Figure 6 shows how the percent of steady-state HTO concentration in the surface water varies with time. There is little dependence on surface water depth and negligible dependence on whether the release is into the atmosphere or surface water.
FIG. 8. Steady-state HTO concentration in atmospheric water vapor for three cases of surface water depth with release into atmosphere (---) or surface water (-----) in latitude belt 40°–50° N.

FIG. 9. Steady-state HTO concentration in surface water for three cases of surface water depth with release into atmosphere (---) or surface water (-----) in latitude belt 40°–50° N.
In Figure 7 the percent of steady state HTO concentration in the precipitable water vapor versus time is plotted. This plot shows that the build up of concentration is very fast with larger surface water depths for releases into the atmosphere.

Figures 8 and 9 show the distribution of HTO concentration over the earth for a release in the 40-50° N latitude belt. All values of concentration have been normalized to the concentration in the source latitude belt. Thus in Figure 8 for release into the atmosphere the curves are normalized by the water vapor concentration in latitude belt 40-50° N; and for release into the surface water the curves are normalized by the surface water concentration in latitude belt 40-50° N. Figures 8 and 9 show that the concentrations are significant only locally for larger surface water depths.

CONCLUSIONS

For routine releases of tritium the Gaussian model developed shows that surface water concentrations on the regional scale and atmospheric concentrations on the local scale are most important. During precipitation high concentrations in the surface water can occur for an accidental release due to scavenging of the tritium.

Global tritium concentrations in the atmosphere and surface water are highly dependent on the selection of the surface water depth used in the model, with higher concentrations occurring with smaller depths. This suggests an advantage to siting CTR's near large bodies of water. The model results also show that it is more advantageous to release tritium directly into the atmosphere, although differences in concentrations between these two release models are not large.

More sophisticated modeling techniques for transport over several hundreds of kilometers are required, particularly if complex terrain and land-sea contrasts are to be included. For example, some success has been obtained using numerical boundary layer models to estimate tritium concentrations in the vicinity of the Savannah River Laboratory [19]. The manner in which HTO enters into the hydrologic cycle, and the similarities between HTO and H₂O need further quantification. This becomes particularly important when examining the scavenging process of HTO by precipitation, and the subsequent return to the atmosphere via the evapotranspiration process. Research into the probabilities of large accidental releases during unfavorable meteorological episodes needs to be undertaken.

A more refined global model would consider the actual land and ocean distributions. One step in this direction would divide each latitude belt into longitude-latitude areas. Each area could then be considered entirely land or ocean. Also based on some atmospheric tritium measurements [11], [12] a two layer model of the atmosphere could be developed.

REFERENCES


LASER FUSION

Status, future and tritium control*

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California,
United States of America

Abstract

LASER FUSION: STATUS, FUTURE AND TRITIUM CONTROL.

The laser fusion programme today is being advanced by remarkable new developments in laser technology and new understandings of fusion physics. Steady progress in these areas has formed a solid basis for scientific feasibility demonstrations which will lead to the development of reactor scale facilities. At Livermore the 10 kJ, 20–30 TW Shiva facility is now operational and producing regular new fusion results. Design work has begun on a 200–300 TW laser designed to carry the programme through the first breakeven demonstration experiments in the mid-1980s. Confidence in reaching this goal is based on the significant progress that has been made in state-of-the-art, high-power Nd:glass laser technology, in experimental laser fusion and laser plasma interaction physics, and in theoretical and analytical computer codes which reliably model and predict experimental results. For all of these experiments, a variety of fusion targets are being fabricated in the laboratory, and the control and handling of tritium is now a regular and routine part of ongoing inertial fusion experiments. Target design with gains of about 1000 have been studied and the means to mass produce such pellets at low cost are also being developed. Although the reality of laser fusion power is still a long way off, laser fusion reactor systems are being visualized and specific process hardware is being designed and modelled. The complete fuel cycle from the manufacture of pellets loaded with deuterium and tritium to the recovery and recycling of unburned products is being studied and preliminary results are available. As reactor class systems approach, it will be necessary to demonstrate each element of the tritium life cycle. From the work done so far, one can begin to have confidence in the control and management of the tritium source term in future laser fusion power plants.

The concept of laser fusion, also referred to as inertial confinement fusion, is that a sequence of tiny fuel pellets containing deuterium and tritium are projected towards the centre of a reaction chamber where high power laser pulses strike each pellet, heating its fuel and releasing thermonuclear energy (Fig.1). The kinetic energy of neutrons from the fusion reactions is converted in an absorbing blanket to thermal energy which is coupled to make electricity through a normal

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FIG. Inertial confinement fusion concept.
**FIG. 2.** Schematic diagram of a laser fusion power plant and its energy cycle.

**TABLE I. CONDITIONS FOR BURNING DEUTERIUM TRITIUM FUSION FUEL**

<table>
<thead>
<tr>
<th></th>
<th>Magnetic confinement</th>
<th>Inertial confinement</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Lawson breakeven</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>$10^8$ K (10 keV)</td>
<td>$10^8$ K (10 keV)</td>
</tr>
<tr>
<td>$\eta \tau$</td>
<td>$10^{14}$ s $\cdot$ cm$^{-3}$</td>
<td>$10^{14}$ s $\cdot$ cm$^{-3}$</td>
</tr>
<tr>
<td>$\eta$</td>
<td>$\sim 10^{14}$ cm$^{-3}$</td>
<td>$10^{22}$ cm$^{-3}$</td>
</tr>
<tr>
<td>$r$</td>
<td>$\sim 1$ s</td>
<td>$10^{10}$ s</td>
</tr>
<tr>
<td><strong>Fusion power plant</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>$10^8$ K</td>
<td>$10^8$ K</td>
</tr>
<tr>
<td>$\eta \tau$</td>
<td>$\sim 10^{15}$ s $\cdot$ cm$^{-3}$</td>
<td>$\sim 10^{15}$ s $\cdot$ cm$^{-3}$</td>
</tr>
<tr>
<td>$\eta$</td>
<td>$\sim 10^{14}$ cm$^{-3}$</td>
<td>$\sim 3 \times 10^{22}$ cm$^{-3}$</td>
</tr>
<tr>
<td>$r$</td>
<td>$\sim 10$ s</td>
<td>$\sim 3 \times 10^{11}$ s</td>
</tr>
</tbody>
</table>
**FIG. 3.** Laser fusion energy yield projections.

**FIG. 4.** The Argus laser system. The target chamber is at the far end of the room between the two laser chains.
thermal cycle. A schematic representation of this process is shown in Fig. 2. The driver can be a laser, an electron or heavy ion beam, or other appropriate source of energy. The driver rapidly heats the surface of the target, forming a surrounding plasma, which transports energy inward, compressing and heating the fuel until the fuel at the core reaches densities dozens of times that of lead and temperatures of the order of 100,000,000°C. At these temperatures the fuel ignites and thermonuclear reactions spread, multiplying by many times the input energy of the driving source.

Although the conditions required for burning deuterium-tritium fuel are similar in both magnetic fusion and inertial fusion, the means by which those conditions are obtained are quite different. In both approaches to fusion energy, ion temperatures of the order of 10 keV must be reached. In magnetic confinement fusion, the high ion temperature is maintained for relatively long times in a low-density plasma (Table 1). On the other hand, in inertial fusion the fuel is compressed to densities of a 100 grams per cm³ or more but only for a few trillionths of a second. The product of this very high density and short inertial confinement time leads to the same criteria or ‘Lawson’ number as is required in magnetic fusion.

A sequence of increasingly powerful lasers (Fig. 3) has been constructed at Livermore to explore the physics of the interaction of lasers with matter and to achieve key fusion milestones. The sequence began with the Janus laser system in 1974 and progressed through Janus to the Cyclops and Argus laser systems first operational in 1975 and 1976 respectively. The first demonstration of laser-driven thermonuclear burn was achieved at Livermore in 1975 with the Janus
2 μm Glass 22 μm CF₂

On a production basis glass shells are coated with the following specifications

<table>
<thead>
<tr>
<th>Coating</th>
<th>Total surface variation</th>
<th>Mandrel</th>
<th>Concentricity</th>
<th>Ambient surface</th>
<th>Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>CF₁,₃</td>
<td>0.1 – 0.2 μm</td>
<td>140 μm glass shell DT filled</td>
<td>3 – 5%</td>
<td>100 – 200 Å</td>
<td>1 – 30 μm</td>
</tr>
</tbody>
</table>

*FIG. 6. Target quality CF₂ coated glass shell.*

laser system. Cyclops was the laser system testbed on which components for future systems were first prototyped and tested. In particular, the technique of spatial filtering to avoid propagating and amplifying optical noise down each laser chain was first demonstrated on Cyclops. Success with this approach led to multiple spatial filters on Argus and Shiva. A view of the Argus laser is shown in Fig. 4 with the master oscillator, beam splitter and early preamplifier components in the foreground. As each beam is amplified, it is allowed to grow in diameter to avoid damage to optics. At the far end of the room the two beams of 20-cm aperture pass into a target chamber and are focused by lenses on to an experimental target mounted in the centre of the chamber. Argus is still operational and continues to produce many important physical results. Specific target experiments on Argus have yielded up to 2 × 10⁹ neutrons. Since high density is a requirement for inertial fusion, experiments on Argus during 1978 have been carried out with the aim of attaining densities in the range of 10 to 100 times liquid density.

The Shiva system was first operational in late 1977 and has delivered over 10 kJ of energy in a one nanosecond pulse and over 26 TW in shorter pulses. The twenty beams of Shiva are focused in two f/1 clusters of ten beams each which strike the target from opposite sides. Specific targets on Shiva have produced as much as 3 × 10¹⁰ neutrons, and future experiments are also planned for Shiva to
achieve fuel compressions in the 10 to 100 times liquid density range. The achievement of ignition conditions in the thermonuclear fuel with target gain of the order of $10^{-3}$ is termed significant thermonuclear burn and is expected to be achieved first at low density, later at high density, with the Shiva laser system. A further advance, termed 'scientific breakeven', exists when the thermonuclear energy released from a pellet equals the light energy input to the pellet (see Fig.3). To reach scientific breakeven conditions and the first demonstrations of scientific feasibility, the full capabilities of the Nova laser system are required. Nova is an extensive upgrade of Shiva scheduled to begin construction in 1979.

Many simple laser fusion experiments employ deuterium-tritium (DT)-filled glass microspheres as the target. Early targets of this type were as small as 40 microns in diameter. As our laser systems have increased in power and energy, so also the targets have increased in size, and today glass microsphere targets 200–400 microns in diameter are common. Such targets are typically DT-filled to a density of 2 mg/cm$^3$ (approximately 150 psi)$^1$, with the largest

---

$^1$ 1 psi = 1 lbf/in$^2$ = 6.895 $\times$ 10$^3$ Pa.
only 64 nanograms of DT gas. More advanced targets involving successive layers of various materials are required for high performance fusion milestones. In particular, Teflon-coated glass microballoons are now being fabricated for use in target experiments designed to achieve 10 to 100 times liquid density. Surface finish is an important requirement for both the starting glass microsphere in the targets and for successive layers of other materials. We can now routinely achieve surface finishes of some 100 angstroms, hundreds of times better than that available through commercial sources. Coatings must not only be applied with good finish but with uniform concentricity and thickness and uniform composition throughout (Figs 5 and 6).

Results from each experiment depend on a full campaign of target design and fabrication, laser system and diagnostics readiness. Figure 7 shows a specific result from a recent target experiment, an X-ray photomicrograph of the intensity of X-rays emitted from a fusion target. The original target appears as the outermost circular contour in the photomicrograph. The hot-looking central region reveals the most intense portion of the thermonuclear reaction. Within the LLL Program, we are now able to image and analyse spectrally a variety of particles from fusion reactions and are beginning to be able to time-resolve these emissions.

One measure of progress over the last few years is a steadily increasing set of neutron yields from larger and more sophisticated targets. Those neutron yields are shown in Fig. 8 for various target designs alphabetically listed for convenience and for the laser system on which each result was produced. The pre-experiment prediction developed with the aid of the LASNEX physics code is
plotted alongside each experimental point. Over a range of seven orders of magnitude, we have been able to model adequately the output of many different target designs. This capability is important to continued success with high yield targets on Shiva and Nova. If one plots the best of these results from the Argus laser system in terms of confinement density, time and ion temperature, that result can be compared with similar results in the Magnetic Fusion Program (Fig.9). Although both programmes still have many orders of magnitude to go, we can see that today they are producing roughly comparable fusion conditions.

The leading contender to produce scientific breakeven is the Nova laser facility pictured in Fig.10. It consists of approximately 40 beams extending longer into larger apertures than Shiva and constructed as a replicative enlargement of the existing Shiva system. Also using improved laser glass, Nova is being designed to deliver over ten times the power and energy of Shiva. The facility is expected to be fully operational in 1984, making possible first demonstrations of breakeven conditions in inertial fusion by the mid-1980s. In addition to providing the driving energy necessary for scientific breakeven conditions, Nova will also provide a facility where reactor class targets can be studied on a single
FIG. 10. Nova laboratory facility.
FIG. 11. Hylife converter concept.

FIG. 12. Liquid lithium 'waterfall' fuel cycle.
### TABLE II. THE SOCIAL COSTS (HEALTH, ENVIRONMENT AND SAFETY) OF GENERATING 1 GW(e) OF ELECTRICITY

<table>
<thead>
<tr>
<th></th>
<th>Coal</th>
<th>LWR</th>
<th>Breeder reactor</th>
<th>Solar</th>
<th>Magnetic fusion</th>
<th>Laser fusion</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Land use (acres)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Power plant</td>
<td>300</td>
<td>100 - 200</td>
<td>100 - 200</td>
<td>6000 - 19000</td>
<td>100 - 200</td>
<td>100 - 200</td>
</tr>
<tr>
<td>Fuel cycle (30 years life)</td>
<td>3000 - 12000</td>
<td>600 - 12000</td>
<td>15</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td><strong>Transportation (tonne/a) requirements</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3 x 10^6</td>
<td>200</td>
<td>23</td>
<td>Unknown</td>
<td>100 - 400</td>
<td>10</td>
</tr>
<tr>
<td><strong>Gaseous effluents</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nonradioactive (tonne/a)</td>
<td>40000</td>
<td>21</td>
<td>5</td>
<td>Unknown</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Radioactive (Ci/a)</td>
<td>0</td>
<td>15700</td>
<td>2000</td>
<td>0</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>Tritium</td>
<td>?</td>
<td>350000</td>
<td>150000</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td><strong>Solid waste</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Negligible</td>
<td>Negligible</td>
</tr>
<tr>
<td>Nonradioactive (tonne/a)</td>
<td>1 x 10^6</td>
<td>91000</td>
<td>9100</td>
<td>Unknown</td>
<td>450</td>
<td>30</td>
</tr>
<tr>
<td>Radiactive (m^3/a)</td>
<td>0</td>
<td>340 - 1100</td>
<td>450 - 1200</td>
<td>0</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Safety</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Accident consequences</td>
<td>Low</td>
<td>Very high</td>
<td>Very high</td>
<td>Very low</td>
<td>Low</td>
<td>Low</td>
</tr>
<tr>
<td>Accident probability</td>
<td>High</td>
<td>Very low</td>
<td>Very low</td>
<td>Very low</td>
<td>Very low</td>
<td>Very low</td>
</tr>
<tr>
<td>Premature deaths/a</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Occupational</td>
<td>0 - 8</td>
<td>0.1 - 0.9</td>
<td>?</td>
<td>?</td>
<td>0.004</td>
<td>0.003</td>
</tr>
<tr>
<td>Public</td>
<td>2 - 111</td>
<td>0.01 - 0.2</td>
<td>?</td>
<td>?</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>Total</td>
<td>2 - 120</td>
<td>0.1 - 1</td>
<td>?</td>
<td>?</td>
<td>0.007</td>
<td></td>
</tr>
<tr>
<td>Proliferation hazard</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>

Note: The values represent estimated costs or impacts, with ranges for some sources of uncertainty.
shot basis. Other experiments on Nova will include studies of chamber materials, activation and radiation effects, first wall experiments, and focusing optics survivability tests.

From the success with inertial fusion experiments and calculations so far, we can begin to visualize the laser fusion power plant of the future. In one concept in particular, the reactor chamber uses a thick liquid lithium 'wall' operating at 500°C (Fig.11). An array of 548 liquid lithium jets, each 20 cm in diameter, creates an effective thickness of 1 m of lithium, reducing the neutron fluence at the first structural steel wall to roughly one twentieth of that without the lithium. The lithium in the High Yield Lithium Injection Fusion Energy (HYLIFE) design reduces long-lived radioactive waste by a factor of ten or more, and eliminates the need for replacement of the first wall and reactor structure within the thirty-year lifetime of the power plant. The lithium fall also provides an environment for tritium breeding, and breeding ratios (atoms produced per fusion event) as high as 1.7 are calculated for this design. That is, for every three atoms of deuterium and tritium, one is burned, two are unburned and recovered, and 1.7 atoms are bred, processed and recycled. An important feature of the liquid lithium injection design is that the breeding ratio can be adjusted, providing flexibility for the use of tritium either at the source reactor or at other potential user sites.

Figure 12 shows a schematic of a possible tritium fuel cycle from the initial fabrication of individual pellets to the breeding, separation and recycling of deuterium-tritium fuel.

To extract tritium from the liquid lithium, two methods are being studied: diffusion through a refractory material and molten salt extraction cells. In addition, tritium must also be extracted from gases circulating in the reactor chamber, including helium that is being produced at the same rate as tritium in the breeding process. Also, inert cover gas and double-wall containment gas will need to be processed to remove tritium. Once again, several methods exist for tritium extraction: circulation through an aluminium-zirconium getter, preferential diffusion of the carrier gas through a ceramic, and molecular sieving of the tritium after conversion to tritiated water. In a nominal 1000 MW(e) laser fusion power plant of the HYLIFE type, approximately 64 grams of tritium are processed each hour (1.5 kg/day). A review of the inventory of tritium in the HYLIFE power plant shows that approximately 5 kg are held and continuously recycled in the liquid lithium loops. Approximately 1.25 kg of tritium are required each day to fuel a day’s supply of reactor pellets. This figure also determines the daily throughput of tritium found within the tritium reprocessing system itself, say 1.5 kg, since the amount of tritium dissolved in the extraction system is negligible. Finally, assuming a 1.7 breeding ratio and a ten-day supply of tritium bottled for feed to pellet fabrication, roughly 12.5 kg of tritium would also be stored in inventory to allow for reasonable down times. Thus, of a total inventory
of 19 kg of tritium, only a relatively small quantity, 1.5 kg, is found in the 'active' portion of the fuel cycle. The major portion of the total tritium is either bottled for future targets or captured in the lithium flow within the primary reactor cooling system.\(^2\)

Tritium handling, control and storage are perhaps best considered in the light of reducing the overall social cost burden presented to society by large-scale electric power production. Table II lists estimates of social costs for various present and future energy options. The coal and LWR columns give data derived from actual operation of these systems, whereas the remaining four columns present estimates for advanced sources of power. Fission breeders, solar electricity plants and the two types of fusion are commonly referred to as 'inexhaustible sources' because their supply of fuel is either somewhat regenerative or so abundant as to last for the foreseeable future of man on earth.

It is clear that any source of relatively inexhaustible energy is likely to be more expensive than present electrical generation sources: for example, the cost of a laser fusion power plant or a large breeder reactor of the 1000 MW(e) class is expected to be roughly 2 billion (2 thousand million) 1978 dollars. However, the ultimate selection of one or more of these alternatives may well depend on the social cost factors inherent in the design as much as in the capital investment required.

The typical 1000 MW(e) light-water fission reactor operating today discharges over 15 kCi of tritium as a gaseous effluent each year. In contrast, conceptual designs for fusion power plants show that their effluent can be limited to 1 kCi yearly by applying present-day tritium technology. In perspective, since the present (October 1978) installed capacity of nuclear power plants in the United States of America is 51.421 MW(e), a total of 807 fusion reactors of 1 GW(e) each could be operated with the same total curies of tritium effluent. This generating capacity is 1.5 times as great as the entire United States installed power today. It appears that tritium control, although an important factor to be considered in the design of future laser fusion systems, is a manageable characteristic. When coupled with the other social cost advantages indicated, including the low discharge of radioactive and inert wastes of all kinds, laser fusion has great potential as a safe and environmentally acceptable source of long-range energy.

\(^{2}\) In contrast, the total DT inventory required for the LLL laser fusion experiments is less than 0.5 gram (4800 Ci).
DISCUSSION

S. PORTER: In the comparison you made in your presentation of 1000 Ci/a of $^3$H release/GW(e) for a laser fusion reactor as against 15700 Ci/a for a light-water reactor, are you assuming for the laser reactor the same methods of off-gas stream processing as for LWRs?

P.E. COYLE: No. Light-water reactors involve different physical processes and different systems than those postulated for laser fusion reactors.

S. PORTER: Then you are comparing completely different situations?

P.E. COYLE: Yes, in the sense that the sources and their intensities are very different. Undoubtedly light-water reactor release rates can be improved also.

S. PORTER: Can you give me an estimate, for a fusion reactor versus a LWR, of $^3$H releases with comparable $^3$H purification systems?

P.E. COYLE: The figures quoted are for the state of the technology as it is being implemented today in light-water reactors and for what we believe we could achieve, given today's technology, in future laser fusion systems.
MEASUREMENT OF TRITIUM

Session 3, Part 1
HT-HTO SAMPLING AT
THE NEVADA TEST SITE

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Nevada Test Site,
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Abstract

HT-HTO SAMPLING AT THE NEVADA TEST SITE.
A method is proposed for field collection of gaseous tritium and tritiated water vapour
with a unit suitable for operation in remote desert areas. Its primary feature is its capability
of operating without supplementary hydrogen. The development of the sampling programme
and testing of the sampler adopted by Reynolds Electrical and Engineering Co., Inc. (REECo)
at the Nevada Test Site is discussed.

BACKGROUND

The Nevada Test Site, where the United States Department of Energy
tests nuclear devices, is located 105 kilometres northwest of Las Vegas,
Nevada, and covers an area of 350,000 hectares, much of it at the northern
fringe of the Mojave Desert. Most of the personnel employed there
are concentrated in three main camps leaving large areas of the site
relatively unoccupied. An extensive network of water and air sampling
stations is maintained there as part of a longstanding environmental
surveillance program, and recently it was decided that the determination
of tritium concentrations, in gaseous and vapor forms, in certain parts
of the site, would be worthwhile. This report will describe how the
sampler was developed.

The site is subdivided into numbered areas, some of which are ex-
tensively cratered. Although none of these craters are releasing sig-
nificant amounts of radionuclides, it is known from earlier REECO
studies that one of them is releasing measurable amounts of tritiated
water vapor. This is Sedan crater, the most prominent one on site.

This crater was formed by the detonation of a 100-kiloton thermo-
nuclear device in 1962 as a cratering experiment of the Plowshare series
of peaceful nuclear explosions. Soil and vegetation studies have shown
tritiated water vapor to be prominent in the area disturbed by the tre-
mendous throwout of the blast, making the area around the crater attrac-
tive for airborne tritium studies. Investigations there have proven to
be interesting but not surprising. It has been determined that tritium
gas in the area is of no consequence, but tritiated water vapor in con-
centrations as high as 9 X 10^-6 micrometers per cubic metre has been
found in certain areas near the rim of the crater. At a distance of 275 metres from the rim the concentrations typically fall to one-tenth to one-hundredth of the rim values.

**DESIGN**

The unit was designed according to the classical methods of sampling an air stream for gaseous tritium, viz. using the platinum-catalyzed reaction between oxygen and supplemental hydrogen to form collectible water vapor [1, 2]. It was soon discovered, however, that any type of
TABLE I. COMPARISON OF COLLECTION METHODS

<table>
<thead>
<tr>
<th>Sampling period</th>
<th>Tritiated water vapor ($\mu$Ci/m$^3$)</th>
<th>Tritium ($\mu$Ci/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$5.7 \times 10^{-5}$/6.0 X $10^{-5}$</td>
<td>$2.0 \times 10^{-4}$/1.3 X $10^{-4}$</td>
</tr>
<tr>
<td>2</td>
<td>$5.4 \times 10^{-5}$/5.7 X $10^{-5}$</td>
<td>$1.4 \times 10^{-4}$/1.7 X $10^{-4}$</td>
</tr>
<tr>
<td>3</td>
<td>$5.3 \times 10^{-5}$/5.4 X $10^{-5}$</td>
<td>$1.3 \times 10^{-4}$/1.5 X $10^{-4}$</td>
</tr>
<tr>
<td>4</td>
<td>$4.5 \times 10^{-5}$/4.4 X $10^{-5}$</td>
<td>$5.7 \times 10^{-5}$/6.4 X $10^{-5}$</td>
</tr>
<tr>
<td>5</td>
<td>$1.1 \times 10^{-3}$/9.5 X $10^{-4}$</td>
<td>$3.6 \times 10^{-3}$/2.8 X $10^{-3}$</td>
</tr>
<tr>
<td>6</td>
<td>$2.4 \times 10^{-4}$/2.0 X $10^{-4}$</td>
<td>$1.7 \times 10^{-4}$/1.2 X $10^{-4}$</td>
</tr>
<tr>
<td>7</td>
<td>$1.6 \times 10^{-4}$/1.7 X $10^{-4}$</td>
<td>$1.9 \times 10^{-4}$/2.0 X $10^{-4}$</td>
</tr>
<tr>
<td>Ratio average ± Variance</td>
<td>1.0 ± 0.01</td>
<td>1.1 ± 0.09</td>
</tr>
</tbody>
</table>

Supplemental hydrogen delivery system could cause problems in an exposed field unit such as this one. An alternative was devised, tested and found to be an adequate substitute. This will be discussed later.

The sampler consists of a rotameter, two standard Plexiglas drying columns containing solid desiccant with a 47-millimetre Millipore aerosol sampler holder between them, and a small electronic pump. Connections between individual components are made with clear plastic tubing, Figure 1. The metal chamber contains six grammes of five percent platinum on asbestos as a site for the hydrogen-oxygen reaction. No provisions were made for heating the catalyst. All components with the exception of the hydrogen delivery system (when used) are housed in a plywood box measuring 30 centimetres wide X 35 centimetres high X 15 centimetres deep. The box is equipped with a handle, and all surfaces painted with epoxy resin for weather protection. The complete unit, not including the power source, weighs approximately six kilogrammes and can be built for less than $400.

The pumps marketed by Spectrex, Inc., of Redwood City, California, are very practical for such a unit. One model can be powered by a 6-volt battery and draws 0.25 ampere at its maximum flow rate of one litre per minute. A flow rate of 0.5 litre per minute, chosen as the standard sampling rate, is not sufficient to drive any of the total flow meters available so the total volume of air sampled during a given period is determined by calculation. It is known that the flow rate does not fluctuate appreciably over a one-week period, so the volume of air drawn through the system can be determined easily to within five percent.

Silica gel and Drierite are both satisfactory for collection of moisture in this system. It could be modified to work with some liquid dehydrating agent such as ethylene glycol [3], but silica gel is most often used because of availability. It has been found, however, that water vapor will migrate through a bed of this material if dry air is passed through a previously hydrated bed. This is exactly the situation
in the second column, the sampled air stream having been thoroughly dried by the first column; as expected this condition is aggravated by summer temperatures in the desert, and up to 80 percent of the moisture in this column can be lost during the sampling period. Unless it is recognized and proper allowances made, this will of course cause errors in the calculation of tritium concentrations.

A suitable technique was found which permitted the extra hydrogen supply system to be eliminated. It was hypothesized that the simple addition of five millilitres of distilled water to some absorbent material at the inlet of the second drying column might be as effective in aiding the collection of water vapor generated in the metal chamber as would a small amount of hydrogen gas metered into the sampled air stream. By arranging two identical units in parallel, one using the addition of water and the other a supplemental hydrogen delivery system at one cubic centimetre per minute, it was shown that both units yielded similar results. Table I shows the actual results of this investigation at one permanent sampling location.

The samplers, each with a storage battery and a solar panel, are deployed in the field and attended there. The drying columns are changed once a week with preweighed columns, the expended ones taken to the laboratory, reweighed, and the absorbed moisture driven off. Some of this is collected and measured by liquid scintillation counting. The detection limit is 5 X 10^{-8} microcuries per cubic metre at the standard flow rate over a one-week period. After the desiccant has been dehydrated, it is regenerated by heating in a vented oven at 200 degrees Celsius for two hours and reused three or four times with no apparent memory effects.

CONCLUSION

The sampler as designed is useful for the collection of tritium and tritiated water vapor in air. Its portability permits transportation to practically any location, and it is reliable. Studies with stable hydrogen gas have shown it to convert hydrogen to water vapor with satisfactory efficiency, and overall it is believed to be practical and economical in those cases where time-averaged concentrations in the field are to be determined.

REFERENCES

DISCUSSION

R.V. OSBORNE: What is the lifetime of the pumps under your continuous sampling conditions?

R.J. STRAIGHT: These pumps were originally guaranteed for 10,000 hours but I am not sure if this is still the case. At all events our experience has shown them to be operational for only six to nine months in some cases and for almost two years in others. The primary problem with these pumps seems to be the small wire that is attached to the voice coil of the small speaker which actually does the pumping. This wire has a tendency to break at its point of attachment to the coil but can be resoldered, albeit with some difficulty.

A.S. MASON: Do the HT data show a background value of approximately 3 pCi per standard m³ of air?

R.J. STRAIGHT: This is approximately our background level. However, I intend to check the raw data to confirm this point.
A LOW-BACKGROUND LIQUID SCINTILLATION COUNTER FOR MEASURING LOW-LEVEL TRITIUM

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Abstract

A LOW-BACKGROUND LIQUID SCINTILLATION COUNTER FOR MEASURING LOW-LEVEL TRITIUM.

A low-background liquid scintillation counter with an automatic sample changer for measurement of low-level tritium has been developed. The anti-coincidence guard detector consists of a plastic scintillator and two bi-alkali photomultiplier tubes. The plastic scintillator is in the shape of a bottomless box and envelopes the sample chamber which contains a 100 ml Teflon counting vial. To eliminate entirely the slight leakage effect of an external standard source, a removable coin-type source was devised. The source was only utilized when efficiency calibration of the samples was carried out. The automatic changer has a capacity of 15 samples and cyclic repeat counting is easily performed. The background count rate of the instrument is less than 5 counts/min using a 100 ml Instage/water (60/40) cocktail. The sensitivity corresponds to 1 count/min per 25 TR (Tritium Ratio) and is certainly adequate for monitoring programmes on the present northern hemisphere fresh water without any prior enrichment.

1. INTRODUCTION

Measurement of low levels of tritium is becoming important in studies dealing with hydrology, meteorology and monitoring programmes in the field of health physics for tritium releases from nuclear power facilities.

For the detection of soft beta-emitting radionuclides such as tritium or carbon-14, the liquid scintillation method offers potentially a greater sensitivity than the gas-filled counting technique because of the capability of the detector to accept a large sample size derived from the high density of the liquid phase. The large sample size, however, will be associated with a somewhat high background. For low-level counting systems a 'figure of merit' value which is the ratio
of the net sample count rate to the square root of the background \( S/\sqrt{B} \) has been postulated. It is, therefore, well recognized that the background must be minimized to increase the 'figure of merit'.

Background reduction in the gas counter has been effectively achieved by the use of a ring of GM or coaxially enveloping counters surrounding the sample counter because the geometrical volume of the sample is inevitably large. In the liquid scintillation counter, however, the physical sample volume is relatively small, and several previous works \[1, 2\] showed that the effect of anti-coincidence counters on background reduction was less important than was first thought. Recently Noakes et al. \[3\] challenged this theory and developed a low-background liquid scintillation counter which utilized an anti-coincidence guard consisting of a large annular NaI crystal and four photomultipliers. The instrument gave an excellent performance with a low background as low as two counts/min and high efficiency; its disadvantages, however, are lack of sample-changing mechanism and utilization of external standard source. The system has an ideal geometrical arrangement between the sample and the NaI scintillator, and the detector assembly of the system and some of the heavy shielding blocks must be taken apart when a sample is mounted or exchanged. The external standard method for quenching correction is not easily available. Besides these difficulties, the expense of constructing the detector is considerably increased.

The general aim of this work is to develop a counting system with a low background characteristic allowing automatic sample change and external standard channel ratio calibration. This system, therefore, has the great advantage of routine utilization in the measurement of low-level tritium such as monitoring programmes for environmental water. It also means that the system is easily operable by workers inexperienced in operating sophisticated low-level counting instruments.

2. DESCRIPTION OF THE COUNTERS

2.1. A prototype counter

A counter was designed for normal 20 ml vials to obtain fundamental performance data of an anti-coincidence system.

The sample counting detector used the conventional coincidence technique which places a sample chamber between two photomultiplier tubes arranged coaxially. EMI 9635 QB quartz-face tubes were selected for their excellent low dark current characteristics. The sample chamber was made of oxygen-free copper (OFC) providing low radioactivity contamination. The detector described above was surrounded by 10-cm thick iron which was obtained from armoured material from the battleship "Mutsu", a ship sunk by an explosion in World War II which
FIG. 1. Dimensional layout of detector of prototype counter (dimensions in mm).

TABLE 1. EFFECTS OF THE EXTERNAL STANDARD (NON-OPERATIVE POSITION) ON THE BACKGROUND COUNT RATE (counts/min)\(^a\)

<table>
<thead>
<tr>
<th></th>
<th>Vial with black ink</th>
<th>No vial</th>
<th>Empty vial</th>
<th>Vial with scintillator</th>
</tr>
</thead>
<tbody>
<tr>
<td>A (equipped)</td>
<td>3.10</td>
<td>8.40</td>
<td>12.83</td>
<td>20.53</td>
</tr>
<tr>
<td>B (removed)</td>
<td>2.87</td>
<td>6.20</td>
<td>12.10</td>
<td>16.37</td>
</tr>
<tr>
<td>A − B</td>
<td>0.23</td>
<td>2.20</td>
<td>0.73</td>
<td>4.16</td>
</tr>
</tbody>
</table>

\(^a\) Aloka LSC 601B, 20 ml vial, \(^3\)H window.

FIG. 2. Block diagram of electronic circuitry of the prototype counter.
was salvaged after the war. This material is absolutely free from atomic bomb fall-out contamination. The anti-coincidence guard detector consisted of a plastic scintillator (C.I. Kogyo, Tokyo), 24 cm square and 7 cm thick, and two RCA 8575 photomultiplier tubes with bi-alkali photocathodes selected for their high quantum efficiency. This guard detector was set directly above the iron shield for simplicity of fabrication and sample mounting. An additional lead shield of 5 cm thickness was mounted above the guard detector. The dimensional layout of the detectors and the massive shield is shown in Fig.1.

Sample transfer into the counting chamber is achieved by the following four mechanical steps:

1. The turntable rotates on which the samples are circularly arranged.
2. The elevator is lowered.
3. The rotating conveyor moves through an angle of 180°.
4. The elevator moves upward into the counting chamber.

An external standard source which is conventionally utilized for quenching correction has become an indispensable device in liquid scintillation counting. The preliminary experiment, however, revealed that the radioactivity source created an increment in the background count of several counts per minute even in the non-operating position. The result obtained from a commercially available counter is listed in Table I as an example. To eliminate this leakage effect completely, a removable rod source was designed. A plastic sealed radioactive source tip containing 10 mCi of $^{137}$Cs was screwed into one end of a 30-cm steel rod. An oblique hole whose length was 27 cm, penetrating the iron shield, was drilled. A dummy steel stick the same size as the source rod was inserted into the hole in place of the source rod as the shielding material during the sample counting run. The electronic circuitry following the detector is, as shown in Fig.2, the conventional one.

In recent years the suspension of water in aromatic solvent using a non-ionic surfactant (emulsion systems) has become popular. These systems are capable of incorporating up to 40% water and this technique is widely used for low-level counting of tritium. Pre-mixed aromatic solvent and surfactant dissolving scintillation flours are now commercially available. Using these mixtures, the preparation of the water sample becomes quite rapid and simple. The systems Instagel or Aquasol-2 (60%) and water (40%) were used in this work.

The background count rate in the optimum $^3$H window of the prototype counter is 2.8 counts/min on the average using the emulsion system of Instagel : water (12ml : 8ml) with low potassium glass vials. The water used as blank is underground water obtained from a deep well. The corresponding $^3$H counting efficiency is 15%. The performance of the counter is expressed concisely by Moghissi's Y-value [4] which is the minimum limit of detection at a 68.3% confidence level and one minute counting time. The value correlates background B
(counts/min), efficiency E (counts/min per disintegrations/min), and the amount of water in the scintillation systems M (g or ml):

\[ Y \text{ (pCi/ml)} = \sqrt{\frac{B}{2.22 \times E \times M}} \]

Substituting \( B = 2.8 \), \( E = 0.15 \) and \( M = 8 \) in the above equation gives a Y-value of the prototype counter that is 0.63 pCi/ml. Another expression of the sensitivity for low-level counting of tritium is the value of the counts/min equivalent Tritium Ratio:

\[ S \text{ (TR/cpm)} = \frac{1}{(7.2 \times 10^{-3} \times E \times M)} \]

where \( 7.2 \times 10^{-3} \) is disintegrations/min per ml water of 1 TR. The S-value of the counter is 116 TR/cpm.

2.2. A modified counter for large samples

In the measurement of the low-activity sample which gives a count rate near to that of background, the minimum detectable activity is inversely proportional to the figure of merit (fm) expressed as

\[ fm = E \times M / \sqrt{B} \]

where E is the counting efficiency, M is the amount or volume of the sample and B is the background count rate. To achieve a high value of fm, a large volume of the sample should be incorporated in the detector. In conventional counting systems, however, the increase in the sample size also causes an increase in the background count rate. As an anti-coincidence guard system in general works more
effectively on a larger sample size, a modified detector was designed which was able to hold a vial as large as 100 ml capacity and to change the sample automatically.

The plastic scintillator of an anti-coincidence guard detector in this system was a rectangular solid whose dimensions were 20 cm square and 22 cm high with a cylindrical cavity of 10.5 cm diameter and height 13.5 cm. The scintillator enveloped the sample chamber which was able to hold a 100-ml vial so as to improve the geometrical detection efficiency for external cosmic ray radiation. Two photomultiplier tubes (RCA 8575) were attached to the opposite side of the surface of the scintillator. A pair of RCA 4501-V3 photomultiplier tubes for the sample detector penetrated the two side walls of the plastic scintillator and faced each other. A dimensional layout of the detector is illustrated in Fig.3.

Such an external standard source as designed for the prototype counter cannot be used in this case because of the impossibility of drilling a hole through the anti-coincidence guard scintillator. A coin-type sealed source was devised to be set by means of a simple magnetic tool in a small circular pit on the pedestal of the sample holder which was transferred into the counting chamber with a sample vial.

Teflon vials of 100 ml capacity, 48 mm diameter (O.D.) and 75 mm height (from the bottom to the shoulder), were selected because of their low radioactivity content, low light absorption and excellent chemical tolerance. Although the vials are commercially available, they are not especially manufactured for liquid scintillation counting and therefore lack uniformity in optical density because of variation in wall thickness. The vial was selected by the external standard channel ratio which approximately correlated with the degree of photon absorption of the vial wall.

Transfer mechanisms of the counting sample are quite similar to those of the prototype counter apart from the number of the sample and the diameter size of the sample holder.

The background count rate in the optimum $^3$H window of the counter is 4.4 counts/min on the average using the emulsion system of Instagel or Aquasol-2 : water (60ml : 40ml) with Teflon vials. Although the background count varies from one lot of vials to another, the variation of the background of the vials in an identical lot shows a random statistical nature.

The tritium counting efficiency in the optimum window is 14% and therefore the Y-value is

$$Y (\text{pCi/ml}) = \sqrt{4.4/(2.22 \times 0.14 \times 40)}$$

$$= 0.17$$

and the S-value is

$$S (\text{TR/cpm}) = 1/(7.2 \times 10^{-3} \times 0.14 \times 40)$$

$$= 24.8$$
The results show that the sensitivity for the measurement of low-level tritium of the modified counter is 4.7 times higher than that of the prototype counter.

The long-term stability of background counting is shown in Fig.4. A slight seasonal variation probably caused by the change of temperature is observed.

3. DISCUSSION AND CONCLUSION

One of the most important aspects of counters is the low counting efficiency. In the prototype counter, the inferiority in the counting efficiency is mainly caused by the low quantum efficiency of the photocathodes of the photomultiplier tubes. Selection of low dark current tubes unfortunately also involves selection of low efficiency tubes. The counting efficiency of the unquenched tritium standard is 45% at best.

No positive evidence of the counting efficiency of the modified counter is available. Possible causes of the drawback are an increase in the distance between the photo-surfaces of the tubes and inferior geometry of the vial for the photocathodes.

The modified counter, designated LB-1, has been continuously operated for five years for measuring tritium in environmental water ranging between 20 and 900 pCi/litre.

A comparison of overall error estimation between the direct counting method and the pre-enrichment method is shown in Fig.5. Horizontal solid lines indicate
a 2σ confidence level of counting statistics only for direct counting. Vertical lines, on the contrary, indicate the twice of the root sum square of the standard error of counting statistics and the error associated with electrolytic enrichment. The enrichment factor, which is the ratio of final tritium concentration to initial tritium concentration, is about 30 and the associated error is estimated to be 5% experimentally. For concentrations higher than 200 pCi/litre, the accuracy of both methods appears to be about the same. For a lower concentration, the pre-enrichment method is apparently more accurate. The direct counting method, however, can be applicable to a sample as low as 50 pCi/litre with reasonable accuracy. The sensitivity is sufficient to analyse recent fresh water samples from the northern hemisphere without any enrichment process which usually consumes a great deal of time and labour.

N.B. A new model of LB-1 modified for a 20-ml vial has been built just before this paper was submitted. Preliminary results indicate a background count rate of 0.96 counts/min with a 33% window by the use of a toluene scintillator and polyethylene vials. Detailed results will be reported in the near future.
REFERENCES


DISCUSSION

G. MASTINU (Chairman): The 100 ml Teflon vials you employed are commercially available and are not specifically intended for liquid scintillation counting. Might not the variability of their physical characteristics have affected the reproducibility of low-activity samples and background fluctuations?

I. IWAKURA: As we mention in the paper, although the background count varies from one lot of vials to another, the variation in the background of vials taken from a given lot exhibits a random statistical character. Therefore, if a lot number is identified, the background fluctuation caused by the physical condition of the vials need not be taken into account. On the other hand, counting efficiency is always slightly affected by physical condition and a shift occurs. This shift, however, can be compensated by external standard channel ratio calibration.

G. MASTINU (Chairman): The counter for large samples you have described is of great assistance to researchers engaged in tritium measurements for environmental and health protection purposes. Actual environmental levels are in fact within the range of tens of hundreds of pCi/litre. Commercially available apparatus, on the other hand, permits significant measurements of tritium only down to 200–100 µCi/litre. Your modified counter may therefore permit significant measurements of tritium at environmental levels, avoiding the need for electrolytic enrichment of samples. This is a very delicate and time-consuming technique, although, incidentally, its application seems unavoidable in geological studies, which often involve samples with a very low tritium level (0.1–10 T.U.).
A RAPID AND QUANTITATIVE METHOD TO DETERMINE THE TRITIUM CONTENT IN DNA FROM SMALL TISSUE SAMPLES

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Abstract

A RAPID AND QUANTITATIVE METHOD TO DETERMINE THE TRITIUM CONTENT IN DNA FROM SMALL TISSUE SAMPLES.

A rapid and quantitative two-step procedure to isolate double-strand DNA from small (10 – 100 mg) animal tissue samples is presented. The method is developed for investigations to evaluate the relative importance of organically bound tritium for the dose factors used to calculate dose commitments due to this nuclide. In the first step the proteins in the homogenized sample are hydrolysed, at a high pH (9.0) and ionic strength (1.5) to dissociate protein from DNA, using immobilized Proteinase K as a proteolytic enzyme. The DNA is then absorbed to hydroxylapatite and separated from impurities by step-wise elution with buffers of increasing ionic strength. More than 90% of the DNA in the samples could be isolated in double-strand form by this procedure. The method has been applied to determine pool-sizes and biological half-life times of tritium in DNA from various animal (mouse) tissues. It has also been shown to be suitable in other radiobiological studies where effects on DNA are investigated.

INTRODUCTION

Tritium in tritiated water may be directly bound to carbon in organic molecules through various metabolic pathways [1]. In these positions it is hardly exchanged with hydrogen in water molecules. Thus when an organism is kept in tritiated water the specific activity of metabolically incorporated tritium (mainly bound directly to carbon) will at steady state approach the
### TABLE I. THE RELATIVE SPECIFIC ACTIVITY (RATIO OF T/H IN ORGANIC COMPOUNDS TO T/H IN BODY WATER) IN ANIMAL TISSUES AFTER HTO INGESTION OR EXPOSURE TO A TRITIATED ENVIRONMENT

<table>
<thead>
<tr>
<th>Compound Exposure</th>
<th>T in organic pool</th>
<th>Lipids</th>
<th>Protein</th>
<th>DNA</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>After THO exposure</td>
<td>1.8</td>
<td>0.3 - 0.5</td>
<td>0.7 - 1.1</td>
<td>0.12</td>
<td>[2]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.5</td>
<td></td>
<td></td>
<td>[3]</td>
</tr>
<tr>
<td>After T exposure through food chains</td>
<td>1.6</td>
<td>1.1 - 1.5</td>
<td>1 - 4&lt;sup&gt;b&lt;/sup&gt;</td>
<td></td>
<td>[5]</td>
</tr>
<tr>
<td>from THO</td>
<td></td>
<td>0.9&lt;sup&gt;c&lt;/sup&gt;</td>
<td></td>
<td></td>
<td>[6]</td>
</tr>
</tbody>
</table>

*Weight fraction of T in organic compound relative to T in water at statistical T-distribution*

|            | ~ 0.7 | 1.09 | 0.64 | 0.32 |


<sup>b</sup> Nucleoprotein.

<sup>c</sup> Steady state value not obtained.

Specific activity of tritium in the tritiated water. This applies only for those C-H bonds that can participate in the metabolism of the organism and in the absence of isotope effects. Recently experimental evidence for this behaviour has been presented [2]. Mice were given tritiated water for two years. The specific activity of tritium in the nucleic acids was then approximately equal to the corresponding activity in the body water. The relative activity of tritium in a protein, haemoglobin, was however lower as the C-H bonds in the essential amino acids do not participate in the metabolism.

The organically bound tritium can either be incorporated in the organic pool in higher trophic levels or be involved in the energy metabolism yielding tritiated water. Thus an organism kept in a generally tritiated surrounding where tritium initially is present as HTO has two sources for organically bound tritium:

1. an inorganic source (HTO);
2. an organic source from lower trophic levels.
FIG.1. The ratio $A_{260}/A_{230}$ for DNA-basic protein mixtures.

- Chymotrypsinogen;
- Lysozyme

When these sources can be considered to be independent of each other in the metabolism, a higher specific activity of tritium in the organic pools is expected than when only the inorganic source is present. There are, however, only a few studies available where the incorporation of tritium from both sources has been studied [3–6] (Table I). The data reported there indicate that the tritium content in the organic pools increases along a food chain starting from HTO.

The critical target for tritium decay in cells is the nucleus, and here one of the most sensitive targets is DNA. The nucleus of mammalian cells may contain up to 10% (wt/wt) of DNA [7, 8]. Only the dose from tritium in the water is accounted for in the procedures recommended by the ICRP to calculate the dose from tritium released to the environment [9]. The figures given above, the data in Table I and results that show that RBE for tritium in DNA is 1.5 times the RBE for tritium in the water compartment, indicate that these procedures to calculate the dose may underestimate the real dose from tritium [10]. As tritium, as well as $^{14}$C, $^{88}$K and $^{129}$I, is a nuclide that gives a major contribution to the population doses caused by the nuclear fuel cycle, a re-evaluation of the dose factors used to calculate the dose from tritium seems justified.

For this re-evaluation the relative specific activities and pool sizes of tritium in the different organic pools (compartments) and their biological half-life times must be determined. The activities and pool sizes must be determined after a steady state has been reached in the different trophic levels exposed to tritiated water. Published data that are relevant here mainly consider the inorganic tritium source and cover only one trophic level (Fig.1). They are therefore only of relevance to estimate the dose in the first trophic level exposed to tritiated water. Further studies that consider both inorganic and organic tritium sources are therefore necessary. DNA is the organic pool with the longest biological half-life time, and an important target for the tritium decay. The relative importance of the dose from tritium in DNA must therefore be determined.
To determine pool sizes, specific tritium activities and biological half-life times of tritium in DNA, a method that allows a quantitative isolation of DNA from a tissue sample must be used. The available methods are rather time consuming and consist of many extraction steps that make quantitative isolation of DNA from small tissue samples difficult. We have therefore developed a rapid two-step procedure for quantitative DNA isolation that is suitable for these purposes [11]. The method and some of its applications will be presented.

MATERIALS AND METHODS

Materials

DNA (Sigma, D-1501), bovine serum albumin (Serva 11920), RNA (Boehringer 15363), chymotrypsinogen (Worthington CGC), lysozyme (Worthington LYSF), pronase (Merck), proteinase K immobilized to cellulose (Merck), and H-3 TdR (Amersham TRK-120) were used as purchased. All other chemicals were of reagent grade.

Animals and cells

Mice (NMRI) were obtained from Eklund's Animal Farm, Täby, Sweden.

Tissue homogenization

The mice were sacrificed by cervical dislocation. Immediately thereafter tissues were prepared and frozen on dry ice. Samples of tissues (10–100 mg) were then sonicated in 2 ml homogenization buffer (0.15M Na-phosphate, 0.024M EDTA, pH = 7.9) at 4°C for 3 min at 50 W (Branson sonifier B 15).

Deproteinization of the homogenate

The homogenate was added to 3 ml of a suspension of proteinase K immobilized to cellulose (20 mg immobilized proteinase per 100 mg tissue (wet weight)) in the following buffer (0.15M Na-phosphate, 2M NaCl, pH = 9.0) and incubated overnight (16 h) at 40°C. The suspension was agitated throughout the incubation. The sample was then filtered to remove the immobilized proteinase K.

Isolation of DNA

The filtrate from the deproteinized sample was diluted to an ionic strength of 0.05 and shaken 10 min with ~ 0.5 g DNA-grade hydroxylapatite (Biorad). DNA and RNA are then adsorbed. RNA (and residual protein), and DNA were
desorbed by washing the hydroxylapatite with phosphate buffer (pH = 6.8) of ionic strength 0.15 and 0.50 respectively. A filter manifold (Millipore with EHWP filters) was used to treat up to 30 samples simultaneously. The filtrates were collected. The isolated DNA was analysed with respect to RNA [12] and protein content chemically [13] and spectroscopically, using the ratio of the absorbance at the absorption maximum to the absorbance at 230 nm. The double strandedness was estimated from the melting curve. A Gilford spectrophotometer (250) equipped with a thermoprocessor was used for the spectrophotometric measurements.

The tritium content in the DNA and other filtrates was determined by liquid scintillation counting.

RESULTS AND DISCUSSION

Yield and properties of the isolated DNA

The procedure to isolate pure DNA as described under materials and methods was found to give optimal DNA yields. In the deproteinization step in which chromosomal and non-chromosomal proteins are hydrolysed the use of immobilized proteases was found to be essential to prepare DNA free from protein. Large quantities of proteases can then be used. They do not hydrolyse each other as they are immobilized, and they can be re-used. The complex formation between DNA and basic proteases that is frequently observed [14] does not interfere here as the proteases are easily filtered off under conditions where the complex formation is suppressed (high pH and ionic strength). For some tissues (pancreas, intestine, spleen), protein-free DNA could be prepared when the deproteinization was carried out at a lower pH (7.0) using Pronase immobilized to spherical gel particles (Sepharose) [11] as a protease. For other tissues (brain, kidney, liver) this procedure did not yield protein-free DNA. In order to obtain protein-free DNA from these tissues it was found necessary to perform the deproteinization at a higher ionic strength and pH, where the chromosomal proteins are dissociated from DNA, and using proteinase K as a proteolytic enzyme. This demonstrates that there are tissue-specific differences in the interactions between DNA and the chromosomal proteins. Preparation of protein-free DNA from tissues with a high content of chromosomal proteins [7] requires the procedure with proteinase K as described here. This procedure was used throughout this study. This method allows a nearly quantitative DNA isolation (Fig.2). More than 90% of the $^3$H counts in tissues and cells that had incorporated $^3$H TdR was found in the DNA-fraction. The isolated DNA was mainly double stranded as it was desorbed from the hydroxylapatite at an ionic strength where double-stranded DNA is desorbed. This is also supported by the determination of the hyperchromicity (Table II). The molecular weight is, however, only $10^6 - 10^7$ daltons. Higher
FIG. 2  Distribution of tritium counts, after $^3$H TdR exposure, in various fractions obtained in the DNA isolation procedure. Single-strand DNA is desorbed by 0.15M phosphate buffer, double-strand DNA by the 0.5M buffer (HA = hydroxylapatite).

A – C  Tissues from mice that had been given $^3$H TdR. Shaded area gives the range of observed data, for 10 samples.

D  Isolated DNA with tritium was applied to a tissue homogenate from an animal that had not received $^3$H TdR.

molecular weight DNA can be prepared when the homogenization is carried out by grinding the tissue sample in liquid nitrogen [15]. This procedure is time consuming and not essential for the purposes for which the method described here was developed. These required a rapid and quantitative method to isolate double-stranded DNA from small tissue samples (10 – 100 mg). The method described here meets these requirements.

The amount of isolated DNA is directly determined from its UV spectrum as it is essentially free from protein and RNA [16]. This requires knowledge of the base composition. The nucleotide absorbancy and the amount of DNA were determined as described in Ref.[17].

The amount of protein in the isolated DNA can be determined from the ratio of the absorbance at the absorption maximum and at 230 nm (Fig.1). This can be used for direct determination of the radiation-induced crosslinking
### TABLE II. AMOUNT OF DNA ISOLATED FROM ANIMAL TISSUES AND ITS PROPERTIES\(^a\)

<table>
<thead>
<tr>
<th>Tissue</th>
<th>DNA-content (mg/g wet tissue)</th>
<th>(A_{260}/A_{230})</th>
<th>Protein content (%)</th>
<th>RNA content (%)</th>
<th>Hyperchromicity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>This study</td>
<td>Literature values [8]</td>
<td></td>
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</tr>
<tr>
<td>MOUSE (NMRI)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Small intestine</td>
<td>4.8</td>
<td>3 - 4</td>
<td>2.30</td>
<td>(&lt; 1)</td>
<td>(&lt; 1)</td>
</tr>
<tr>
<td>Liver</td>
<td>3.8</td>
<td>2.3 - 2.9</td>
<td>2.23</td>
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<td>(&lt; 1)</td>
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<tr>
<td>Pancreas</td>
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<td>3</td>
<td>2.28</td>
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<td>–</td>
<td>2.12</td>
<td>5</td>
<td>2</td>
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<tr>
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<td>–</td>
<td>2.27</td>
<td>1</td>
<td>1</td>
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<td>RAT (Wistar)</td>
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<td></td>
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<tr>
<td>Brain (whole)</td>
<td>4.5</td>
<td>1 - 3</td>
<td>2.15</td>
<td>2</td>
<td>2</td>
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<tr>
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<td></td>
<td>2.21</td>
<td>(&lt; 1)</td>
<td>(&lt; 1)</td>
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<tr>
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<td>2.16</td>
<td>5</td>
<td>3</td>
</tr>
<tr>
<td>Liver</td>
<td>3.9</td>
<td>2.4 - 3.5</td>
<td>2.20</td>
<td>(&lt; 1)</td>
<td>(&lt; 1)</td>
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<tr>
<td>Pancreas</td>
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<td>3.7 - 5</td>
<td>2.26</td>
<td>(&lt; 1)</td>
<td>(&lt; 1)</td>
</tr>
</tbody>
</table>

\(^a\) Reproducibility of triplicates (from the same organ) ± 3%.

of protein to DNA [18] as the deproteinization as carried out here does not hydrolyse protein that is bound to DNA.

The yield and properties of the DNA isolated from various tissues are given in Table II. The yields are consistently higher than those obtained using other methods to isolate DNA [8]. This also demonstrates the advantage of the DNA isolation method described here.

**Applications**

*Determination of the biological half-life times of tritium in DNA of various animal tissues*

The method described to isolate DNA has been applied in a preliminary study to determine the biological half-life time of tritium in DNA. Mice were
given $^3$H TdR subcutaneously and in their drinking water for a short time. The tritium content in the DNA from various tissues was then determined as a function of time after they received $^3$H TdR. Some results are given in Fig.3. After 10 days more than 90% of the tritium counts were found in the isolated DNA. The size of the metabolic $^3$H TdR pool is then negligible, and the data for longer times can be used to determine the biological half-life time of the tritium in DNA. The results indicate as expected that the half-life time is longer in tissues with a low proliferating capacity than in those proliferating rapidly [19]. The observed differences in the sizes of the tritium content in DNA show that there are also metabolic sex differences in the rates with which $^3$H TdR is incorporated. It is also readily incorporated in DNA in those tissues where the cells proliferate slowly.

A small (≈ 10%) but consistent decrease in the ratio $A_{\text{max}}/A_{260}$ of the isolated DNA was observed with time after $^3$H TdR incorporation. This indicates that the tritium decays induced a crosslinking between protein and DNA. More data and an investigation on the possible repair of this crosslinked DNA are necessary to substantiate this observation further.

FIG. 3. The specific tritium activity in DNA from mice tissues after a single intake of $^3$H-thymidine in mice. The mice (NMRI, age 30 days) each received 10 μCi $^3$H-TdR subcutaneously and 10 μCi through the drinking water at time zero. Tissues from 3 – 5 animals were used for each data point. $A_{260}$ = Absorption at 260 nm from DNA in the sample used in the scintillation counting of tritium.
Applications in cellular radiation biology and radiation therapy

The method to isolate DNA quantitatively has also been used to study the protection of healthy cells in radiotherapy with depression of capillary blood flow [20]. Because of the anoxic conditions the amount of DNA in the protected tissue (small intestine) was larger than in unprotected tissue. In the latter cell death and degradation of radiation-damaged DNA reduced the amount of DNA in the rapidly proliferating mucosal layer. Radiation-induced DNA protein crosslinking due to incorporated $^3$H and $^{14}$C and its repair were also studied.

Further studies

It has been shown that a tritium pool with a long biological half-life time — > 100 days — is tritium incorporated in DNA. On this basis a longer study on the relative importance of this tritium pool for the total dose commitment from tritium (and $^{14}$C) is in progress. This requires that both the half-life times and the pool sizes, at steady state, from inorganic and organic tritium sources in DNA must be determined. The possible increase in the T/H ratio along a food chain, as observed in some studies (Table I), can then be investigated under more controlled conditions. The DNA isolation method described here is a valuable tool in this context.

A re-evaluation of the dose factors for tritium (and $^{14}$C) [21] requires co-ordination of studies carried out in different laboratories. Procedures and basic data used in this work should be standardized as much as possible. It is also necessary to discuss whether the nucleus should be considered as a critical organ for the decay of incorporated tritium and $^{14}$C.

ACKNOWLEDGEMENTS

This study has been supported by the Zentrale Forschungskommission der Universität Bremen (V.K., R.Z.), and the Swedish Atomic Research Council (H.A., L.N.).

REFERENCES

Y. UENO: I am rather doubtful about the efficiency when you use tritiated water for labelling DNA, owing to the fact that the tritium is then loosely bound.

V. KASCHE: We did not use tritiated water here, in any case this would not influence the yield in the isolated DNA, although of course the activity would be much lower.

Y. UENO: The half-life in DNA is shorter in the small intestine, which has a fast cell cycle, than in the pancreas, but the specific activity is lower in the former than in the latter. Could this be a cause of a discrepancy in your results?

V. KASCHE: We were only interested in the longer biological half-life and did not determine the tritium content in DNA for the first ten days. The rapidly proliferating cell compartment in the small intestine probably has a higher specific tritium activity in its DNA than does the pancreas.
D.J. MEWISSEN: Have you any experience of using your method for assessing the incorporation of tritium into DNA following exposure to tritiated water?

V. KASCHE: No, not yet.

D.J. MEWISSEN: Is there any possibility that isolated DNA might be contaminated by tritiated protein in the final stage of your method?

V. KASCHE: The protein content in the isolated DNA is so small (see Table II) that such contamination, if it occurred, could not be significant.

A.A. MOghISSI: Both the deproteinization and phosphate buffer treatment are processes associated with the potential of protium activation and thus hydrogen exchange. You may have quantitatively recovered the DNA, but how can you be sure that you have quantitatively recovered tritium?

V. KASCHE: May I first refer you to Fig.2D of the paper? Labelled DNA that had been isolated using the procedure described was here added to a cold homogenate. The DNA isolation method was carried out as described. More than 95% of the labelled DNA was recovered in the DNA fraction. Thus the exchange of tritium from DNA due to the method cannot be a serious matter.

A.A. MOghISSI: You appear to support the hypothesis that the T/H ratio increases in the food chain. The evidence, however, points overwhelmingly in the opposite direction, namely towards a 'constant' T/H ratio. Some years ago we fed rabbits tritiated food and water and observed them over three generations. The T/H ratio remained approximately constant throughout. The discrepancies in the environmental values relate to the changes in T concentration resulting from weapons testing and differences in Tb for various components of the biosphere and the biological system.

V. KASCHE: You may be right, but I feel that carefully planned experiments are needed in order to provide a final answer to this question. At present we have too few data at our disposal.
TRITIUM IN THE AQUATIC ENVIRONMENT

Session 3, Part 2, Session 4
and Session 5, Part 1
TRANSFER AND INCORPORATION OF TRITIUM IN AQUATIC ORGANISMS

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Abstract

TRANSFER AND INCORPORATION OF TRITIUM IN AQUATIC ORGANISMS.

Aquatic plants and animals were studied under natural environmental as well as in laboratory conditions. In *Scenedesmus obliquus*, the tritium in the free water and the tritium organically bound are found to be dependent on the ambient concentration of THO. Rates of uptake and release of tritium by the aquatic plant *Hydrilla verticillata* were studied, and the significance of the persistence patterns of the aqueous and organic phases of tritium in the plant was established under different periods of exposure. Three species of marine algae (*Chlamydomonas reinhardtii, Acetabularia mediterranea* and *Acetabularia crenulata*) have been used to study pollution by tritium. Laboratory experiments were conducted with freshwater fishes (*Tilapia mossambica*) to study the distribution pattern of tritium in the body water of edible tissues from HTO exposure. Differential treatment with starving and feeding the organisms showed no significant difference in the TBT/TFWT specific activity ratio. Tritium incorporation into fishes (*Gymnopterus affinis, Poeciliopsis occidentalis, Salmo gairdneri* and *Ictalurus lacustris*) living in triated water and eating non-tritiated food was also investigated. Rate of uptake and release of tritium by the body water and tissues in *Katsayis sp.* and the body-water turnover rates in shellfish *Anadara granosa* were established. In the case of natural environmental conditions, plants and animals grown in contaminated water were periodically sampled for investigation of tritiated water and organically bound tritium content. The tritium in effluents was in both water and organic forms; the biological availability of the latter forms is discussed.

* Part of the IAEA Co-ordinated Research Programme on the Behaviour of Tritium in the Environment.
1. INTRODUCTION

The main aims of the Co-ordinated Research Programme are to obtain information on the residence time, pattern of movement and distribution of tritium in typical ecosystems, its incorporation into biological compounds and its transfer along food chains to determine the biological significance of this radionuclide.

As aquatic ecosystems in certain parts of the world may be an important source of human foods, research projects concerned with aquatic organisms have been encouraged where marine and fresh water laboratory facilities were available. Aquatic plants and animals reared in laboratory aquaria under controlled conditions were studied for uptake and release patterns of tritium; these laboratory experiments were conducted in Belgium, India and the United States of America.

The ecosystem study involves a small river in which the liquid wastes from a number of nuclear facilities, located in Mol (Belgium), are released. The tritium in effluents was in both water and organic forms. The whole aquatic environment has been prospected and samples have been collected and measured, in particular for the tritium incorporated in the organic matter.

As the detailed data concerning the material and methods used by the participating laboratories are either already published or in press, full information will not appear in the present report which deals mainly with the chief results and their discussion.

2. STUDIES PERFORMED UNDER LABORATORY CONDITIONS

2.1. HTO exposure of freshwater plants

2.1.1. Experimental conditions

In the Lawrence Livermore Laboratory, Harrison and co-workers [1] maintained cattails (Typha angustifolia L.) and filamentous algae (Pithophora sp.) in an experimental pool containing tritiated water (initial concentration 80 μCi/l); the concentrations of tritium were determined for eight months in the pool water and in the water and organic matter of the tissues of the organisms.

In other participating laboratories, Scenedesmus obliquus [2, 3], Hydrilla verticillata [4] and Chlamydomonas reinhardtii [5] were grown in their respective media with different tritium concentrations (range 0.05 –50 μCi/ml); the exposure time of the algae in the active media varied from 12 days to 42 days in order to determine the period of time required for effective tritium transfer between the two main hydrogen pools of the system: plant water and the organic fraction in the plant. The chemical procedure employed to study the distribution of tritium in different organic constituents of Scenedesmus has been described by Krishnamoorthy and co-workers [2].
2.1.2. Results and discussion

2.1.2.1. Tritium in the free tissue water (TFWT)

The TFWT of the rhizomes and leaf bases of cattails reached 65 to 70% of the tritium concentration of the pool water during the eight months of the experiment, the lack of equilibration being attributed to exchange with atmospheric water by the leaves. On the contrary, in the filamentous algae the TFWT concentration was approximately the same as that of the pool water, the ratio being 1.02.

Similarly, in Scenedesmus experiments [2], the tritium specific activity (tritium/hydrogen) of TFWT was always close to that of environmental water, the values ranging between 0.94 to 1.09.

The rate of uptake and release of tritium for short intervals of time was also investigated [4]. Hydrilla verticillata reached an equilibrium concentration corresponding to 80% of the medium water concentration, when normalized for moisture content of the plant (0.76 wt/wt), within a period of one hour. The release pattern showed three components according to least-square fit: the mean residence time corresponding to TFWT phase was 0.36 h and the rate of uptake can be calculated as 0.028 ml/min per g.

2.1.2.2. Tissue-bound tritium (TBT)

The specific activity of the organically bound tritium in cattail tissues was less than that of the pool water (40 to 65%); no strong relationship was apparent between plant part and level of TBT. The ratios of TBT to TFWT were also less than unity (0.62 to 0.99) except for the rhizomes where it was about unity. In the filamentous algae, the specific activity of the TBT was about the same as that of the water and the ratio of TBT to TFWT was essentially unity. Thus, the algae appeared to be in a steady state of uptake and loss of tritium with the water, which might be expected in a short-lived simple organism [1]. Likewise the values of the ratio of the specific activity of TBT in Scenedesmus obliquus to the medium water were respectively 0.91 [3] and 0.67 [2] in algae exposed during 20 days to an active medium.

Data reported by Bonotto and co-workers [5] demonstrate that when Chlamydomonas algae are cultured in the presence of tritiated water, the amount of $^3$H incorporated in the organic matter is related to its concentration in the culture medium, whereas the concentration factor ($\mu$Ci/g dry weight: $\mu$Ci/ml medium) remained constant around 0.40 for Scenedesmus and Chlamydomonas. The nature of the organic compounds into which $^3$H is incorporated in Chlamydomonas remains to be studied.

The relative fractions of tritium incorporated in the three phases of the bound and free forms of tritium in the aquatic plant Hydrilla verticillata [4], were found to be 100:3.27:0.77. This shows that 3.27% of tritium is in the
TABLE I. DISTRIBUTION OF TBT IN DIFFERENT ORGANIC FRACTIONS EXTRACTED FROM *Scenedesmus* [2]

<table>
<thead>
<tr>
<th>Solvents</th>
<th><em>Scenedesmus</em> (% extraction at room temp.)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lipid extraction (acetone, CHCl₃ and CH₃OH)</td>
<td>58.3</td>
<td>Mostly lipids, pigments and fatty acids</td>
</tr>
<tr>
<td>Ether</td>
<td>ND</td>
<td>Ether soluble substances</td>
</tr>
<tr>
<td>80% ethanol</td>
<td>1.1</td>
<td>Free amino acids and mostly carbohydrate</td>
</tr>
<tr>
<td>6N HCl</td>
<td>34.0</td>
<td>Protein hydrolysate</td>
</tr>
<tr>
<td>Residue</td>
<td>6.6</td>
<td></td>
</tr>
</tbody>
</table>

Labile TBT form and 0.77% is in the non-labile TBT form; these values agree well with those obtained by Bruner [6].

The sequential solvent extraction procedure has been used to study the distribution of tissue-bound tritium in the various classes of organic constituents of the *Scenedesmus obliquus* cells. The results (Table I) show all constituents to be significantly labelled. The relative specific activity ratio (RSA) calculated with respect to medium concentration indicates that greater proportions of hydrogen atoms were labelled in nucleic acids compared with lipid and lipid fractions, the mean RSA values being respectively 1.13:0.56:0.44 [2].

2.2. HTO exposure of marine plants

2.2.1. Experimental conditions

Four species of marine algae have been used in the Belgian participating laboratory: *Acetabularia mediterranea, Dunaliella bioculata, Porphyra* sp. and *Ulva lactuca*. The first two species were grown in the laboratory [7, 8], the others were harvested on the North Sea beaches and maintained in culture in the presence of chloramphenicol (10 µg/ml) to prevent the growth of marine bacteria. The exposure time of these algae in the active media varied from two days to 27 days, and the tritium concentration in seawater was about 0.03 µCi/ml.

DNA was prepared by phenol extraction as described by Heyn and co-workers [9] or by pronase treatment as reported by Lurquin and co-workers.
FIG. 1. Incorporation of $^3$H in the dry matter of green algae, Ulva lactuca.

[10]; sepharose-sieved DNA has been analysed for its buoyant density by ultracentrifugation in CsCl gradient, in the presence of marker DNAs (Clostridium perfringens: $\rho = 1.691$ g/cm$^3$; Streptomyces coelicolor: $\rho = 1.730$ g/cm$^3$). Tritiated DNA was precipitated with cold 5% TCA in the presence of 100 $\mu$g of serum bovine albumin, collected on Millipore filters and counted in a liquid scintillation counter.

The unicellular algae Chlamydomonas sp. were grown in tritiated seawater, previously filtered, autoclaved and slightly enriched in nutrients [2]. Experiments performed in the Bhabha Atomic Research Center were carried out with varied tritium concentration in the medium from 0.6–4.1 $\mu$Ci/ml of seawater. A sequential solvent extraction procedure has been used to study the distribution of tissue-bound tritium in the various organic fractions.

2.2.2. Results and discussion

2.2.2.1. Incorporation and distribution of tritium in algae

The uptake and incorporation of tritium in the green algae Ulva and in the red algae Porphyra are illustrated in Fig. 1 and Fig. 2 respectively.
FIG. 2. Penetration of $^3$H in red algae, *Porphyra* sp., and incorporation in the dry matter.

### TABLE II. RATE OF UPTAKE OF TBT IN *Chlamydomonas* sp. CELLS [2]a

<table>
<thead>
<tr>
<th>Time of growth (days)</th>
<th>Cell doubling</th>
<th>Activity in the cells (µCi/ml combustion water)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>3.56</td>
<td>0.28</td>
</tr>
<tr>
<td>7</td>
<td>5.55</td>
<td>0.31</td>
</tr>
<tr>
<td>9</td>
<td>6.87</td>
<td>0.44</td>
</tr>
<tr>
<td>10</td>
<td>7.11</td>
<td>0.54</td>
</tr>
<tr>
<td>13</td>
<td>7.81</td>
<td>0.65</td>
</tr>
<tr>
<td>15</td>
<td>7.50</td>
<td>0.70</td>
</tr>
</tbody>
</table>

a $^3$H concentration: 1.18 µCi/ml seawater.
FIG. 3. Acetabularia mediterranea. Molecular sieve chromatography on sepharose 4B of a crude extract (6.5 ml) of isolated chloroplasts. Column size: 2.5 X 36 cm; eluting solution: 2M NaCl; flow rate: 36 ml/h; fraction size: 1.8 ml; room temperature: 22°C. (From Ref. [5].)

The rate of incorporation of TBT in the cell culture of *Chlamydomonas* sp. in relation to cell doubling is given in Table II. The cultures had grown to a maximal bloom, attaining a cell doubling factor of 7–8 within two weeks, and further additional culture time did not significantly increase the population sizes whereas the cells lost their mobility and sank to the bottom of the flask. The TBT in the dry cells of *Chlamydomonas* sp. reached a level of 0.70 μCi/ml combustion water in a medium of 1.18 μCi/ml seawater corresponding to an average of 63% of the medium concentration.

The lipid extraction procedure with acetone and CHCl₃–CH₃OH mixtures removed about 40–45% of TBT; incorporation of tritium in the lipid fraction was an excellent index of fatty acid biosynthesis [11].
Another major fraction of TBT was obtained with 6N HCl extraction which primarily consisted of protein hydrolysate [12], the amino acids obtained from the protein hydrolysate forming 70–80% of the total protein content of the algae. The appearance of tritium in the protein fraction reflected the extent of incorporation of tritium in non-exchangeable protons of peptide amino acids, though proteins are known for their ability to acquire tritium by exchange with media protons [13].

The penetration and distribution of tritium in the different constituents of algae *Acetabularia mediterranea* and *Acetabularia crenulata* have also been studied [14]. The separation of the radioactivity taken up and incorporated, carried out by conventional biochemical methods, has shown that an important part of tritium is present in the fraction including the small molecules (water, amino acids, . . . ); however, an appreciable part is recovered in the fractions formed by nucleic acids (DNA and RNA) and proteins. The amount of tritium incorporated by the algae increases as a function of the concentration of tritium in the seawater and the duration of exposure.

Purified chloroplast of *Acetabularia mediterranea* were processed as reported previously [10], and their extract submitted to molecular sieving on a sepharose 4B column. Figure 3 shows that a distinct radioactive peak is eluted from the column in the fraction 21–26 and that it is followed by a second radioactive peak. In order to check the nature of the material present in the first radioactive peak, further experiments have been performed [5], and that the labelled material was indeed DNA was demonstrated by ultra-centrifugation in a CsCl gradient, where it gave a single peak having a buoyant density of 1.703 g/cm³, which is characteristic of *Acetabularia* chloroplast DNA.

2.3. HTO exposure of freshwater animals

2.3.1. Experimental conditions

2.3.1.1. Aquaria

The following fish species were studied in the various laboratories participating in the IAEA Co-ordinated Research Programme:

*Carassius auratus* (goldfish) and *Salmo trutta forma fario* (trout), Department of Radiobiology (Mol); *Gambusia affinis* (mosquito fish), *Poeckilopsis occidentalis* (Gila topminnow), *Salmo gairdneri* (trout) and *Ictalurus lacustris* (catfish) in the EPA Laboratory (Las Vegas); *Tilapia mossambica* in the Bhabha Atomic Research Center Laboratory (India) [15]. Fish were exposed in tritiated water from 10–30 days (Belgium), up to 32 days (India) and up to 203 days (U.S.A.); the fish were fed commercial food (Belgium, U.S.A.) and tritiated food (India, Belgium).
2.3.1.2. Outdoor pools

In the Lawrence Livermore Laboratory a fibre-glass pool [1] was filled with 2000 gallons of stream water and tritiated water added to a concentration of 80 μCi/l; the pool was stocked with rhizomes of cattails, filamentous algae and mixed plankton obtained from a natural pond, several hundred freshwater clams (Anodonta nuttalliana Lea), 25 crayfish (Astacus sp.) and 25 goldfish (Carassius auratus). Six plastic-lined pools [16], constructed at the EPA Experimental Farm on the Nevada Test Site, were filled with 3700 litres of tritiated water. Inorganic fertilizer and green algae were introduced. Windblown dirt, debris and hay from the farm entered the pools, and a variety of insect life was observed in the pools before mosquito fish (Gamusia affinis) were introduced. Four pools were stocked in mid-July and the fish harvested in mid-October.

2.3.2. Results and discussion

It was observed [1, 15, 17] that the tritium in the free tissue water (TFWT) remained close to the tritium concentration of the medium water; after one hour of exposure to tritiated water the TFWT concentration in Tilapia attains 70% of the medium concentration [15]. Some fish remaining at the end of the uptake experiment were returned to non-tritiated water for observation of their rates of loss of tritium. The biological half-life is found to be 48 ± 6 min in the Tilapia experiment; in goldfish and in clams the rate of loss was very rapid and the curve is characterized by at least two components [1]: the short-lived component, which represents more than 95% of the tritium, had a half-life of about one day and the long-lived component had a half-life of about 15 days.

2.3.2.1. Tissue-bound tritium (TBT)

The clams obtained tritium from the water as well as from their food, i.e. from the micro-organisms and the detritus in suspension in the water. The concentration of TBT changed with time and at different rates for different tissues [1]. It increased up to about Day 90, when the maximum values were observed in most tissues. The concentrations were highest in the visceral mass and lowest in the calcareous tissue. Since the crayfish could be sampled only twice, at Days 5 and 26, the maximum TBT values probably were not observed. They were highest in the digestive gland and lowest in the carapace. The TBT concentrations for crayfish muscle were about the same as for clam visceral mass.

2.3.2.2. Specific activities

In Table III, selected results are presented to show the ratio between the tritium specific activity in dried fish tissue and that in water in which the fishes
### TABLE III. SPECIFIC ACTIVITY OF TRITIUM IN DRY TISSUE OF FISH GROWN IN AQUARIA OR IN OUTDOOR POOLS

<table>
<thead>
<tr>
<th>Species</th>
<th>Exposure conditions</th>
<th>Specific activity ratio&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>2&lt;sup&gt;H&lt;/sup&gt;-food</td>
<td>Commercial food</td>
</tr>
<tr>
<td><strong>Tilapia mossambica</strong></td>
<td>Aquarium</td>
<td>30</td>
<td>0.025&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td><strong>Carassius auratus</strong></td>
<td>Aquarium</td>
<td>32</td>
<td></td>
</tr>
<tr>
<td><strong>Salmo gairdneri</strong></td>
<td>Pool</td>
<td>173–214</td>
<td>0.15&lt;sup&gt;e&lt;/sup&gt;</td>
</tr>
<tr>
<td>Dead eggs</td>
<td>Aquarium</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Dead fry</td>
<td>Aquarium</td>
<td>32</td>
<td></td>
</tr>
<tr>
<td>Fish</td>
<td>Aquarium</td>
<td>140</td>
<td></td>
</tr>
<tr>
<td><strong>Salmo trutta forma fario</strong></td>
<td>Aquarium</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>Adult</td>
<td>Aquarium</td>
<td>10</td>
<td>0.16&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td><strong>Gambusia affinis</strong></td>
<td>Aquarium</td>
<td>21</td>
<td></td>
</tr>
<tr>
<td>Young</td>
<td>Pool</td>
<td>11</td>
<td>0.36</td>
</tr>
<tr>
<td>Young</td>
<td>Pool</td>
<td>39</td>
<td>0.56</td>
</tr>
<tr>
<td>Adult</td>
<td>Aquarium</td>
<td>60</td>
<td>0.37</td>
</tr>
<tr>
<td>Adult</td>
<td>Aquarium</td>
<td>203</td>
<td>0.52</td>
</tr>
<tr>
<td>Adult</td>
<td>Pool</td>
<td>58</td>
<td>0.63</td>
</tr>
<tr>
<td>Adult</td>
<td>Pool</td>
<td>93</td>
<td>0.88</td>
</tr>
<tr>
<td><strong>Procleiopsis occidentalis</strong></td>
<td>Aquarium</td>
<td>88</td>
<td></td>
</tr>
<tr>
<td>Young</td>
<td>Aquarium</td>
<td>185</td>
<td></td>
</tr>
<tr>
<td>Adult</td>
<td>Aquarium</td>
<td>79</td>
<td></td>
</tr>
<tr>
<td><strong>Ictalurus lacustris</strong></td>
<td>Aquarium</td>
<td>3</td>
<td>0.51</td>
</tr>
<tr>
<td>Dead eggs</td>
<td>Aquarium</td>
<td>133</td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup> Tritium specific activity in dry tissue divided by that in medium water.

<sup>b</sup> Fertilized eggs placed in tritiated water when received.

<sup>c</sup> Edible part only (muscle).
TABLE IV. TURNOVER CONSTANTS AND BIOLOGICAL HALF TIMES FOR TRITIUM EXCHANGE IN TISSUES OF MARINE INVERTEBRATES

*Values determined during period of tritium loss*

<table>
<thead>
<tr>
<th></th>
<th>K</th>
<th>$T_\frac{1}{2}$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>Mya arenaria</em> (fed)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Body water hydrogen</td>
<td>0.33 h$^{-1}$</td>
<td>2.1 h</td>
<td>[18]</td>
</tr>
<tr>
<td>Organic matter hydrogen</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rapidly exchangeable</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Muscle</td>
<td>0.28 h$^{-1}$</td>
<td>2.5 h</td>
<td></td>
</tr>
<tr>
<td>Viscera</td>
<td>0.29 h$^{-1}$</td>
<td>2.5 h</td>
<td></td>
</tr>
<tr>
<td>Slowly exchangeable</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Muscle</td>
<td></td>
<td>290 d</td>
<td>[22]</td>
</tr>
<tr>
<td>Viscera</td>
<td></td>
<td>120 d</td>
<td>[22]</td>
</tr>
<tr>
<td><em>Crassostrea gigas</em></td>
<td></td>
<td></td>
<td>[18]</td>
</tr>
<tr>
<td>Body water hydrogen</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pooled (fed)</td>
<td>0.97 h$^{-1}$</td>
<td>0.72 h</td>
<td></td>
</tr>
<tr>
<td>Pooled (no food)</td>
<td>0.52 h$^{-1}$</td>
<td>1.34 h</td>
<td></td>
</tr>
<tr>
<td><em>Cancer productus</em> (no food)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Body water hydrogen</td>
<td></td>
<td></td>
<td>[18]</td>
</tr>
<tr>
<td>1.8 h$^{-1}$</td>
<td>0.37 h</td>
<td></td>
<td></td>
</tr>
<tr>
<td><em>Katelysia opima</em> (no food)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Body water hydrogen</td>
<td></td>
<td></td>
<td>[20]</td>
</tr>
<tr>
<td>0.7 h$^{-1}$</td>
<td>1.0 h</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Organic matter hydrogen</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rapidly exchangeable</td>
<td>0.24 d$^{-1}$</td>
<td>2.9 d</td>
<td></td>
</tr>
</tbody>
</table>

were exposed and grown. A ratio greater than unity would indicate concentration of tritium within the organic constituents of fish relative to the water environment. No ratio greater than unity was measured in the experiments reported. No significant differences are found in the specific activity ratios by feeding the *Tilapia* with tritiated food or starving them, probably because the maximum ratio attained is itself very low in both cases. Moreover no significant difference is observed between the values of the specific activity ratio in the muscle of trout fed commercial or tritiated food: probably the intake was too low during the short exposure period. Results of experiments performed in the EPA Laboratory [16] show that the consumption of food grown in the tritiated environment increases the tritium specific activity in tissue of mosquito fish to levels about 50 to 90% higher than in fish grown in tritiated water and fed commercial foods.
<table>
<thead>
<tr>
<th></th>
<th>Time of exposure (days)</th>
<th>Mean moisture content (%)</th>
<th>Activity (nCi (^{3}\text{H}/\text{g})</th>
<th>Incorporation (%)</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Seawater</td>
<td>Wet matter</td>
<td>Dry matter</td>
</tr>
<tr>
<td><em>Asteria rubens</em></td>
<td>2</td>
<td>64.3</td>
<td>29</td>
<td>10.7</td>
<td>0.22</td>
</tr>
<tr>
<td><em>Crangon vulgaris</em></td>
<td>10</td>
<td>80.3</td>
<td>33</td>
<td>17.7</td>
<td>0.80</td>
</tr>
<tr>
<td><em>Mytilus edulis</em></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Whole animal</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soft tissues</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shell</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><em>Anadara granosa</em></td>
<td>14</td>
<td>80.0</td>
<td>67.6</td>
<td>43.8</td>
<td>2.7</td>
</tr>
<tr>
<td>Soft tissues</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Limited data are available on the loss of tissue-bound tritium after transfer of fish to a less tritiated environment. It is clear, however, that a portion of tritium in tissue is not excreted rapidly [16]. This finding is discussed in the conclusions of the present report.

2.4. HTO exposure of marine animals

As little information was available on the rate of tritiation of body water in the marine organisms and its exchange to the organic form, investigations were initiated in three participating laboratories (Lawrence Livermore Laboratory, Bhabha Atomic Research Center, Belgian Nuclear Centre) to study the rate of incorporation and loss of tritium in the body water and in the organic fraction of various marine invertebrates.

2.4.1. Experimental conditions

The clam *Mya arenaria* and the crab *Cancer productus* were obtained from San Francisco Bay, the Japanese oyster *Crassostrea gigas* was obtained from Tomales Bay; these animals were placed in tritiated (126 nCi/ml) seawater in a fibre-glass pool previously described [18], a dense resident population of Chlorella-like organisms being present in the water. *Katelysia opima* and *Anadara granosa* were collected near Bombay and transferred into glass aquaria filled with seawater, and the tritiated water was added to a concentration of 67.6 nCi/ml. The animals were given no food during the experimental period [19, 20]. *Crangon vulgaris, Asteria rubens* and *Mytilus edulis* were obtained from the Belgian North Sea coast [21] and maintained in artificial seawater during the experimental period, the animals being fed with commercial food.

2.4.2. Results and discussion

2.4.2.1. Turnover of body water

The specific activity of tritium in the body water of clams that had been maintained in tritiated water for seven days decreased rapidly when they were removed to filtered, continuously flowing, tritium-free water. The loss curve had only one component; the rate constant was 0.33 h⁻¹ and the biological half-time was 2.1 h (Table IV). These results indicate that the tissue-free water behaves as a single pool, and that the turnover rate is very rapid [18]. In the oyster, another filter feeder, the turnover rate of tissue-free water was measured for comparison with clams. As can be seen from Table IV, the turnover rate constants determined for pooled samples differed in the presence (0.97 h⁻¹) and absence (0.52 h⁻¹) of food organisms in the water. As a further basis for comparison, turnover rates were measured for the tissue-free water of the crab
body fluid: the rate constants calculated were 1.8 h\(^{-1}\) during loss and the corresponding half time was 0.37 h. *Katelysia opima* reached nearly 80\% of the medium specific activity within an hour, and body water tritium gradually increased further in 48 h to 88\%, followed by the decrease in the medium during the remaining of the experiment. Equilibrated animals when transferred to the inactive medium released tritium quickly within a few hours; the loss curve had only one component. The rate constant was found to be 0.7 h\(^{-1}\) and the biological half time was 1 h (Table IV).

2.4.2.2. Turnover in organic material

Changes in specific activity were followed also in the rapidly and slowly exchangeable compartments of the organic material of muscle and visceral tissue of *Mya arenaria* (Table IV), these tissues being chosen for study because they usually differ considerably in their rates of hydrogen turnover. Muscle and viscera tissue were essentially the same in their rates of decrease in specific activity with time in the rapidly exchangeable compartments (Table IV). Hydrogen turnover was followed also in the slowly exchangeable compartment of the organic material of clam muscle and viscera: virtually no change was observed in this slowly exchangeable hydrogen compartment. This is consistent with the results of an earlier experiment [22]: the $T_1$ was about 290 days for muscle and about 120 days for viscera (Table IV).

In Table V, data on the build-up of organic bound tritium in various marine invertebrates show that the level of tritium accumulation in the organic tissue is very low compared with body-water concentration. The retention of tritium in the body tissue depends upon the chemical form and in the present cases the animals are fed with tritiated water.

2.5. Biological availability of tritium released in liquid effluents

Tritium in liquid effluents may have multiple origins and may prevail in different physico-chemical states. Certain organic tritium compounds may constitute a particular risk due to their preferential absorption by living organisms.

Algae (*Scenedesmus obliquus*) cultivated in effluents can be used to detect such molecules [3] and consequently for the evaluation of the importance of this contaminating fraction. The culture of algae on effluents constitutes therefore a biological test.

Results of application of this method, on laboratory effluents originating from synthesis of labelled molecules and effluents released from nuclear power plants of the PWR type, are presented in Table VI. In the case of effluents released by a radiochemical laboratory, the results showed that the tritium is preferentially incorporated in the organic matter of the algae but the percentage
TABLE VI. SPECIFIC ACTIVITY OF TRITIUM IN DRY TISSUE OF ALGAE
(Scenedesmus obliquus) GROWN IN LIQUID EFFLUENTS\(^a\) RELEASED BY A
RADIOCHEMICAL LABORATORY AND BY TWO PWR NUCLEAR POWER
PLANTS

<table>
<thead>
<tr>
<th>Origin of effluent</th>
<th>Initial (^3)H conc. in culture medium (nCi/ml)</th>
<th>(^3)H content in combustion water of dry tissue ((\mu)Ci/ml)</th>
<th>Specific activity ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radiochemical Lab.</td>
<td>52</td>
<td>1.41</td>
<td>27.1</td>
</tr>
<tr>
<td>Radiochemical Lab.</td>
<td>76.5</td>
<td>4.10</td>
<td>53.6</td>
</tr>
<tr>
<td>Radiochemical Lab.</td>
<td>70.3</td>
<td>3.08</td>
<td>43.8</td>
</tr>
<tr>
<td>Nuclear power plant</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SEMO</td>
<td>5.1</td>
<td>4.60</td>
<td>0.90</td>
</tr>
<tr>
<td>SENA</td>
<td>16.6</td>
<td>14.50</td>
<td>0.87</td>
</tr>
</tbody>
</table>

\(^a\) The effluents were filtered on Millipore (0.45 \(\mu\)m) before mixing with culture medium.

of \(^3\)H biologically available is lower than 1% of the total \(^3\)H present in these effluents. This small fraction is, however, important for the contamination of the food chain.

The values of the specific activity ratio are about one if the effluents released by the PWR nuclear power plants are examined; this means that there was no organic tritium biologically available present in the filtered effluents studied; however, some \(^3\)H was present in the insoluble organic fraction which contained more particularly micro-organisms.

A freshwater fish (*Salmo trutta forma fario*) has been also used as biological test. The results showed that the values of the specific activity ratio (tritium specific activity in dry tissue divided by that in medium water) varied from 1.55 to 27.6, depending on the organs analysed, when the fishes were grown (4 to 8 days) in water in which an effluent released by a radiochemical laboratory was added to a final concentration of 2.4 nCi/ml and 0.24 nCi/ml respectively in the two experiments. Results of experiments performed on the same fish species exposed [17] in tritiated water (1 \(\mu\)Ci/ml) showed that the values of the specific activity ratio ranged from 0.03 to 0.19 for the various organs.
TABLE VII. SPECIFIC ACTIVITY (nCl $^3$H/gH) OBSERVED IN SAMPLES COLLECTED IN A RIVER AND A POND DOWNSTREAM OF A RADIOACTIVE DISCHARGE

A. RIVER

<table>
<thead>
<tr>
<th>Water</th>
<th>Aquatic plants (combustion water)</th>
<th>Eggs (ducks)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximal values</td>
<td>Mean values</td>
<td>Maximal values</td>
</tr>
<tr>
<td>3.15</td>
<td>1.12</td>
<td>35.8</td>
</tr>
</tbody>
</table>

B. POND (No. 4)

<table>
<thead>
<tr>
<th>Water</th>
<th>Fish (dry matter) (combustion water of various organs)</th>
<th>Fish (free water tissue)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.50</td>
<td>0.24</td>
<td>5.73</td>
</tr>
</tbody>
</table>

3. STUDY OF THE TRITIUM TRANSFER IN A NATURAL ENVIRONMENT

The ecosystem studied is a small watercourse receiving liquid effluents from complex nuclear facilities located in Mol (Belgium). Tritium in effluents was in water as well as in organic forms. The river feeds intermittently a series of ponds used for fish rearing. The ecosystem was investigated for about one year, the tritium concentration in water and sediment samples and in living organisms being determined. The emphasis was placed on measurement of the tritium incorporated in the organic matter, the specific activity of the combustion water being determined in each case.

Table VII presents some general values of specific activity observed in the samples collected in the river and in one of the ponds. When these values are examined it appears that those of the tritium incorporated in organic matter are higher than those of the river water by a factor as high as 20. This unexpected observation was in conflict with results from experiments in laboratory aquaria in which tritium was present as THO. Therefore one must look for biologically available tritiated compounds in the effluents released in the ecosystem under study. The presence of these compounds were experimentally proved later and their biological availability evaluated (see Section 2.5). It would appear that the physico-chemical state of the tritium is a sensitive parameter of particular importance for predicting and evaluating population exposure [23].
REFERENCES


DISCUSSION

R.V. OSBORNE (Chairman): The work that you have just reported is, I believe, part of a continuing programme. What are you proposing now for the next stage of this programme?
R. KIRCHMANN: We should like to evaluate the specific activities of the organic matter present in various compartments (sediments, plants and animals) of freshwater ecosystems. One of the sites under study receives routine releases of liquid effluents from nuclear installations, while the second type of site investigated is contaminated only from 'fall-out' tritium. The results so far show that $^3$H-bound tissue (fish) accounts for some 60–70% of the total tritium ingested through consumption of fish caught in both types of freshwater ecosystem investigated.

D.J. MEWISSEN: What would be the relative content of proteins in the dry residue?

R. KIRCHMANN: In the dry tissue of the fish muscle, which is what is being considered here, the content of proteins is nearly 90%.
MICROBIAL ACTIVITY MEASUREMENTS
BY MEANS OF
TRITIUM-LABELLED SUBSTRATES*

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Abstract

MICROBIAL ACTIVITY MEASUREMENTS BY MEANS OF TRITIUM-LABELLED
SUBSTRATES.

Two improved methods to determine microbial activity at nearly in situ conditions are
reported. The first is valid for the identification of active bacteria metabolizing on easily
degradable organic compounds by means of $^3$H-micro-autoradiography. The second method
provides reliable uptake rates of $^3$H-substrates with one substrate concentration added to the
sample. Uptake velocities were calculated from the Lineweaver-Burke equation. Measurements
of active bacterial numbers and biomass as well as heterotrophic activities obtained by applying
these methods to a brackish water environment are discussed. A correlation matrix between
bacterial variables and ecological relevant parameters is established. An average annual number
of $970 \times 10^3$ ml$^{-1}$ active bacteria were found in the research area, that is 41% of the total
number of bacteria. Active bacterial biomass averaged $10.3 \mu g \, C \, l^{-1}$ in comparison with
19.3 $\mu g \, C \, l^{-1}$ total bacterial biomass. Turnover times of substrates ranged between 7 and 147 h
for amino acids. Uptake velocities ($V_m$) of four $^3$H-substrates (amino-acid mixture, leucine,
glucose, acetate) exhibited a close relationship with chlorophyll a concentrations and water
temperature. The similarity of $V_m$ plots indicates the presence of uptake properties for
different substrates in bacteria living in areas with insufficient nutrient supply.

INTRODUCTION

The uptake of dissolved organic substrates from aqueous solutions and of
particulate matter (after hydrolysis by exo-enzymes), is a fundamental process in
nature which is dominated by bacterial activity. The main components of
naturally produced organic compounds are carbon, hydrogen, oxygen and nitrogen.
In the basic process of photosynthesis water is involved as well as carbon dioxide
and inorganic nutrients.

Therefore primary production of phytoplankton can be traced by $^{14}$C as well
as by $^3$H. The former approach introduced by Steeman-Nielsen [1] is widely

* Research supported by the Deutsche Forschungsgemeinschaft, Bonn - Bad Godesberg.
INCUBATION

WATER SAMPLE 1 - 4 ml

INCUBATION OF 3h AT 20°C OR IN SITU TEMPERATURE

FORMALDEHYDE (35%)  

3h SUBSTRATE

ISOTONIC NaCl SOLUTION

FILTRATION THROUGH NUCLEOPore FILTERS (0.2μm PORE SIZE)

NUCLEOPore FILTER

FILTER HALVES ON WET RUBBER LAYER

AUTORADIOGRAPHY

DEVELOPMENT AND FIXATION OF THE FILM PLATE

FILM PLATE WITH FILTERS EXPOSED FOR 14 DAYS

X-RAY FILM PLATE PRESSED ON THE FILTERS IN THE DARKCHAMBER

MEASUREMENT

SPOTS CAUSED BY BACTERIA

PLUMBICON CAMERA ON MICROSCOPE

AUTOMATIC IMAGE ANALYSER (TYPE QUANTIMET 720)

ON-LINE CALCULATOR

CALCULATION

AREA, INTERCEPT, PERIMETER AND COUNT OF SPOTS CAUSED BY BACTERIA.  
CALCULATION OF ACTIVE BACTERIA PER ml, ACTIVE BACTERIAL BIOMASS AND 'SPOT-VOLUME' AS A RELATIVE MEASUREMENT OF BACTERIAL ACTIVITY.

FIG.1. Determination of active bacteria.
applied, and the latter has been realized by McKinley and Wetzel [2]. The converse reaction to photosynthesis, breakdown and uptake of organic substrates by heterotrophic bacteria may also be followed by both $^{14}$C and $^{3}$H. Substrate uptake of $^{14}$C-labelled substances according to the procedure suggested by Wright and Hobbie [3] has been adopted by several investigators [4-7]. Tritiated substrates for this purpose have been used by Azam and Holm-Hansen [8], Dietz, Albright and Tuominen [9] and Hoppe [10]. The determination of heterotrophic microorganisms involved in the carbon cycle by micro-autoradiography has almost exclusively been performed using tritiated substances [11-13]. In this study two improved methods for determining microbial heterotrophic activity are described briefly. An annual cycle of heterotrophic activity and active bacteria numbers and biomass has been recorded from an eutrophic brackish water environment. The relationship between microbial activity and relevant ecological parameters is discussed.

METHODS

Water samples were taken under sterile conditions from an eutrophic brackish water biotope of the Kiel Fjord (Western Baltic Sea) at monthly intervals throughout the year. The main microbiological parameters determined were: number and biomass of active bacteria, uptake velocity and turnover time of different tritiated compounds, number and biomass of total bacteria and colony-forming units. In addition, the chlorophyll a concentration, composition of the phytoplankton population, DOC, and various inorganic chemical as well as hydro-graphical and meteorological data were collected. The number of actively metabolizing bacteria was determined by means of $^{3}$H-micro-autoradiography. The method is designed for routine work and can be easily standardized. The dark chamber procedure is reduced to a minimum. Special materials required are: $^{3}$H-substrate, Nuclepore filters and X-ray stripping film (Kodak AR 10). $^{3}$H-amino-acid mixture of high specific activity was chosen as a substrate to label bacteria at near in situ conditions. The basic approach has been published by Hoppe [10, 13]. The automated version is depicted in Fig.1.

From a water sample an aliquot (1-4 ml) is pipetted into a glass vial and supplemented with 0.1-10 $\mu$Ci ml$^{-1}$ $^{3}$H-amino-acid mixture (Amersham - Buchler, Braunschweig). One of the parallels serves as a blank and is fixed immediately with 20 $\mu$l ml$^{-1}$ of formalin to check adsorption and contamination of the label. After 3 h incubation at standard (20°C) or in situ temperature the test samples are fixed with the same amount of formalin, and 30 ml of isotonic water is added to dilute radioactivity and bacteria in the sample. The bacteria in the sample are collected on a 0.2 $\mu$m Nuclepore filter by gentle filtration. One half of the filter (the other may be used for fluorescence staining) is placed on a moistened foam
Preliminary: 3H-substratum, 0.5μCi 100 μl⁻¹ in each bottle.

Final concentration of added substratum (μCi l⁻¹): 0.0073, 0.0073, 0.0073, 0.0073, 0.0073, 0.0073.

Water sample, pre-filtered (50 μm mesh), 50 μl in each bottle.

Incubation: Incubation of 1 to 3 h at in situ temperature.

Measurement of radioactivity: Transfer of wet filters.

Scintillation cocktail, 10 μl.

Calculation:

Data input: Standard (cpm), incubation time (h), substrate concentrations of bottles 1–6 (μg C l⁻¹), blanks (bottles 2, 3) and samples (bottles 1, 4–6) (counts/min).

Data output: $\tau_0$ (h), calculated from bottles 1 and 2; $V$ (μg C l⁻¹ h⁻¹), $K_0 + S_0$ (μg C l⁻¹), calculated from bottles 3–6.

Fig. 2. Determination of heterotrophic activity by means of tritium-labelled substrates.
rubber sheet. Twenty-five half filters can be positioned in a defined order on a space of 12.0 × 16.3 cm. This means that these filters are treated in the same manner in the subsequent process (seldom achieved in autoradiography). In the dark chamber a wetted stripping film plate (Kodak AR 10) is gently pressed on to the filters on the foam rubber. When the stripping film plate is separated from the foam rubber the filters adhere to the film plate in close contact. Subsequent exposition took 14 days. Development was 3 min with Kodak D 19 developer, followed by a 0.5-min water bath and 3 min of fixation. Evaluation of the stripping film was performed with a bright field microscope and an automatic image analyser (Quantimet 720).

Substrate uptake parameters (V_m, T_t, K_t + S_n) were measured by means of tritiated substrates (3H-amino-acid mixture, 3H-leucine, 3H-glucose, 3H-acetate). There are two advantages in favour of using tritium-labelled compounds rather than 14C-substrates: (i) Tritiated substrates are commercially available with very high specific activities (e.g. 3H-leucine, 100 Ci/mmol). This allows the addition of minute quantities of label in comparison with the natural substrate concentration. The rate of substrate uptake can therefore be directly calculated without the need for extrapolation as in 14C-experiments. (ii) At high ambient nutrient concentrations, 3H-substrates of high specific activity provide sufficient measurements of uptake rate and of V_m. Thus the use of 3H-substrates enlarges the applicability of the uptake kinetical approach to nutrient-poor and nutrient-rich water samples.

In the procedure (see Fig.2), aliquots of 50 ml samples were supplemented with 0.5 μCi of 3H-substrate (final concentration 0.0059 μg C l⁻¹ leucine) for uptake rate measurements; 0.5 μCi of 3H-substrate plus increasing quantities of unlabelled substrate (final concentration 10, 30 and 50 μg C l⁻¹) were added to determine the uptake velocity using the kinetic model. Controls were fixed before 0.2% formalin was added to the radioactive substrate. After incubation for 1 to 3 h at in situ temperature on a laboratory shaker, test samples were fixed as described above. The samples were filtered under low vacuum pressure (0.2 atm) through low quench cellulose-nitrate filters (Sartorius, 0.2 μm pore size, 22 mm diameter) and the bacteria-bearing filters were transferred into scintillation vials containing 10 ml of scintillation cocktail (5.5 g Permabland Packard, 120 g naphthalene in 1 litre dioxane). Using a 3H-standard provided by Amersham — Buchler (Braunschweig) the efficiency of scintillation counting was found to be 41%. Calculations of the uptake rate (f/t) and uptake velocity (V_m) were derived from the following equations:

\[
\frac{f}{t} \times 100 = \frac{v}{S} \quad \text{(uptake rate in % h⁻¹) (according to Ref. [8])} \quad (1)
\]

\[
\frac{t}{f} = \frac{S}{v} \quad \text{(turnover time (h))} \quad (2)
\]
where \( f \) = fraction of the incorporated radioactivity as measured dis/min
incorporated versus dis/min initially added to the sample
\( t \) = time of incubation (h)
\( v \) = uptake velocity of the substrate at ambient substrate concentration
(\( \mu g \; C \; L^{-1} \; h^{-1} \))
\( S \) = ambient substrate concentration (\( \mu g \; C \; L^{-1} \))

\[
\frac{S + A}{v} = \frac{A}{V_m} + \frac{K + S}{V_m} \quad \text{(Lineweaver-Burke modification of Michaelis-Menten equation)}
\]  

(3)

\[
\frac{S + A}{v} = \frac{C \mu t}{c}
\]  

(4)

where \( A \) = substrate added to the sample (\( \mu g \; C \; L^{-1} \))
\( V_m \) = maximum velocity of substrate uptake (\( \mu g \; C \; L^{-1} h^{-1} \))
\( K \) = half-saturation constant (\( \mu g \; C \; L^{-1} \))
\( C \mu t \) = radioactivity added to the sample (dis/min)
\( c \) = radioactivity incorporated by the bacteria of the sample (dis/min)

If values for \( A \) are plotted against \( C \mu t/c \), \( V_m \) may be calculated from the slope of the regression line (Eqs 3 and 4). When investigations with \(^3\)H are reported, an objection often raised is that high rates of self-decomposition of the label should be taken into account. The extent of self-decomposition, however, should not be overestimated. According to the data of Evans [14] for \(^3\)H-amino acids and \(^3\)H-carbohydrates of high specific activity, it is less than 1–2% per month (at +2°C and 2% ethanol as a radical scavenger). Tritiated water, as formed by exchange on storage of tritium-labelled compounds in aqueous solutions, is only one of seven general features responsible for loss in activity. If necessary, tritiated water may be evaporated under vacuum and the radioactive residue may be redissolved for immediate application.

RESULTS AND DISCUSSION

In this study the \(^3\)H-tracer methods described above were used to follow the links between phytoplankton development and pollution effects on the one hand and heterotrophic uptake of dissolved organic compounds by microorganisms on the other. From the microbiological point of view emphasis was placed upon the elucidation of interrelationships between bacterial numbers, bacterial biomass and uptake activity parameters. The tritiated substrates used for this purpose were \(^3\)H-amino-acid mixture, \(^3\)H-leucine, \(^3\)H-glucose and \(^3\)H-acetate. The characteristics of the sampling area (Inner Kiel Fjord) are as follows: Annual fluctuations of salinity and temperature from 11 to 26 %; S and −1.2°C to 22°C respectively.
Average values of primary production about 200 g Cm⁻²y⁻¹, of chlorophyll a 5.6 µg l⁻¹, of phosphate 2.98 µg at PO₄³⁻—P l⁻¹ and of nitrate 7.3 µg at NO₃⁻—N l⁻¹. The area is influenced by the polluted river Schventine, rainwater drainage and heavy ship traffic.

Mean values and ranges of microbiological data measured during the investigation period 1976–77 are listed in Table 1. As can be calculated from the recorded data, the proportion of active bacteria averages up to 41% of the total number whereas about 0.9% could be isolated on ZoBell Agar (2216 E) of 23% S. In comparison with the number of active bacteria the colony-forming units amounted to 1.9%. From the raw data it was concluded that during the warmer seasons up to 74% of the bacteria present in a water sample revealed uptake properties for the substrate offered. During the winter sometimes only about 10% of the bacteria were actively metabolizing when they were incubated at in situ temperature. Biomass of the total number of bacteria decreased from summer to winter down to 8.3% (summer value = 100%) whereas the active bacterial biomass was reduced to 0.7%. In order to justify the application of relatively high nutrient concentration in our autoradiographic experiments (300 µg C l⁻¹ of amino-acid mixture) experiments were performed with minimized nutrient additions (30 and 3 µg C l⁻¹ amino-acid mixture). With 30 µg C l⁻¹ only a slight decrease in spots counted on the film plate was observed whereas at 3 µg C l⁻¹ 63% of the spots counted at 300 µg C l⁻¹ could still be determined.

The annual cycle of active bacterial number and relative biomass, substrate uptake velocity of four substrates and chlorophyll a in the Kiel Fjord is depicted in Fig.3. The seasonal fluctuations of active bacterial numbers (Fig.3a) show a minimum during autumn and winter with maxima in spring and summer. It is remarkable that the relative active microbial biomass does not follow the trend indicated by the numbers in every case. It is obvious that we have a very high number of active bacteria in June corresponding to a relative low biomass, whereas the maximum of bacterial biomass is reached in July when numbers already decrease. We conclude from this that there are considerable alterations in the species composition of natural occurring bacterial populations within short periods. Additional microscopical observations indicated the dominance of very small free living bacteria (0.2–0.4 µm in diameter) in the June sample, whereas large cells organized in chains determined biomass measurements in July. Plots of substrate uptake velocities of the four substrates (Fig.3b) show a similar pattern of curves. A pronounced peak in July is obvious in all cases as is the minimum in winter and the increase in spring. The close correlation between the four curves may be explained by the suggestion that autochthonous bacteria in a water body with relatively low nutrient supply (in comparison with polluted freshwater) must have more uptake properties ready at the same time to satisfy their nutritional requirements. The comparison of Fig.3a and Fig.3b shows a strikingly close correlation between uptake velocity and active bacterial biomass. Numbers alone do not always present a complete picture of the activity expected.
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>TNB</td>
<td>2035 x 10^3</td>
</tr>
<tr>
<td>Biomass</td>
<td>19.3</td>
</tr>
<tr>
<td>CPU Number</td>
<td>1.8 x 10^3</td>
</tr>
<tr>
<td>V_m</td>
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<td>Glucose</td>
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<tr>
<td>ASM</td>
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<td>Leucine</td>
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</tr>
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<td>0.14</td>
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<tr>
<td>K_1 + S_0</td>
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</tr>
<tr>
<td>T_1</td>
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</tr>
<tr>
<td>Glucose</td>
<td>6.3</td>
</tr>
<tr>
<td>Acetate</td>
<td>1.7</td>
</tr>
<tr>
<td>Chlorophyll</td>
<td>5.6</td>
</tr>
<tr>
<td>DOC</td>
<td>2.44</td>
</tr>
</tbody>
</table>

**Notes:**
- **TNB** = total number of bacteria per ml.
- **MAR** = active bacteria per ml, as determined by 3H-autoradiography.
- **CPU** = colony-forming units per ml (sapphire bits).
- **V_m** = maximal uptake velocity (mg C/l·h).
- **T_1** = turnover time (h).
- **K_1 + S_0** = uptake constant plus natural substrate concentration (mg C/l).
- **DOC** = dissolved organic carbon (mg C/l).
- **ASM** = amino-acid mixture.

Range values are in parentheses.
An explanation of the findings may be provided in Fig. 3c. Chlorophyll a concentrations show the same general features as bacterial biomass and substrate uptake velocity. Although measurements of primary production have not been performed it may be concluded from data of light intensity that samples with high chlorophyll a content also showed high rates of productivity. In blooming algal populations a considerable number of cells had an unhealthy appearance and were heavily populated by bacteria. Excretion of healthy cells and products released by decaying algae may have enhanced microbial development and activity.

To summarize the results obtained, a correlation matrix was established, based on a linear correlation of randomly distributed data. Calculations with the Spearman rank correlation coefficient valid for non-randomly distributed data gave similar results. The general features of the correlation analysis are illustrated in Fig. 4 (for explanation of the symbols see caption). Parameters within the individual compartments (MAR, \( V_m \), \( T_1 \), TNB, \( K_t + S_n \)) are in most cases correlated with one another. This has already been demonstrated for the uptake velocity (\( V_m \)) of the four substrates (ASM, leucine, glucose, acetate). Despite the temporary differences between bacterial numbers and biomass a correlation between these two parameters was found when viewed over the whole year (MAR and TNB). The individual compartments (MAR, TNB, \( K_t + S_n \), \( V_m \)) are very closely linked together as was proved by various correlations at the 99% confidence level. Factors determining bacterial development and activity seem to be chlorophyll a content (as a measure related to planktonic excretion) and water temperature, whereas DOC exhibited almost no relationship with the other parameters measured. DOC gives only the momentary level of an unstable parameter which depends on the current supply of nutrients and the bacteria acting on them. Furthermore, only a small fraction of DOC is available for bacterial breakdown. Colony-forming bacteria (Sapro.) have little influence on activity parameters, but they are closely correlated with the phosphate concentration in the water sample, which is influenced by pollution effects.

Environmental factors influencing the area of investigation vary over a wide range. Therefore bacteria from various origins introduced into this environment may not always find the conditions favourable. Rheinheimer [15] found a close relationship between salinity and the bacterial flora in the western Baltic Sea. At low water temperatures, bacteria from land and sewage showed a longer period of survival in brackish waters than at higher water temperatures occurring in summer. Our autoradiographic observations support the hypothesis recently put forward by Stevenson [16] 'that the physiological state of a significant portion of the bacterial community in most aquatic environments can be described as dormant'. On the other hand we did not find that the small forms 'represent manifestations of exogenous dormancy' [17], as cited in Stevenson [16]. Fractionated filtration as well as microscopical observations indicated that the small forms in particular seem to be well adapted to nutrient-poor conditions in brackish waters. Our annual cycles in the heterotrophic potentials of four substrates
FIG. 3. Annual cycle of active bacteria, uptake velocity and phytoplankton chlorophyll $a$ in a brackish water environment.

(a) Annual cycle of active bacteria (●) and relative biomass of these bacteria (○) as measured by $^3$H-micro-autoradiography at 1 m depth.

(b) Annual cycle of the uptake velocity ($V_m$) of four $^3$H-substrates at 1 m depth.

(c) Annual cycle of chlorophyll $a$ concentrations at 1 m (○), 10 m (●) and 18 m (+) depths.
FIG. 4. Interrelationships between the variables measured during an annual cycle.

MAR = compartment involving active bacteria as determined by $^3$H-autoradiography; auto = automatic counting of active bacteria (Quantimet 720); man. = visual counting of active bacteria; µg C l$^{-1}$ = biomass of active bacteria (automatically measured, Quantimet 720); spot vol. = 'volume' of spots on the X-ray film as calculated from their area (a relative measure of uptake activity); TNB = compartment involving parameters derived from the determination of total bacteria by fluorescence-staining; TNB = total bacteria counts; µg C l$^{-1}$ = total bacterial biomass; Sapro. = colony-forming units (saprophytes); Chloro. = chlorophyll a; DOC = dissolved organic carbon; $K_1 + S_n$ = compartment concerning the parameter $K_1 + S_n$ as calculated from uptake kinetic experiments of the four substrates $^3$H-amino-acid mixture (ASM), $^3$H-leucine (Leu.), $^3$H-glucose and $^3$H-acetate (Ac.); temp. = water temperature; $T_1$ = compartment involving the parameter $T_1$ as calculated from uptake kinetic experiments (see $K_1 + S_n$); $V_m$ = compartment involving the parameter $V_m$ as calculated from uptake kinetic experiments (see $K_1 + S_n$). Arrows and lines indicate that a correlation between the parameters or compartments was found. Black spots on the arrows and lines indicate a correlation at the 99% confidence level, 0 indicates a correlation at the 93% confidence level.
differ from observations made by Albright [18] in the lower Fraser River, its estuary and Georgia Strait. Albright found a good correlation between DOC and heterotrophic activity in Georgia Strait water. On the contrary, the Kiel Bight system seems to be determined by phytoplankton development and temperature. This statement is supported by Gocke’s investigations [19] in the same region, where bacterial activities lagged behind the chlorophyll a development. During the summer, blue green algae may favour temporarily extremely high bacterial numbers and activities. According to Horstmann [20], blue green algae tend to accumulate in the upper water layer when weather is calm, a condition normally occurring in this area during late summer. The results for bacterial numbers in the brackish water are comparable to those of Overbeck [21], obtained from lake water samples: generally less than 1% of the total bacteria numbers could be isolated on nutrient agar plates. According to Jannasch’s chemostat investigations [22], these large bacteria capable of growth on agar plates belong to the type which is normally ‘inactive in natural seawater but surviving’. We found that during periods of sufficient nutrient supply these bacteria may become important in inshore waters. However, also a considerable fraction of well-adapted bacteria must be temporarily inactive (or ‘dormant’, as in Stevenson [16]) especially during the cold season, as could be demonstrated by autoradiography.

ACKNOWLEDGEMENTS

The author wishes to thank M. Rieper (Heligoland) and R. Dawson (Kiel) for their comments on the manuscript.

REFERENCES


DISCUSSION

S. STRACK: You mentioned that in your samples only 41% of the bacteria showed an active metabolism. Is it possible that in samples with seawater containing, for example, a higher concentration of organic substances, a different proportion would be found?

H.-G. HOPPE: The autoradiographic method described makes it possible to measure bacteria actively metabolizing on easily degradable organic substances in the water. The remaining 59% of bacteria which were not detected may be composed of dead or extremely inactive bacteria, specialists and autotrophic bacteria. The average figure of 41% active bacteria was not significantly different in waters with different nutrient concentrations. This finding may be explained by the adaption of the autochthonous bacterial populations to environmental conditions. Only the small group of bacteria growing on nutrient agar plates was found to be strongly affected by the nutrient concentration in the water. We have no information on the situation in waters extremely poor in nutrients.
INCORPORATION OF TRITIUM INTO PLANCTONIC ALGAE IN A CONTINUOUS CULTURE UNDER DYNAMIC CONDITIONS

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Abstract

INCORPORATION OF TRITIUM INTO PLANCTONIC ALGAE IN A CONTINUOUS CULTURE UNDER DYNAMIC CONDITIONS.

For the purpose of modelling the ecologic behaviour of organically bound tritium (OBT) in aquatic food chains under dynamic conditions (i.e. by changing tritium concentrations), a continuous culture of algae was chosen to which tritium was added by a single injection as tritiated water (HTO). The culture was working according to the chemostatic principle where the concentration of cells is in a steady state. Therefore, according to the growth of algae, tritium is incorporated into the organic substance, while in a parallel process HTO and algae are eliminated from the system at the same rate. From these two processes of first-order kinetics, a special function resulted for the concentration process of OBT in the fermenter that is well known in the field of drug kinetics. Initially it increases until it reaches a maximum value where it intersects the elimination curve of HTO, then decreases and asymptotically approaches the time axis — in the same manner as the elimination curve — only at a superior level. A comparison of this theoretically calculated function with the concentration actually found shows that also under dynamic conditions tritium is undergoing discrimination because of isotopic effects up to a ratio of $I = 0.80$. The calculation of the ratios $R = (\text{OBT})/(\text{HTO})$ in the continuous culture by comparing the function for OBT with the elimination curve for HTO shows a linear increase of R-values during the experiment. At maximum tritium concentration in the algae, the ratio becomes greater than one, and at the end of the experiment it reaches a value of about 6. However, by extrapolating to a time of 40 half-lives, when the absolute concentration of HTO has already decreased by a factor of $10^{-12}$, a ratio of about 25 was found. The discrimination enters the estimation of R-values at a constant factor of 0.80.
1. INTRODUCTION

An indication for the ecologic behaviour of organically bound tritium (OBT) in aquatic and terrestrial food chains is the ratio \( R \) of specific activities of organically bound tritium to tritium in the surrounding water, in particular in each respective correlation to the hydrogen content of both phases:

\[
R = \frac{\text{specific activity, organically bound (OBT)}}{\text{specific tritium activity as water (HTO)}}
\]

When interpreting those \( R \)-values determined in environmental samples, it must be considered that normally dynamic conditions exist in the natural ecologic systems, i.e. constantly changing tritium concentrations are usually found.

In order to investigate the ecologic behaviour of organically bound tritium under such dynamic conditions, controlled biological experiments are necessary, accompanied by mathematical models, to detect biokinetic regularities in the behaviour of tritium in natural eco-systems.

For our experiments we used a continuous culture of single cell planctonic algae as the biological system which may be considered as a single element of an aquatic ecological system, i.e. that of the primary producers in rivers or lakes. In a continuous culture (chemostat) the algae grow in a balanced flow basically closely related to normal conditions of growth in natural waters. Here, too, the continuous growth of algae encounters a steady cellular decline and deterioration whereas the concentration of algae cells remains relatively constant disregarding fluctuations of seasonal origin.

In addition, a continuous culture represents a characteristic model for yet another important biological system playing a major role connected with the problem of radiation exposure to man via tritium: the blood circulatory system in man and animals [1]. In this context, a large number of red corpuscles constantly aggregate in the red bone marrow and break down in the liver and spleen, so that the erythrocyte concentration in blood is fairly constant. An extended connection between the blood circulatory system and the test described here reveals itself in the assessment.

2. MATERIALS AND METHODS

In this experiment, we used the unicellular planctonic algae *Scenedesmus quadricauda* (Institute for Water-Soil-Air Hygiene, Berlin (West)). They were cultures in an inorganic nutrient solution under sterile conditions. As the culture vessel, we used a fermenter of 1 litre volume. In this fermenter 732 ml of algae suspension was continuously stirred and illuminated. Four fluorescent lamps
surrounding the fermenter provided illumination of up to about 14000–18000 lx light intensity at the surface of the container. The culture was kept at a temperature of 27°C, and was ventilated with sterile air enriched by 2% of CO₂. Culture growth was controlled by measuring turbidity.

At steady state, the culture was continuously supplied with sterile nutrient solution by a peristaltic pump. The outflow of culture medium (equal to the rate of addition of fresh nutrient) was achieved by an overflow tube. The medium was removed from the container and transferred via a rubber tube into a bottle which was kept in darkness at a temperature of 4°C. From this bottle, samples were taken at intervals of 3, 6 and 12 h consecutively.

The sample algae were separated from the culture medium in a chilled centrifuge. Tritium activity of the medium was directly measured in a liquid scintillation spectrometer. The cells were washed three times with tritium-free water and then freeze dried. After combustion, the activity of organically bound tritium was determined.

To estimate the loss of easily exchangeable tritium for OBT employing this procedure, a simple counter test was made. Tritium-free algae were washed three times in tritiated water, then freeze dried and measured following combustion. Thus, the 'entered' activity could be determined according to the isotope exchange during treatment with tritiated water. It amounted to less than 1% of the activity of the cleansing water [2]. This led to the conclusion that the loss of exchangeable tritium for OBT is negligible.

The hydrogen portion of the algae matter was measured after the method of Pregl. It amounted to 6.0%.

3. RESULTS

3.1. Growth of algae

After the usual lag phase and a relatively short exponential growth phase, the algae propagated at a linear growth rate until the culture was converted into a continuous process at a cellular concentration of about 0.45 g dry substance/litre. With this relatively high concentration of cells for a continuous culture, the specific growth rate \( \mu \) amounted to a value of about \( \mu = 0.04 \text{ h}^{-1} \), suitable for a culture generation time of \( T = 17 \text{ h} \).

To maintain a stable steady state, the dilution rate constant \( D \) of a continuous culture must correlate precisely with the specific growth rate \( \mu \),

\[
\mu = D
\]

so that the cell concentration in the fermenter remains constant. However, during the course of the experiment it was apparent that the growth rate was not
altogether stable. Because of minor oscillation the dilution rate constant had accordingly to be changed three times in order to maintain a constant amount of cells, i.e. the bio-mass in the system. These corrections were taken into consideration when calculating all the theoretical test parameters.

For the entire length of the test the mean growth rate and the dilution rate amounted to

\[ \mu = D = 0.0413\ h^{-1} \]

3.2. Tritium in the aqueous phase

To the continuous algae culture about 5 mCi tritium contained in 2 ml water were added to the culture nutrient by a single injection after being kept about 24 h in a stable steady state. During the process of addition, the in- and outflow was briefly interrupted so that HTO was able to undergo a homogeneous distribution in the fermenter before being eliminated by the outflow as a result of constant dilution. The tritium activity in the fermenter decreased purely exponentially, and consequently that in the outflow, in accordance with a first-order process of (HTO) = (HTO)_o \cdot e^{-D \cdot t}. According to the duplication time t_d of exponential cell growth, the time for half-life T_{elim} is thus calculated as T_{elim} = (Ln2)/D (h) for elimination whereby at any given time 50% of the available substance is eliminated by the fermenter. The measured values of individual samples result in a straight line, applied semilogarithmically as nCi/g, over more than three decimal exponents with those angles caused by the above-mentioned change in the rate of flow (Fig.1).

The following values characterize a single compensating line without angles placed over all measured values:

\[ D_o = 0.0413\ h^{-1} \]

\[ T_{elim} = 16.78\ (h) \]

The initial concentration (HTO)_o of tritium resulted from the point of intersection of the first part of the curve with the y-axis and amounted to 1.08 \times 10^7\ pCi/g.

Consequently, until the end of the test more than 10 half lives later, the activity had decreased to a value of 7.2 \times 10^3\ pCi/g.

3.3. Bound tritium in the algae cell substance (OBT)

In accordance with the growth of algae, tritium is organically bound by photo-synthesis, and the tritium-labelled substance increases exponentially with
the rate of growth. As mentioned before, the latter is numerically exactly identical with the dilution rate constant D, so that the abbreviated formulation k will be used for both constants of speed.

A simplified comprehensive model demonstrates the dry substance of algae growing in a balanced flow as a compartment receiving tritium from the nutrient solution at a calculated invasion constant k₁, and releasing tritium at a calculated elimination constant k₂.

We are not really dealing with a genuine 'entry and exit' process, since the algae dry substance in this context does not represent an identically consistent bio-mass, but is newly developed constantly and eliminated together with the incorporated tritium. However, for purpose of calculation, this model is quite adequate. The following differential equations express tritium concentrations as

\[
\frac{d[B]}{dt} = k \cdot [A] - k \cdot [B]
\]

\[
\frac{d[B]}{dt} = k \cdot [A]_0 \cdot e^{-kt} - k \cdot [B]
\]

According to Bieberbach [3], this equation can be resolved by the following integral:

\[
[B] = e^{-\int_0^t k \cdot dt} [B_0 + \int_0^t k \cdot e^{-kt} \cdot dt]
\]

with the final result of

\[
[B] = A_0 \cdot k \cdot t \cdot e^{-kt}
\]
or else in terms used in the test:

\[
[\text{OBT}] = [\text{HTO}]_0 \cdot D \cdot t \cdot e^{-Dt}
\]

This function describes the special case of a curve playing an important role in drug kinetics as the blood level curve: the curve resulting from two parallel processes of first-order kinetics reflecting in blood the concentration of an orally administered drug that is transported by the invasion constant \(k_1\) via the gastrointestinal tract into blood and from there by the elimination constant \(k_2\) via the kidneys into urine. In 1910 it was first developed by Bateman to calculate the concentration of a daughter element within a radioactive decay series, therefore it is also called the 'Bateman function'. Whereas the equation above represents a special case where both processes occur at the same speed, i.e. \(k_1 = k_2\), the general equation is [4,5]

\[
N_2 = N_0 \cdot \frac{k_1}{k_2 - k_1} \left( e^{-k_1 \cdot t} - e^{-k_2 \cdot t} \right)
\]

It is structured as follows: initial increase to maximum value, intersecting the elimination curve of HTO, the decrease and asymptotic approach of the time axis, in the same manner as the elimination curve only at a superior level.

We receive the following values from discussing the curve: the maximum curve is

\[
t_{\text{max}} = \frac{1}{D}
\]
FIG. 2. Tritium concentrations in the continuous culture (curves calculated in theory and actual concentrations).

and lies exactly at the point intersecting the elimination curve. The maximum value is

$$OBT_{\text{max}} = \frac{(HTO)_0}{e}$$

The point of reflexion is

$$t_{\text{flex}} = \frac{2}{D}$$

Figure 2 shows curves calculated in theory (dotted line) as well as the concentrations actually found for OBT.

On the one hand, the dispersion of measured values renders the unequivocal recognition of the maximum compensating curve impossible, but on the other hand it represents the expected Bateman function, only shifted in the direction of the abscissa by one proportional factor. However, this corresponds with the expected result since our tests [6] and those of other authors [7–10] show a discrimination of isotopic origin in batch cultures when incorporating tritium.
FIG. 3.  R-values in the continuous culture.

As a result of isotopic effects, the ratio I is obtained in the continuous culture by comparing the theoretic and actual function for OBT

$$I = \frac{(OBT)_{\text{actual}}}{(OBT)_{\text{theoretical}}}$$

This represents a curve purposely adjusted for the Bateman function taking ratio I into consideration.

$$(OBT)_{\text{actual}} = ((HTO)_0 \cdot D \cdot e^{-D \cdot t}) \cdot I$$

is also shown in Fig. 2. An adjustment of the curve using the least square fit of the standard deviation resulted in a ratio of

$$I = 0.802$$

Accordingly, a discrimination against tritium of 20% is recognizable in the continuous culture.

3.4. Calculation of R-values in the continuous culture

R-values result from a comparison of the elimination curve for HTO with the function for OBT.
### TABLE I. CALCULATION OF R-VALUES

<table>
<thead>
<tr>
<th>Effective half-life values</th>
<th>Residual specific R-values</th>
<th>Calculated with or without discrimination (I = 0.80)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Activity (%)</td>
<td>Without</td>
</tr>
<tr>
<td>1</td>
<td>50</td>
<td>0.69</td>
</tr>
<tr>
<td>10</td>
<td>0.1</td>
<td>6.96</td>
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<tr>
<td>40</td>
<td>$10^{-10}$</td>
<td>27.63</td>
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</tbody>
</table>

*FIG. 4. Cumulative functions for OBT.*

Both curves have a mutual intersecting point — taking into consideration discrimination or not — from which point on the concentration for OBT is higher than that of HTO, i.e. in this system the R-values become greater than 1 at a certain point of time. Figure 3 shows the R-values against time as measured in the samples. They show a linear increase as early as the time when the R-values are still smaller than 1.

After about 30 h they become greater than 1 until, at the end of the test, about 10 half-lives later, an R-value of R = 5.85 is reached. Since the system's functions for the elimination and for the OBT curve are known, R-values can be calculated for an optional length of time.

Table I contains the calculation of R-values up to 40 half-lives, corresponding to a period of 28 days.
Straight lines resulting from the calculation are also contained in Fig.3. The consideration of \( I = 0.80 \) results in a straight line that corresponds well with the R-values of the samples.

4. DISCUSSION AND CONCLUSIONS

The determination of R-values shows the following obvious result. In spite of discrimination, the ratio R becomes greater than 1 in the culture at a pre-determined point of time and then continues to increase linearly without detectable limit. In addition, the discrimination has the effect of a constant factor by which the values of the theoretical straight lines are shifted in the direction of the X-axis. This result is remarkable since such ratios can easily be found in connection with such concepts as concentration or accumulation, particularly with tritium storage in a compartment against attenuating concentration, similar to iodine stored in the thyroid gland or calcium in bone. Under prevailing dynamic conditions, as in the system 'continuous culture', R-values result from two exponential processes taking a parallel course at the same speed but in opposite directions, as shown in the model.

The expression 'retention' is best fitted for this process where in a system's compartment (during the elimination of tritium from this system) a larger concentration of tritium is retained than in the surrounding area. However, it must be emphasized that this retention is not a consequence of isotopic effects, as Rambeck and Bassham [10] describe, i.e. it is not a 'discriminatory retention'. In the model described, a retention favoured by isotopic effects was not in evidence.

In conclusion, we would like to point out an interesting aspect occurring in the dynamic system when tritium is added periodically. The kinetics in this particular case can be derived from the experiment described.

Figure 4 shows two cumulative functions for OBT originating from tritium doses administered at varying time intervals. The special interest lies in the fact that the occurrence of very different maximum limits depends on the length of the time intervals. Dost [4] developed the curves as blood level curves for a drug administered periodically, yet these are just as suitable for the continuous algae culture described.

REFERENCES

TRITIUM BEHAVIOUR IN AQUATIC PLANTS AND ANIMALS IN A FRESHWATER MARSH ECOSYSTEM

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Abstract

TRITIUM BEHAVIOUR IN AQUATIC PLANTS AND ANIMALS IN A FRESHWATER MARSH ECOSYSTEM.

Ten curies of tritium as tritiated water (HTO) were experimentally added to an enclosed 2-ha Lake Erie marsh on 20 October 1973. Tritium kinetics in selected plants and animals were determined over a one-year period. Tritium levels in the marsh bottom sediment averaged 1.8 times the marsh water levels during this period, but little evidence of tritium concentration above the marsh water tritium levels was observed in the flora and fauna. The unbound tritium : marsh water tritium ratios in smartweed (Polygonum lapathifolium) and pickerelweed (Pontederia cordata) (both emergents) were lower than the same ratio for pondweed (Potamogeton crispus) (a submersent), presumably because of a differential loss of HTO over H2O in the two emergents. There was some evidence of bound tritium buildup in midsummer, particularly in the pondweed, possibly because of increased photosynthetic activity and plant growth at this time, resulting in more tritium being tied up in organic constituents. Tritium uptake into the unbound compartments of crayfish (Procambarus clarkii), carp (Cyprinus carpio) and bluegills (Lepomis macrochirus) was rapid. For crayfish, maximum HTO levels were observed on days 2 and 3 following treatment for muscle and viscera respectively. Unbound HTO in carp muscle peaked in four hours and the level in carp viscera reached a maximum in two days. Maximum levels of unbound HTO in bluegill muscle and viscera were observed on day 1. Unbound HTO in all species decreased following peak levels, paralleling marsh water HTO activity. Tritium uptake into the bound compartments was not as rapid nor were the levels as high as for unbound HTO in the fauna. The peak bound level in crayfish muscle was observed on day 10 (bound : unbound ratio of 0.34) and the maximum level in viscera was noted on day 20 (bound : unbound ratio of 0.23). Bound tritium in carp muscle and viscera reached maximum levels on day 20 (bound : unbound ratios of 0.25 and 0.39 respectively). In bluegills, peaks were reached on days 5 and 7 (bound : unbound ratios of 0.35 and 0.38 for muscle and viscera respectively). Bound tritium in all species decreased following maximum levels.

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INTRODUCTION

The amount of electricity generated from nuclear power plants in the United States of America is planned to increase from 600 MW in 1970 to an estimated 800 000 MW by the year 2000 to help meet the public demand for energy [1]. At present there is a demand for additional conventional nuclear power plants to come on line. Such plants routinely release low-level radioactivity into the environment during normal operations, and the single radionuclide released in the largest quantities is tritium. The environmental burden of tritium from the United States nuclear power industry is projected to increase from 0.3 MCi in 1970 to 600 MCi in 2020 [2].

Tritium is important from a biological/environmental standpoint because relatively large quantities are released into the environment from nuclear power plants (over 90% of released radioactive material is tritium) and associated nuclear fuel reprocessing facilities. Tritium has a long physical half-life, is ubiquitous, and is known to follow protium pathways in biological material. At the present time there is no economical control technology available for reducing environmental releases.

The environmental aspects of tritium have been considered only in the last few years. Jordan and co-workers [3] studied tritium movement in a tropical ecosystem, and concluded that the resident half-time of tritium in the soil was longer than in plants and air, and therefore was the rate-limiting step in movement of tritium through the soil, plants and air. Koranda and Martin [4] obtained similar results in a desert ecosystem. Jordan and co-workers [5] described tritium kinetics in an old field grassland in Illinois. Harrison and Koranda [6] investigated tritium movement in organisms in an experimental freshwater pool and Harrison and co-workers [7] studied its movement in organisms in an experimental marine pool.

One of the most complete studies of environmental tritium at an operating nuclear power plant was conducted by Cohen and Kneip [8]. These investigators defined the relative concentration of tritium in several compartments of an estuarine environment. Measurements were made of tritium in the loose and organically bound water in bottom sediments, rooted aquatic plants, fish, and ambient water. They found significant incorporation and retention of tritium in the bound state in bottom sediment and biota. Tritium expressed in terms of pCi/litre was found to be, on the average, ten times higher in sediment, four times higher in fish and three times higher in rooted aquatic plants than the ambient tritium concentration in water. In some instances, the tritium content of the free or loose water of these components of the ecosystem was also one to two times higher than ambient water.

Jacobs [9], Elwood [10] and Bruner [11] reviewed the literature concerning tritium kinetics in the environment, and Rohwer and Wilcox [12] reviewed the
radiological aspects of environmental tritium. Elwood noted the lack of information available on tritium behaviour in temperate, freshwater ecosystems.

To increase our understanding of tritium behaviour in the environment, we designed a study to investigate tritium kinetics in a temperate freshwater marsh ecosystem under as near natural conditions as possible. This present report is limited to a discussion of plant and animal data from this study. For a discussion of tritium kinetics in the marsh water, sediment, and air over the study area see Refs [13, 14].

METHODS

Study area

Field work for the study was carried out on a 2-ha enclosed Lake Erie marsh, known locally as Navarre Marsh, an area owned jointly by the Toledo Edison and the Cleveland Electric Illuminating Companies. Located in northwestern Ohio, the marsh is about 32 km east of Toledo, at the site of the Davis-Besse Nuclear Power Station. The two utility companies have leased Navarre Marsh to the U.S. Fish and Wildlife Service for management as a wildlife refuge. Human disturbance of the area was minimized throughout this study because of security precautions at the plant site.

The study area, a lentic system, was enclosed by an earthen dike, the integrity of which was maintained throughout the investigation. During the study, the unit contained about 10 000 m³ of water at a mean depth of 50 cm. The water surface area was approximately 20 000 m². For a further description of the study area, and the method of tritium application to the marsh, see Ref. [13].

Ten curies of tritiated water (HTO) were added to the marsh on 29 October 1973, and tritium behaviour in selected aquatic plants and animals was determined over the following one-year period.

Sampling procedure

Aquatic animals sampled in the study were crayfish (Procambarus blandingi), carp (Cyprinus carpio) and bluegill sunfish (Lepomis macrochirus). Three days following tritium application, three 30-in (76.2-cm) square enclosures (wood frame and hardware cloth), one containing crayfish, average weight 28.3 g, one containing bluegills, average weight 7.2 g, and one containing carp, average weight 14.6 g, were placed in the study area from an adjacent uncontaminated marsh unit. Samples of four individuals were taken from each enclosure 0.5, 1, 2, 3, and 4 h following placement in the area. Daily samples then were taken
during the first week, followed by weekly sampling through March 1974. After this date, samples were collected twice each month.

Sample material generally was maintained in the enclosures with the aid of 20 Herter's live bait traps (Herter's Inc., Waseca, Minnesota 56093) systematically placed in the marsh. As organisms were trapped they were placed in the appropriate enclosure, care being taken not to overpopulate any one enclosure. Immediately following collection, sampled animals were placed in plastic bags containing a small amount of marsh water, frozen in liquid nitrogen, and transported to the laboratory where they were kept frozen (−15°C) until analysed.

Weekly sampling of marsh vegetation was initiated in June 1974. Leaf samples were collected from nodding smartweed (Polygonum lapathifolium), pickerelweed (Pontederia cordata) and curly-leaf pondweed (Potamogeton crispus). Samples were placed in airtight containers, transferred in a dark environment on dry ice to the laboratory, and stored in a freezer (−15°C) until analysed.
Radioassay

For radioassay, samples were lyophilized by the technique of Stewart and co-workers [15] to separate free HTO (unbound tritium) from tritium incorporated into organic material (bound tritium). A 0.5 ml aliquot of the extracted sample water was added to 15 ml of Insta-gel\textsuperscript{®} liquid scintillation cocktail (Packard Instrument Company, Downers Grove, Illinois 60515), in a low-potassium scintillation vial. A 0.1 g subsample of the freeze-dried sample material was combusted in a tritium-oxidizer (Model 300 Tri-Carb\textsuperscript{®}, Packard Instrument Company), and the water of combustion was collected in a low-potassium liquid scintillation vial to which 15 ml of Insta-gel\textsuperscript{®} were added. Samples prepared in the above manner were counted for tritium activity in a Model 3320 Automatic Tri-Carb\textsuperscript{®} Liquid Scintillation Spectrometer (Packard Instrument Company).

All reported activities are those calculated for the day the sample was collected from the marsh. Sample activity then was expressed on a per millilitre basis (for unbounded tritium) or a per gram dry weight basis (for bound tritium). Conversion to a per gram hydrogen (g H) basis for unbound HTO was accomplished by dividing the calculated activity per millilitre of water by 11.1\% (wt\% of hydrogen in water). Conversion to a per gram H basis for bound tritium of plants and animals was accomplished by dividing the calculated activity per gram dry weight by 5.2 and 7.5\% respectively (assumed wt\% of hydrogen in the sample material).

RESULTS AND DISCUSSION

Plants

Unbound and bound tritium levels in the three plant species generally followed the marsh water tritium level (Fig.1). The ratio of unbound tritium in plants to marsh water tritium was significantly (p < 0.01) less than unity for all three plant species as determined by the sign test [16]. In only one case did the ratio exceed unity; a value of 1.03 for pondweed on day 262. These results indicate no concentration of tritium in the water of the plants relative to the marsh water.

There is a basic difference among the species of Fig.1 in terms of the ratio of unbound to marsh water tritium. The largest ratio of unbound : marsh water tritium was observed in curly-leaf pondweed. A Friedman's rank sum test [16] indicated there were significant differences among these species in terms of this ratio (S = 19.5, p < 0.001). The associated multiple comparisons procedure (which is based on an additional probability statement) indicated a significant difference between smartweed (median 0.33) and pondweed (median 0.675),
and between pickerelweed (median 0.375) and pondweed (p = 0.012 for both comparisons). Based on the limited probability values available for this procedure, no difference could be detected between smartweed and pickerelweed (p < 0.038). We think there is a reasonable explanation for the observed differences between the two emergents (smartweed and pickerelweed) and the submergent (pondweed) in terms of the unbound tritium : marsh water tritium ratio. Horton and co-workers [17] experimentally determined that HTO is lost differentially over H_2O from a body of water to the atmosphere if the relative humidity is greater than 8%, because atmospheric moisture is virtually free of HTO. The differential loss of unbound HTO from the two emergents could explain the lower unbound tritium : water marsh tritium ratios observed in these species. The pondweed, not directly exposed to the atmosphere, might be expected to show a higher unbound tritium : marsh water tritium ratio, which it does.

We tested the June and July data (days 215–275) against the August and September data (days 276–337) for each species with a Wilcoxon rank sum test [16] to determine if there was a trend in the ratio of unbound to marsh water tritium with growing season. The two emergents strongly indicated lower unbound tritium to marsh water ratios late in the season (pickerelweed, p = 0.018; smartweed, p = 0.001) whereas pondweed showed no change (p = 0.155). One explanation for this observation is that the emergents gradually dried out as the season progressed, resulting in lower plant water volumes (thus greater surface area to volume ratios for the plant water pools). Pickerelweed went from an average moisture content of 82.2% (June-July) to 79.8% (August-September) and smartweed from 77.3% to 73.9% (both changes significant at p < 0.005). Moisture content in the submergent (pondweed) increased slightly (83.3% for June-July to 84.5% for August-September), but this change was not considered significant (p > 0.05). We suggest that a greater surface area to volume ratio in the two emergents later in the season may have resulted in an increased differential loss of tritium at this time in these species.

There is some evidence of bound tritium buildup in midsummer, particularly in the pondweed. Bound tritium levels peaked in the latter part of July, and then dropped, resulting in no tritium concentration above the marsh water tritium levels (Fig.1). The reason for this buildup in midsummer may be due to increased photosynthetic activity and plant growth at this time, resulting in more tritium being tied up in organic constituents. Harrison and Koranda [6] pointed out that, because the source of hydrogen for aquatic plants in a tritiated water medium is water, bound tritium levels may be expected to rise as photosynthetic activity increases. Bound tritium that had been incorporated into organic molecules during the rapid plant growth stage of early summer exchanged quite readily following cessation of rapid plant growth (Fig.1). Cohen and Kneip [8] observed a buildup of bound tritium in Potamogeton perfoliatus in late summer and early autumn, with a peak in the middle of September in their estuarine
study area. These investigators found that the tritium peak occurred at the
time of the year when the fresh water flow was at a minimum and the salt water
intrusion moved upstream. As the fresh water flow increased, the activity in
the tissue appeared to decrease. We feel that a good understanding of bound
tritium kinetics must await detailed biochemical studies. Weston [18] pointed
out there is a lack of data on this subject.

Tritium was applied to the marsh water approximately seven months before
the growing season and vegetation was exposed to tritium in both the marsh
water and bottom sediment throughout the growing season in 1974. Unbound
tritium levels in the bottom sediment averaged 1.8 times the marsh water levels
during this period. We attribute this concentration factor to the slower turnover
of tritium from sediment to water compared with turnover from the water to the
atmosphere. It is quite significant, we think, that this concentration of
tritium did not show up in the vegetation. This deviates from the findings of
Cohen and Kneip reported earlier. The concentration factors observed by these
investigators may possibly be explained by changes in the tritium content of
the environmental medium and different turnover times for tritium in different
components. Higher tritium levels in sediment and vegetation were observed by
these investigators in the Hudson River below the Indian Point Nuclear Power
Station following tritium release from the plant. Tritium turnover in the water
could be attributed to loss to the atmosphere and to the flushing capacity of the
river. This might well be expected to be faster than loss from other aquatic
components of this system, resulting in concentration of tritium. The past
history of the two study areas also may have influenced the results of the studies.
The Indian Point Nuclear Power Station has been periodically releasing tritium
into the Hudson River since it began operating in 1962. In the present study
there was a single input of tritium into an enclosed marsh.

Harrison and Koranda [6] maintained cattails (Typha angustifolia) (an
emergent) in a tritiated experimental pool for nine months after which time
samples of leaves (outer and inner), rhizomes, and roots were analysed for
tritium content. At the time of sampling the unbound tritium : pool water HTO
ratio did not approach unity in any of the plant parts. Their data indicated a
trend toward a smaller ratio from the plant roots to the outer leaf blades (from
0.67 to 0.58). This they attributed to exchange with atmospheric water by the
leaves. When the plants were supplied with pre-dried air, the unbound HTO of
the leaves approached that of the solution applied to the roots. The data from
the present study, collected over one growing season, concur with these results.

Animals

Uptake and loss of tritium over an 11-month period in crayfish, carp and
bluegill sunfish were determined. Tritium uptake into the unbound compart-
ments of both muscle and viscera tissue was rapid for all species studied
FIG. 2. Uptake and loss of tritium in crayfish at Navarre Marsh in 1973–74. The dashed line is the observed marsh water tritium concentration, and the data points for crayfish represent means of 1–6 samples. Note change of scale on the abscissa.

(Figs 2–4). For crayfish, maximum HTO levels were observed on days 2 and 3 for muscle and viscera respectively. Unbound HTO in carp muscle peaked in 4 h, whereas levels in carp viscera reached a maximum in 2 d. Maximum levels of unbound HTO in bluegill muscle and viscera were observed on day 1. Unbound HTO in all species decreased following peak levels, paralleling marsh water HTO activity.

The rapid uptake of tritium into unbound compartments of the organisms was expected, and agrees with work reported by others [6, 7, 19–21]. Theoretically, the concentration of body-water tritium in aquatic organisms should (under equilibrium conditions) equal the concentration of environmental water tritium. However, in the present study, the ratio of unbound tritium in the animals to marsh water tritium was significantly less than unity (p < 0.01) as determined by the sign test [16]. Because of the differential loss of HTO over H₂O to the atmosphere when the relative humidity is greater than 8% [17], some body-water tritium would be expected to be lost during sample preparation. An effort was made to eliminate tritium loss from sample material during
storage in the freezer, but slight loss also may have occurred there. These losses would account for ratios less than unity under equilibrium conditions. The rapid uptake of HTO into the body-water compartments of all organisms most probably can be attributed to the very close association between the environmental medium and the internal fluid content of the organism at the gills. The fact that unbound HTO in the organisms closely followed the marsh water HTO levels (Figs 2–4) also indicates the association is very close and exchange is rapid.

The crayfish and bluegill bound tritium in muscle tissue was reported earlier [13]. However, subsequent to this report sample preparation for tritium assay was changed in our laboratory. The lyophilization technique used in the present study is the more standard method for differentiating between bound and unbound tritium, and data obtained from this technique are more directly comparable with data reported by other investigators employing similar techniques. In addition, the hydrogen content of the animal tissue is assumed to be 7.5% in the present study. This value was changed from 5.5% [13] to be
FIG. 4. Uptake and loss of tritium in bluegills at Navarre Marsh in 1973–74. The dashed line is the observed marsh water tritium concentration, and the data points for bluegills represent means of 1–4 samples. Note change of scale on the abscissa.

more in line with a reported value of 7.4% for crayfish (Astacus sp.) and 7.6% for goldfish (Carassius auratus) [6]. Therefore, the crayfish and bluegill muscle data are included here (Figs 2, 4) for comparison with the rest of the animal data. The effect of the two changes discussed above is to reduce the overall magnitudes of bound tritium levels from those previously reported [13]. The general pattern in the data remains the same.

Tritium uptake into the bound compartments was not as rapid nor were the levels as high as for unbound HTO in any of the species (Figs 2–4). Peak bound levels in crayfish muscle were observed on day 10 (bound : unbound ratio of 0.34) and maximum levels in viscera were noted on day 20 (bound : unbound ratio of 0.23). Bound tritium in carp muscle and viscera reached maximum levels on day 20 (bound : unbound ratios of 0.25 and 0.39 respectively). In bluegills, peaks were reached on days 5 and 7 (bound : unbound ratios of 0.35 and 0.38) for muscle and viscera respectively.

As can be seen, tritium was readily incorporated into organic material of the organisms studied. The similarity of the uptake and loss patterns in the two different tissues for all organisms is striking. Under somewhat more con-
trolled circumstances, Tucker and Harrison [22] found that clam muscle and viscera tissue were essentially the same in their rates of decrease in specific activity of tritium with time in what they termed a rapidly exchangeable bound compartment.

The relatively low uptake of bound tritium in all species may be an indication of lack of hydrogen exchange in this compartment. Certainly, with unbound levels being much higher, the chance for further exchange was present. We did not measure the proportion of the organisms' bound tritium pools that consisted of exchangeable hydrogen sites. However, the data indicate that not all organic hydrogen is exchangeable. This is indicated by the fact that bound tritium levels remained low and decreased following peaks even though marsh water tritium levels remained high. If we assume all organic hydrogen sites are exchangeable then tracer kinetic theory [23] would predict that bound tritium would continue to increase until it reached equilibrium with unbound and marsh water tritium, and then all three would decrease. Non-exchangeability of some of the organic-bound hydrogen also is supported by the literature. Siri and Evers [24] stated that exchangeable tissue-bound hydrogen is 15–30% of the total tissue hydrogen, variability being due to the amount of fatty tissue which has a low level of exchangeable hydrogen. Tucker and Harrison [22] stated that the exchangeable portions of tissue hydrogen in clam (Mya arenaria) viscera and muscle were 26 and 22% respectively. Hatch and Mazrimas [25] presented similar values in reviewing other work including their own with laboratory mice and kangaroo rats. If we apply these values to our data we see that at the peaks most tritium exchange may well have taken place. Harrison and co-workers [7] observed similar results in the longjaw mudsucker (Gliclichthyis mirabilis). Maximum bound tritium levels in muscle were observed in about 30 d, after which time levels declined even though unbound tritium remained considerably higher.

Following peak levels, bound tritium in all species and tissues decreased, obviously at the influence of the marsh water tritium concentration. Our conclusion is similar to that of Hatch and Mazrimas [25] that bound tritium in tissues of animals exposed to HTO over substantial periods of time has a specific activity of one-quarter to one-third that of tritium in tissue-free water. In most other environmental tritium studies a lack of tritium concentration has been observed [6, 7, 20, 21, 26–28].

CONCLUSION

This study was conducted over a one-year period under as near natural conditions as possible. The data concur with the findings of other investigators working under more controlled conditions consisting of small experimental pools [6, 7, 22] and freshwater microcosms [19, 26].
No significant concentration of tritium was observed. The small value reported for sediment was attributed to a slower exchange rate between sediment and marsh water compared with the rate between marsh water and the atmosphere. Our data indicate that of the total amount of tritium applied to the marsh, practically all eventually ended up in the atmosphere, where global distribution occurs [9]. Thus, though tritium loss from the flora and fauna was rapid, essentially following loss from the marsh water, practically all of it remains in the biosphere for possible further incorporation into biological material, the amount lost from the global system being due to the physical half-life of the isotope.

ACKNOWLEDGEMENT

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REFERENCES

DISCUSSION

Y. UENO: You show in your Fig. 4 that in bluegill (Lepomis macrochirus) the bound \(^3\)H level is higher than the marsh water \(^3\)H level at about 250 days, but you say that this phenomenon is not observed in other fish. Is the phenomenon therefore biologically specific to bluegill?

L.W. ADAMS: I have not attached any great significance to these levels in bluegill, primarily because they did not persist for very long. One could speculate, however, that since the bluegill is carnivorous (and thus on a higher trophic level than the herbivorous carp), and since the period involved is late spring-early summer, increased food consumption (consisting of a tritiated food source) and growth by the fish may have resulted in more tritium being retained in the bound state. However, without more data on the subject I would not ascribe too much importance to the observation at present.

S. STRACK: In your Fig. 1 showing the tritium loss from smartweed and pickerelweed there is a relatively large difference between the tritium concentrations in the marsh water and in the unbound fraction. I should have expected a rapid exchange between these two compartments, and I wonder how you explain the difference.

L.W. ADAMS: First of all I think it important to point out the difference in unbound tritium between the two emergents (smartweed and pickerelweed) and the submergent (pondweed) of Fig. 1. You will note that unbound tritium in the pondweed more closely approximates the marsh water tritium than does the unbound tritium of either smartweed or pickerelweed. I believe the reason for this to be that the pondweed remained in direct contact with the water medium, which facilitated exchange between the two compartments. The smartweed and pickerelweed leaf samples were taken from the plants above the marsh water surface. I think there was a differential loss of HTO compared with \(\text{H}_2\text{O}\) from these leaves, which resulted in lower unbound tritium levels. I did not sample underwater parts of the emergents, but from the work of Harrison and Koranda with cattails, which is cited in the paper, one might expect the ratio of unbound to marsh water tritium to be higher beneath the water surface.

J.A. HETHERINGTON: Can you amplify your remarks concerning the uptake of tritium by sediments, especially in regard to the pre-measurement treatment you gave the sediments? Were you measuring both the solid and interstitial water phases together, or if you were measuring them separately, did the solid phase still contain the bound water and the water associated with the interlattice spaces some days later?

L.W. ADAMS: A discussion of tritium behaviour in the bottom sediment was presented at the Fourth Radioecology symposium (see citation in the paper), and so I have not discussed it in detail here. Approximately 92% of the
tritium in the sediment was unbound and thus readily available for uptake by
the plants. The remaining 8% was classified as bound tritium. This was tritium
that remained following a sample drying process equivalent to 24 hours in an
oven at 100°C.

L. FARGES: In the section of your paper entitled "Sampling procedure"
you state the number of individuals and the sampling time-table. Could you
justify this sampling procedure for us?

L.W. ADAMS: I wanted a sample size large enough to yield a meaningful
estimate of variability, and so in most cases a sample size of four was taken.
On the other hand I recognized that a large number of samples would be generated
from the study, and so from the practical standpoint of getting samples prepared
and analysed in a reasonable time I did not want the sample size to be too large.

I sampled at shorter time intervals early in the experiment because, judging
from work performed by other investigators, I expected a rapid uptake of
unbound tritium in the organisms followed by a slower uptake of bound tritium,
both of which I wanted to measure. The time interval was lengthened to give
semi-monthly sampling later in the experiment, because no rapid changes were
expected in tritium dynamics then. Again, from a practical standpoint, this
helped keep the number of samples to be processed within acceptable limits.

R. KIRCHMANN (Chairman): I understand that you followed the tritium
content in the ecosystem concerned for one year, ending in 1974. Have you any
more recent data on the residence time of organic tritium in your system?

L.W. ADAMS: No, I have no more recent data. When I terminated the
experiment, tritium levels in the biota, particularly those of bound tritium, were
so low that owing to natural variation they could not be distinguished signifi-
cantly from the levels in control organisms. Primarily for this reason I did not
sample further.

J. DELMAS: Have you observed any variation in the activity of tritium
bound to organic matter, as between the various links of the food chain?

L.W. ADAMS: I observed no differences that I would attribute to trophic
level effects. The point that in my opinion emerges most clearly from this study
is the similarity of tritium behaviour in the different species.
DISTRIBUTION OF TRITIUM IN A CHRONICALLY CONTAMINATED LAKE*

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Abstract

DISTRIBUTION OF TRITIUM IN A CHRONICALLY CONTAMINATED LAKE.
White Oak Lake located on the U.S. Department of Energy’s Oak Ridge Reservation receives a continuous input of tritium from operating facilities and waste disposal operations at the Oak Ridge National Laboratory. The purpose of this paper was (1) to determine the distribution and concentration of tritium in an aquatic environment which has received releases of tritium significantly greater than expected releases from nuclear power plants, and (2) to determine the effect of fluctuating tritium concentrations in ambient water on the concentration of tritium in fish. Aquatic biota from White Oak Lake were analysed for tissue water tritium and tissue bound tritium. Except for one plant species, the ratio of tissue water tritium to lake water tritium ranged from 0.80 to 1.02. The tissue water tritium in Gambusia affinis, the mosquito fish, followed closely the significant changes in tritium concentration in lake water. The turnover of tissue water tritium was very rapid; Gambusia from White Oak Lake eliminated 50% of their tissue water tritium in 14 minutes. The ratio of the specific activity of the tissue bound tritium to the specific activity of the lake water was greatest for the larger species of fish but never exceeded unity. The radiation dose to man from tritium which could be acquired through the aquatic food chain was relatively small when compared with other pathways. The whole body dose to a hypothetical person taking in concentrations of tritium measured in White Oak Lake was 1.8 mrem yearly from eating fish and 10.0 mrem yearly from drinking water.

1. INTRODUCTION

Tritium is considered one of the least hazardous of the radionuclides and is routinely released in the liquid effluents of nuclear power plants and nuclear fuel cycle facilities [1]. Increased environmental releases of tritium are anticipated as the number of nuclear power plants increase. Another potential source of environmental tritium is the development of fusion energy. Under normal operations tritium will be the major source of radioactivity in fusion reactors [2]. Because tritium releases to the

TABLE I. MONTHLY DISCHARGE OF TRITIUM FROM WHITE OAK LAKE FOR 1977

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</table>

...environment are expected to increase, it is important to have site specific data for dose calculations to man. The purpose of this paper is (1) to report on the concentration and distribution of tritium in an aquatic environment which has received a continuous input of tritium significantly greater than the expected releases in the effluents of nuclear power plants, and (2) to determine the effect of fluctuating tritium concentrations in lake water on the tritium concentration in fish.

2. MATERIALS AND METHODS

2.1. Site description

White Oak Lake, a 10.5 hectare impoundment, located on the U.S. Department of Energy's Oak Ridge Reservation, serves as a final settling basin for low-level radioactive effluents from the Oak Ridge National Laboratory. The lake receives a continuous input of tritium from operating facilities and waste disposal operations. Tritium concentrations in White Oak Lake fluctuate with the quantity of tritium entering the lake from White Oak Creek and with changes in the water level due to runoff from rainfall.
Within the lake the concentration of tritium can vary considerably. Sampling in the lake at twelve different locations showed that after a heavy rainfall the concentration of tritium was uniform. However, during a low flow condition, the concentrations of tritium varied from 40 to 600 pCi/ml in samples from adjacent stations. The highest concentrations of tritium and the greatest variation from station to station were detected in the upper end of the lake where White Oak Creek enters the lake. The concentrations of tritium were the most uniform from sampling stations in the vicinity of the dam at the lower end of the impoundment.

The water entering and discharging from White Oak Lake is monitored by continuous, proportional water samplers. Samples are collected weekly and assayed for tritium by the Industrial Safety and Applied Health Physics Division at the Oak Ridge National Laboratory. Annual releases of tritium from White Oak Lake from 1964 to 1976 ranged from 1161 Ci in 1968 to 15 040 Ci in 1973, with an average of 8488 Ci/yr [3]. The monthly fluctuations of the total curies released over White Oak Dam and the concentrations of tritium in the water for 1977 are shown in Table 1.

The average flow over White Oak Dam is 12.5 cfs. Tritium concentration decreases rapidly after the water flows over the dam and is diluted by creek water below the dam. Additional dilution occurs when White Oak Creek enters the Clinch River. A series of 36 water samples taken in June 1974 directly below White Oak Dam and at intervals below the dam to the Clinch River showed that the concentration of tritium decreased from 525 pCi/ml at the dam to 57 pCi/ml at the mouth of the creek where it is accessible to the public.

2.2. Sampling and analysis

Aquatic biota was collected from White Oak Lake and analyzed for tissue water tritium (intracellular and extracellular water removed by freeze-drying) and tissue-bound tritium (tritium remaining in the dried tissue). Fish were collected either by netting or electroshocking, aquatic invertebrates by netting, and aquatic plants and sediments by grab sampling. All biota was maintained in ambient lake water until freeze-drying some two to three hours later.

The tissue water was extracted from whole organisms, except for the larger fish and cattail. Muscle tissue was removed from the larger fish; stem and rhizomes sections were taken from the cattails for freeze-drying. Dried samples were oxidized with a Packard tritium-carbon oxidizer and tissue-bound tritium was collected and counted by standard liquid scintillation counting techniques. Tissue water tritium and lake water which had been filtered through a 0.45μm millipore filter were counted by the same technique.

Sediment samples were freeze-dried to remove the interstitial water, capillary water and some water of hydration. Samples of the dried sediment were oxidized and the bound tritium collected. Subsamples of the dried sediment were heated to 980°C to determine the organic fraction of the sediment.
FIG. 1. Comparison of the concentrations of tritium in the tissue water of Gambusia affinis from White Oak Lake with the concentrations of tritium in lake water.

The uptake of tissue water tritium and tissue-bound tritium in Gambusia was determined by exposing fish to tritiated water at a concentration of 0.1 μCi/ml. Gambusia were sacrificed at various intervals for 17 days and the amount of tissue water tritium and tissue bound tritium was determined by the previously described techniques.

3. RESULTS AND DISCUSSION

3.1. Uptake and elimination of tritium by Gambusia

Tritium concentrations in the tissue water of Gambusia in White Oak Lake closely paralleled the concentrations of tritium in the lake water, Fig. 1. Lake water and ten Gambusia were collected at the same location in White Oak Lake at two or three day intervals for a period of 4 weeks and analyzed for tritium. An analysis of variance showed that concentrations of tritium in both the tissue water and lake water varied significantly with time. The concentration of tritium in the tissue water was approximately the same as the tritium concentration in ambient lake water. The results are presented in terms of the specific activity of tritium in whole body freeze-dried Gambusia and the specific activity of tritium in water.

Other species of fish exchange tissue water tritium with environmental water very rapidly [4, 5, 6]. Tissue water tritium of goldfish (Carassius auratus) introduced into a 2000 gal pool containing tritiated water quickly reached the level of the pool water and decreased at about the same rate as the pool water [4].

Tissue water tritium is rapidly eliminated by Gambusia. Gambusia collected from White Oak Lake and placed in nontritiated water eliminated

\[ \text{eq1} \]
FIG. 2. The uptake of tissue water tritium and tissue-bound tritium by Gambusia exposed to tritiated water.

50 percent of the tissue water tritium in 14 minutes. Ten fish were sacrificed for tritium analysis at 15 minutes intervals for 60 minutes and at varying intervals thereafter. After 165 minutes the concentration of tritium decreased to 1.3 percent of initial value. Elwood [5] found that the tissue water tritium of goldfish from White Oak Lake was eliminated at two exponential rates. The initial fast component, representing 96 percent of the initial level of tissue water tritium, had a biological half-life of 0.2 hr while a slower component, which made up the remaining 4 percent of tritium in the tissue water, had a biological half-life of 0.9 hr.

The uptake of tritium by the tissue water of Gambusia was relatively fast when compared to the uptake of tissue-bound tritium, Fig. 2. Ten Gambusia which had been exposed to tritiated water at a concentration of 0.1 μCi/ml were sacrificed at various times and analyzed for tissue water tritium and tissue bound tritium. Thirty minutes after exposure the concentration of tritium in the tissue water of Gambusia was approximately 73 percent of the aquarium water. After 1.5 hr the concentration had reached approximately the same level as the concentration of tritium in the aquarium water, where it remained for the duration of the experiment. A rapid uptake of tritium by the tissue water was observed for catfish (Ictalurus punctatus) and bluegill (Lepomis macrochirus) [6]. Approximately 30 minutes were required for the tissue water in catfish to reach one-half their maximum burden while 90 minutes were required for bluegill to reach one-half their maximum burden. The rapid uptake and elimination of tritium by Gambusia account for the close correlation of tritium concentrations in the tissue water and in White Oak Lake water.

The uptake of tissue bound tritium in Gambusia increased gradually. After 15 days the concentration of tritium in Gambusia reached a level of approximately 15 percent of the tritium in the aquarium water, Fig. 2. Patzer et al. [7] reported that the consumption of food grown in tritiated water was necessary for Gambusia to attain a ratio greater than 0.50 -- a ratio which compared the specific activity of freeze-dried fish to the
<table>
<thead>
<tr>
<th>Biota</th>
<th>Lake Water pCi/ml</th>
<th>Tissue Water pCi/ml</th>
<th>Tissue Bound pCi/gm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Algae (Cladophora)</td>
<td>609.7 ± 2.6</td>
<td>609.6 ± 6.6</td>
<td>78.9 ± 11.6</td>
</tr>
<tr>
<td>Duckweed (Spirodea polyrhiza)</td>
<td>407.7 ± 8.3</td>
<td>413.0 ± 7.5</td>
<td>95.3 ± 14.1</td>
</tr>
<tr>
<td>Pondweed (Potamogeton foliosus)</td>
<td>423.5 ± 9.9</td>
<td>422.9 ± 2.4</td>
<td>133.4 ± 20.4</td>
</tr>
<tr>
<td>Cattail (Typha latifolia L.)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>stem</td>
<td>646.4 ± 8.3</td>
<td>361.1 ± 4.1</td>
<td>78.6 ± 7.6</td>
</tr>
<tr>
<td>rhizome</td>
<td>646.4 ± 8.3</td>
<td>399.3 ± 2.5</td>
<td>75.4 ± 7.5</td>
</tr>
<tr>
<td>Dragonfly (Plathemis lydia)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>naiad</td>
<td>646.4 ± 8.3</td>
<td>527.1 ± 5.2</td>
<td>138.6 ± 39.0</td>
</tr>
<tr>
<td>Snail (Physa heterostropha)</td>
<td>646.4 ± 8.3</td>
<td>555.7 ± 7.2</td>
<td>76.0 ± 29.6</td>
</tr>
<tr>
<td>Mosquito fish (Gambusia affinis)</td>
<td>646.4 ± 8.3</td>
<td>555.3 ± 10.4</td>
<td>186.3 ± 14.3</td>
</tr>
<tr>
<td>Bluegill (Lepomis macrochirus)</td>
<td>403.2 ± 10.3</td>
<td>409.5 ± 6.7</td>
<td>187.9 ± 18.3</td>
</tr>
<tr>
<td>muscle</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Largemouth bass (Micropterus salmoides)</td>
<td>403.2 ± 10.3</td>
<td>412.6 ± 7.0</td>
<td>166.2 ± 13.9</td>
</tr>
</tbody>
</table>

Specific activity of environmental water. Gambusia exposed to tritiated water and fed food grown in tritiated water attained ratios ranging from 0.50 to 0.90 [7].

In Gambusia from four different locations in White Oak Lake where all organisms are exposed to fluctuating concentrations of tritium, the ratio of tissue-bound tritium to lake water tritium ranged from 0.25 to 0.54. Gambusia collected near the dam where the concentration of tritium fluctuated the least, had the highest ratio, while the lowest ratio was found in the upper end of the lake, where higher concentrations of tritium are found but more fluctuating conditions exist. Continuous changes of tritium concentrations in the lake water could result in a ratio of greater than 1.0 for tissue-bound tritium, if collections were made when the concentration of tritium in the lake water was at the lowest level. However, the fluctuation of tritium in the lake water and the relatively
TABLE III. CONCENTRATION RATIO* OF TISSUE WATER TRITIUM
AND TISSUE-BOUND TRITIUM IN BIOTA FROM WHITE OAK LAKE

<table>
<thead>
<tr>
<th>Biota</th>
<th>Tissue Water</th>
<th>Bound Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Algae (Cladophora)</td>
<td>1.000</td>
<td>0.381</td>
</tr>
<tr>
<td>Cattail (Typha latifolia)</td>
<td>0.618</td>
<td>0.251</td>
</tr>
<tr>
<td>Mosquito fish (Gambusia affinis)</td>
<td>1.057</td>
<td>0.536</td>
</tr>
<tr>
<td>Bluegill (Lepomis macrochirus)</td>
<td>1.016</td>
<td>0.685</td>
</tr>
<tr>
<td>Largemouth bass (Microchirus salmodies)</td>
<td>1.023</td>
<td>0.606</td>
</tr>
</tbody>
</table>

* Tritium specific activity in tissue water and in dry tissue divided by that of lake water.

short biological half-life of the greatest percentage of the tissue-bound tritium [7] prevented Gambusia and other food chain organisms from reaching equilibrium with tritium in the lake water.

3.2. Concentration of tritium in aquatic biota from White Oak Lake

Concentrations of tritium in tissue water and tissue-bound tritium are listed for several species of aquatic biota, Table II. With the exception of cattail the ratio of pCi/ml tissue water to pCi/ml lake water ranged from 0.80 to 1.02. Bluegill and largemouth bass, which had the highest ratios, were collected over a much greater area than were the other organisms. The ratio for the stems and rhizomes of cattails was approximately 0.60 which is similar to the results obtained by Harrison and Koranda [4] when cattails were grown in a large pool containing tritiated water. Since only the roots of the cattail were submerged in tritiated water the tissue water of the plant never reached equilibrium, because of the exchange of tritium with the atmosphere by the leaves of the plants. This phenomenon has been observed by other investigators [8,9].

The accumulations of tissue-bound tritium in biota from White Oak Lake are shown in Table III. The results are presented to show the ratio between the specific activity of tritium in freeze-dried tissue and that in lake water. The ratio indicates the degree of tritiation in the biota in White Oak Lake. A ratio greater than unity would indicate concentration of tritium within organic constituents of the biota relative to the lake water. The highest ratios were observed in freeze-dried tissues of
bluegill and largemouth bass. These fish were collected in the vicinity of the dam where the concentration of tritium in lake water is the most uniform and the tissue-bound tritium in food organisms should be nearer to equilibrium with the lake water. Consumption of food organisms grown in a tritiated environment increases the specific activity in the tissue-bound tritium of fish [7] and could account for the higher concentrations of tritium in the tissues of bluegill and large mouth bass. It is also possible that a portion of the tissue-bound tritium, which is nonexchangeable or turns over very slowly [5,7], represents higher concentrations of tritium that existed in the lake water in the past, Table I. Some of the larger fish were more than three years of age; however a comparison of concentrations of tissue-bound tritium with age showed no significant difference.

3.3. Tritium in sediments

The bound tritium in sediments from White Oak Lake was determined by removing water from the sediments by freeze-drying. Water removed by freeze-drying would include interstitial water, capillary water and some water of hydration. Samples of the dried sediments were then oxidized to collect the bound tritium. The concentration of tritium in the overlying water was 647.6 pCi/ml compared to 449.5 ± 7.2 pCi/ml in the water removed by freeze-drying. The changes of tritium concentration in the lake water and the rate of exchange between sediment water and overlying water could account for the difference in concentration.

The dried sediment contained 17.0 ± 10.2 pCi/gm of tritium. Approximately 11 percent of the dried White Oak Lake sediment was organic material. Assuming that all tritium in the sediment was organically bound, the concentration of tritium in the sediment was within the range of tritium concentrations observed in the tissue-bound tritium of plants and animals in White Oak Lake, Table II. Cohen and Kneip [10] observed an accumulation of tritium in sediments of the Hudson River. The bound tritium and tritium in sediment water contained significantly greater concentrations of tritium than did the ambient water. These higher concentrations were attributed to tritium levels which had existed previously and had permitted nonexchangeable sites to be labeled when higher tritium concentrations existed in the Hudson River.

3.4. Radiation dose to man

A radiation dose to man was calculated for the tritium concentrations in White Oak Lake. The dose was calculated by using the INREM computer code [11] and concentrations of tritium which existed in the lower end of White Oak Lake when the fish samples were collected. Assuming a concentration factor of one for fish [12] and that man eats 20 gm of fish per day, the dose from eating fish would be 1.8 mrem/yr. Using the concentrations of tritium that were measured in fish from White Oak Lake, the estimated dose was 1.1 mrem/yr. The calculated radiation dose for drinking tritiated water from White Oak Lake (1.2 l/day/yr) was 10 mrem/yr. Thus, the potential dose from drinking tritiated water would be more significant than that acquired through the aquatic food chain.
4. CONCLUSIONS

In an aquatic environment where the concentration of tritium may fluctuate from day to day, the concentration of tissue water tritium of most aquatic organisms will follow closely the concentration of tritium in the ambient water. The ratio of tissue water tritium to lake water tritium in White Oak Lake organisms, except for one plant species, ranged from 0.80 to 1.02. The tissue water tritium of Gambusia affinis, the mosquito fish, remained at approximately the same concentration as the lake water during a four week period and followed closely significant changes in the tritium concentration in the lake water. The turnover of tritium in the tissue water of Gambusia is very rapid. Gambusia from White Oak Lake lost 50 percent of their tissue water tritium in 14 minutes, while in laboratory experiments 73 percent of the concentration of tritium in aquarium water was acquired in 30 minutes.

The ratio of the specific activity of tissue-bound tritium in aquatic biota to the specific activity in lake water was determined for several species of aquatic organisms. The highest ratios were found for the larger fish species, but never exceeded one. Tissue-bound tritium, which has a longer biological half-life than tissue water tritium, did not reach equilibrium with the lake water because of the fluctuation of the tritium concentration in White Oak Lake. The concentration of tritium in sediment water was slightly less than the concentration of tritium in the overlying lake water. The organic fraction of the sediment in White Oak Lake was 11 percent, and organically bound tritium in decaying plant and animal life could account for the concentration of bound tritium in the dried sediment.

White Oak Lake is representative of an aquatic environment that receives quantitites of tritium significantly greater than would be released from nuclear power stations. The estimated radiation dose to man from eating fish was 1.8 mrem/yr, which is less than the dose that might be received from drinking water from White Oak Lake.

REFERENCES

DISCUSSION

R. KIRCHMANN (Chairman): The results of this study suggest that the tritium is released in the form of tritiated water. Was the chemical form of $^3$H in the effluents investigated?

B.G. BLAYLOCK: No, this was not determined. However, it was assumed that the form in the lake was tritiated water. Tritiated water is released from waste disposal operations at the Oak Ridge National Laboratory, but other chemical forms are undoubtedly included in the releases from operational facilities.

J.K. MIETTINEN: The specific activity of your organism-bound tritium was about 0.3 of that of the tritium in the lake water. This is in good agreement with our results on specific tritium activity in the tissues of pea plants in comparison with the water in which the plants were grown. Tritium always appears to be discriminated against when penetrating the biological membranes.

Did you observe any radiation effects in your organisms, and what was the dose rate to them?

B.G. BLAYLOCK: In this particular study radiation effects were not investigated and dose rates were not calculated for the aquatic organisms. However, at the concentrations of tritium measured in the water and in the organisms, I do not believe that radiation effects could be detected with our present techniques.

Other radionuclides are released to White Oak Lake and contribute to the radiation dose received by the aquatic organisms. Not only are the organisms exposed to radionuclides in their food and water, but benthic organisms are exposed to radionuclides which accumulate in the bottom sediments. A dose rate calculated in 1970 for the larvae of the midge Chironomus tentans which lives in the bottom sediments, was 11 rad/g. However, I would not expect detectable radiation effects at this dose rate.
MONITORING FOR THE DISPERSION OF DISCHARGED TRITIUM IN SEA-WATER AROUND THE TOKAI-MURA FUEL REPROCESSING PLANT

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Japan

Abstract

MONITORING FOR THE DISPERSION OF DISCHARGED TRITIUM IN SEA-WATER AROUND THE TOKAI-MURA FUEL REPROCESSING PLANT.

Low-level liquid waste effluent has been released batchwise into the Pacific Ocean from the fuel reprocessing plant of PNC sited in Tokai-mura since September 1977. The liquid effluent is discharged from the nozzle located about 17 m below the surface of the sea and is dispersed by the ambient cross stream. Approximately 800 Ci of tritium were released into the sea from October 1977 to June 1978. Tritium concentrations in sea-water were measured in accordance with a special monitoring programme to examine the dilution and dispersion of effluent in sea-water. The results of the tritium measurement showed that the effluent was diluted by two or three orders of magnitude by mechanical jet mixing immediately after the discharge from the nozzle, and was further diluted by one or two orders of magnitude by travelling several hundred metres along the current. It was observed that the concentration of tritium in surface water was raised temporarily after the discharge of more than several tens of curies, but returned to the background level within a day or so.

1. INTRODUCTION

Since the start of the hot-test operation of the fuel reprocessing plant, PNC, at Tokai-mura, the low-level liquid effluent has been released into the Pacific Ocean through the pipeline. The amount of tritium in LWR spent fuels depends on their burn-up, and a large portion of the tritium inventory is released in the liquid stream when the fuels are processed. The monthly discharge and the average concentration of tritium during the test campaign are given in Table I.

A special and extensive environmental survey programme was planned to examine the transport behaviour of the low-level liquid effluent discharged into
TABLE I. TRITIUM DISCHARGED INTO THE SEA

<table>
<thead>
<tr>
<th>Period</th>
<th>Discharged tritium activity (Ci)</th>
<th>Average tritium concentration (Ci/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oct. 1977</td>
<td>3.0</td>
<td>$2.3 \times 10^{-3}$</td>
</tr>
<tr>
<td>Nov.</td>
<td>17</td>
<td>$8.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>Dec.</td>
<td>37</td>
<td>$2.0 \times 10^{-2}$</td>
</tr>
<tr>
<td>Jan. 1978</td>
<td>8.2</td>
<td>$4.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>Feb.</td>
<td>10</td>
<td>$4.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>Mar.</td>
<td>51</td>
<td>$2.0 \times 10^{-2}$</td>
</tr>
<tr>
<td>Apr.</td>
<td>49</td>
<td>$2.0 \times 10^{-2}$</td>
</tr>
<tr>
<td>May</td>
<td>260</td>
<td>$7.2 \times 10^{-2}$</td>
</tr>
<tr>
<td>Jun.</td>
<td>370</td>
<td>$8.8 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

The ocean by measuring the tritium concentration in sea-water. The aims for examining the effluent dispersion in sea-water are as follows:

(a) Dilution of effluent by mechanical jet mixing with sea-water immediately after discharge.
(b) Dilution of effluent plume by the sea-water turbulence and movement down to several kilometres from the discharge point.
(c) Change of tritium concentration in sea-water with time in the intermittent discharge of the effluent.

The results obtained by measuring tritium concentration in sea-water during the hot-test operation of the plant (Nov. 1977–Jun. 1978) are presented here.

2. EXPERIMENTAL

2.1. Sea-water sampling

The pipeline for discharging the low-level liquid effluent extends 2 km from the plant to the discharge point and is installed under the sea-bed. The exit nozzle is located a few metres above the sea bottom at 1.8 km off shore where the sea-water is about 20 m deep, as shown in Fig. 1. The effluent is discharged from the exit upward vertically with an initial velocity of about 7 m/s.

Usually about 200 cm$^3$ of the effluent is discharged at one time after monitoring the activities of major radionuclides such as $^{106}$Ru/Rh, $^{137}$Cs and
$^{144}$Ce/Pr, tritium and gross alpha and beta activities. The authorized discharge limit for tritium is 200 Ci daily and about 50,000 Ci yearly. The concentration of tritium in the effluent is limited to less than 0.67 mCi/cm$^3$.

Three types of sampling were performed according to the experimental purposes as described below.

(a) Sampling for determining the initial dilution

During the effluent discharge, nine sea-water samples were taken at the surface of the sea just above the exit of the pipeline. This type of sampling was performed for twenty discharges.

(b) Sampling across the plume for determining the dispersion

After the initial dilution from the jet mixing, the effluent is dispersed by the currents and eddy motion of sea-water. At about 350 m downstream from the discharge point, samples were taken at grid points in a vertical plane crossing the plume (cross-sectional sampling), as shown in Fig. 2. Since the dispersion of the effluent is not visible, dye solution (uranine) was added at the point of discharge to locate the centre line of the effluent plume. About 40 samples at a cross-section were collected in each release using a Van-Dorn water sampler.

(c) Sampling for examining the variation of the tritium concentration in the sea-water with time after discharge

Seven or thirty samples of surface water were collected at one time at the sampling points illustrated in Fig. 3.
2.2. Measurement of tritium in sea-water

The sea-water samples were distilled and the tritium concentrations were measured directly by the liquid scintillation counting technique. Generally 10 ml of the distillate was measured by a conventional counter, where the detection limit was $1 \times 10^{-6}$ $\mu$Ci/cm$^3$. To measure the lower level of tritium concentration, 50 ml of the distillate was measured by a low-background type counter to obtain the detection limit of $3 \times 10^{-8}$ $\mu$Ci/cm$^3$ [1, 2], corresponding to the present background level in the surface sea-water.

3. RESULTS AND DISCUSSION

3.1. Mixing of a buoyant jet

To examine the degree of mechanical mixing of the effluent with ambient sea-water by a buoyant jet, the dilution rate (DR) or the dilution factor (DF) is defined as follows:

$$\text{DR} = \frac{\text{tritium concentration in sea-water (C}_w\text{)}}{\text{tritium concentration in effluent (C}_0\text{)}}$$

$$\text{DF} = \frac{C_0}{C_w}$$
FIG. 3. Map showing sampling locations of surface water.

The experimental results of 11 runs in which the samples near the centre line of the plume were successfully collected are given in Table II. Current velocities of the ambient cross stream measured using a current meter are also given in the table. The DF values ranged from 270 to 3200, the average being 1200.

The DF value depends upon the length of the trajectory of the jet, which is bent over towards the downstream direction by two main factors. One is the degree of the horizontal component of momentum. The other is the differences in density between the effluent and the surrounding sea-water and between the upper layer of sea-water and the lower one.
TABLE II. DILUTION OF THE EFFLUENT BY BUOYANT-JET MIXING

<table>
<thead>
<tr>
<th>Date</th>
<th>Current velocity (cm/s)</th>
<th>Dilution factor</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3 m layer</td>
<td>10 m layer</td>
<td>Average</td>
<td>Range</td>
</tr>
<tr>
<td>6 Dec. 1977</td>
<td>10</td>
<td>15</td>
<td>790</td>
<td>410 – 1100</td>
</tr>
<tr>
<td>17 Dec. 1977</td>
<td>5</td>
<td>10</td>
<td>580</td>
<td>230 – 1640</td>
</tr>
<tr>
<td>19 Dec. 1977</td>
<td>17</td>
<td>10</td>
<td>1520</td>
<td>780 – 6030</td>
</tr>
<tr>
<td>20 Dec. 1977</td>
<td>5</td>
<td>10</td>
<td>270</td>
<td>180 – 390</td>
</tr>
<tr>
<td>22 Dec. 1977</td>
<td>5</td>
<td>5</td>
<td>680</td>
<td>490 – 1110</td>
</tr>
<tr>
<td>23 Dec. 1977</td>
<td>15</td>
<td>6</td>
<td>2670</td>
<td>2100 – 4800</td>
</tr>
<tr>
<td>13 Jan. 1978</td>
<td>15</td>
<td>15</td>
<td>1390</td>
<td>890 – 3540</td>
</tr>
<tr>
<td>19 Jan. 1978</td>
<td>11</td>
<td>10</td>
<td>3200</td>
<td>1050 – 16000</td>
</tr>
<tr>
<td>7 Feb. 1978</td>
<td>5</td>
<td>5</td>
<td>360</td>
<td>160 – 810</td>
</tr>
<tr>
<td>8 Feb. 1978</td>
<td>5</td>
<td>5</td>
<td>370</td>
<td>320 – 480</td>
</tr>
<tr>
<td>9 Feb. 1978</td>
<td>7</td>
<td>15</td>
<td>1210</td>
<td>430 – 2300</td>
</tr>
</tbody>
</table>

**FIG. 4.** Variation of initial dilution factor with current velocity.
Shudo [3] proposed the equations for determining the dilution effect due to the buoyant jet in a uniform cross stream with homogeneous density using his own experimental data and that of Fan [4] who simulated various buoyant jets in an experimental flume. The dilution ratio \(C_w/C_0\) is expressed as follows:

\[
C_w/C_0 = K^{1/3} Fr_u^{2/3} [x/D + 0.7 Fr_d]^{-4/3}
\]

(1)

\[
y/L = 1.5 [x/L + 0.7 Fr_u^{2/3}]
\]

(2)

\[
L = g \left( \frac{\rho_w - \rho_0}{\rho_w} \right) (W_0/U^3)D^3
\]

\[
Fr_u = U \left( \frac{\rho_w - \rho_0}{\rho_w} g \cdot D \right)^{-1/2}
\]

\[
Fr_d = W_0 \left( \frac{\rho_w - \rho_0}{\rho_w} g \cdot D \right)^{-1/2}
\]

\[
K = W_0/U
\]

where

- \(y\) = vertical distance from the nozzle (cm)
- \(x\) = horizontal distance from the nozzle (cm)
- \(\rho_w\) = density of sea-water (g/cm\(^3\))
- \(\rho_0\) = density of effluent (g/cm\(^3\))
- \(W_0\) = initial velocity of jet (cm/s)
- \(U\) = velocity of cross stream (cm/s)
- \(D\) = inner diameter of the nozzle (cm)

The relationship between the velocity of the ambient cross stream \(U\) and DF value is obtained, as shown in Fig. 4, by substituting the values of 1700 cm, 5 cm, 1.000 g/cm\(^3\) and 1.025 g/cm\(^3\) for \(y\), \(D\), \(\rho_0\) and \(\rho_w\) respectively. 

\(\rho_w = 1.025\ g/cm^3\) is the mean value of the coastal water of this area. The observed values given in Table II are also plotted in Fig. 4.

The observed values except for several points fit the calculated line well. The difference between the calculated and experimental data has resulted from the assumption of a uniform cross stream made in the theoretical calculation.
<table>
<thead>
<tr>
<th>Date</th>
<th>Concentration in effluent (µCi/m³)</th>
<th>Discharge rate (Ci/s)</th>
<th>Sampling point and peak concentration in plume</th>
<th>Dilution ratio²</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Travelling distance (m)</td>
<td>Travelling time (min)</td>
</tr>
<tr>
<td>24 Nov. 1977</td>
<td>3.9 × 10⁻²</td>
<td>5.0 × 10⁻⁴</td>
<td>475</td>
<td>61</td>
</tr>
<tr>
<td>10 Dec. 1977</td>
<td>7.0 × 10⁻²</td>
<td>1.1 × 10⁻³</td>
<td>360</td>
<td>26</td>
</tr>
<tr>
<td>17 Dec. 1977</td>
<td>3.9 × 10⁻²</td>
<td>6.0 × 10⁻⁴</td>
<td>390</td>
<td>317</td>
</tr>
<tr>
<td>19 Dec. 1977</td>
<td>3.8 × 10⁻²</td>
<td>5.6 × 10⁻⁴</td>
<td>325</td>
<td>188</td>
</tr>
<tr>
<td>18 Jan. 1978</td>
<td>5.9 × 10⁻³</td>
<td>8.3 × 10⁻³</td>
<td>350</td>
<td>25</td>
</tr>
<tr>
<td>19 Jan. 1978</td>
<td>1.6 × 10⁻²</td>
<td>2.2 × 10⁻⁴</td>
<td>420</td>
<td>91</td>
</tr>
<tr>
<td>7 Feb. 1978</td>
<td>2.9 × 10⁻³</td>
<td>3.7 × 10⁻⁴</td>
<td>340</td>
<td>114</td>
</tr>
<tr>
<td>9 Feb. 1978</td>
<td>2.3 × 10⁻³</td>
<td>2.9 × 10⁻⁴</td>
<td>385</td>
<td>41</td>
</tr>
<tr>
<td>21 Feb. 1978</td>
<td>2.2 × 10⁻³</td>
<td>3.4 × 10⁻⁴</td>
<td>345</td>
<td>48</td>
</tr>
<tr>
<td>28 Feb. 1978</td>
<td>1.3 × 10⁻²</td>
<td>2.0 × 10⁻⁴</td>
<td>340</td>
<td>42</td>
</tr>
<tr>
<td>3 Mar. 1978</td>
<td>1.2 × 10⁻³</td>
<td>1.7 × 10⁻⁴</td>
<td>225</td>
<td>47</td>
</tr>
<tr>
<td>11 Mar. 1978</td>
<td>1.9 × 10⁻²</td>
<td>2.6 × 10⁻⁴</td>
<td>90</td>
<td>116</td>
</tr>
<tr>
<td>15 Mar. 1978</td>
<td>8.6 × 10⁻³</td>
<td>1.2 × 10⁻²</td>
<td>375</td>
<td>138</td>
</tr>
<tr>
<td>20 Mar. 1978</td>
<td>2.9 × 10⁻²</td>
<td>3.7 × 10⁻⁴</td>
<td>345</td>
<td>125</td>
</tr>
<tr>
<td>2 Apr. 1978</td>
<td>3.3 × 10⁻³</td>
<td>5.2 × 10⁻⁴</td>
<td>220</td>
<td>73</td>
</tr>
<tr>
<td>9 Apr. 1978</td>
<td>1.6 × 10⁻³</td>
<td>2.4 × 10⁻⁴</td>
<td>250</td>
<td>34</td>
</tr>
<tr>
<td>2 Jun. 1978</td>
<td>4.2 × 10⁻¹</td>
<td>6.4 × 10⁻³</td>
<td>150</td>
<td>86</td>
</tr>
<tr>
<td>3 Jun. 1978</td>
<td>3.1 × 10⁻¹</td>
<td>4.8 × 10⁻³</td>
<td>150</td>
<td>31</td>
</tr>
</tbody>
</table>

² A = average concentration/concentration in effluent
B = average concentration/discharge rate.
3.2. Dispersion of the effluent by sea-water turbulence and movement

After the initial jet mixing of the effluent mentioned above, the dispersion caused by the turbulence and motion of the currents in the sea-water takes place. In our experiments complicated characteristics of coastal sea-water movement often make it difficult to find exactly the centre line of the effluent plume. Among 50 trials on the across-the-stream sampling, 18 runs were successfully performed to obtain peak concentration of the plume as shown in Table III.

To evaluate the concentration along the centre line of a plume released from a continuous fixed source, an empirical equation was derived based upon dye-diffusion experiments at the site, and applied to the pre-operational safety assessment of the plant. The equation is expressed as follows:

\[
C(x) = \frac{q}{UH} \cdot \text{erf} \left( \frac{yU}{4\sqrt{\alpha x}} \right) \quad \text{(Ci/cm}^3\text{)}
\]  

where \(C(x)\) is the depth-mean concentration on the centre line at distance \(x\) (cm) from the source, \(q\) is the discharge rate of radioactivity (Ci/s), \(U\) is the current speed (cm/s), \(H\) is the thickness of the vertical mixing layer (cm), \(\alpha\) is the diffusion parameter (cm\(^2\)/s\(^2\)) and \(\text{erf}(y) = \frac{2}{\sqrt{\pi}} \int_0^y e^{-t^2} dt\).

If the distance \(x\) from the source is much larger than the size of the source, the above equation is approximated by the following with \(\alpha = 0.1415\).

\[
C(x) = 0.75 \cdot \frac{q}{xH} \quad \text{(Ci/cm}^3\text{)}
\]  

\(q = C_0 \cdot V\)

where \(V\) is the discharge rate of the effluent (cm\(^3\)/s).

\(H\) and \(V\) were taken to be 460 and \(1.39 \times 10^4\) cm\(^3\)/s (50 m\(^3\)/h), respectively, in the pre-operational safety assessment. If these values are employed, the dilution ratio at a distance \(x\) cm is reduced to the following:

\[
C(x)/C_0 = \frac{22.6}{x}
\]  

The comparison of the observed dilution ratios with those calculated by Eq. (5) is shown in Fig. 5. The observed values are lower than the calculated ones. The difference may be explained by two factors: (1) Sampling was not always carried out on the centre line of the plume, and (2) the application of \(H = 460\) cm in Eq. (4) has resulted in the over-estimation of the depth mean concentration in comparison with the observed value.
**FIG. 5.** Variation of minimum dilution ratio with distance travelled from the discharge point.

**FIG. 6.** Vertical concentration profile in a plume at about 350 m downstream from the discharge point.
TABLE IV. TEMPORAL VARIATION OF THE AVERAGE TRITIUM CONCENTRATION IN SEA-WATER WITHIN SEVERAL KILOMETRES AROUND THE DISCHARGE POINT

<table>
<thead>
<tr>
<th>Date</th>
<th>$^3$H discharged (Ci)</th>
<th>Time elapsed after discharge (h)</th>
<th>Average $^3$H concentration in sea-water ($10^{-6}$ μCi/cm$^3$)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>23 Feb. 1978</td>
<td>1.6</td>
<td>8</td>
<td>1 (30)</td>
</tr>
<tr>
<td>27 Mar. 1978</td>
<td>0.4</td>
<td>9</td>
<td>1 (30)</td>
</tr>
<tr>
<td>11 Apr. 1978</td>
<td>0.5</td>
<td>13</td>
<td>1 (30)</td>
</tr>
<tr>
<td>11 May 1978</td>
<td>0.3</td>
<td>11</td>
<td>0.09 ± 0.03 (4)</td>
</tr>
<tr>
<td>12 May 1978</td>
<td>0.3</td>
<td>11</td>
<td>0.07 ± 0.03 (7)</td>
</tr>
<tr>
<td>13 May 1978</td>
<td>0.1</td>
<td>12</td>
<td>1 (7)</td>
</tr>
<tr>
<td>14 May 1978</td>
<td>0.1</td>
<td>12</td>
<td>1 (7)</td>
</tr>
<tr>
<td>17 May 1978</td>
<td>0.5</td>
<td>4</td>
<td>0.05 ± 0.03 (7)</td>
</tr>
<tr>
<td>20 May 1978</td>
<td>0.1</td>
<td>11</td>
<td>1 (7)</td>
</tr>
<tr>
<td>24 May 1978</td>
<td>22</td>
<td>7</td>
<td>0.04 ± 0.03 (7)</td>
</tr>
<tr>
<td>1 Jun. 1978</td>
<td>190</td>
<td>5</td>
<td>1.9 ± 4.1 (30)</td>
</tr>
<tr>
<td>15 Jun. 1978</td>
<td>1.5</td>
<td>8</td>
<td>0.07 ± 0.06 (7)</td>
</tr>
<tr>
<td>16 Jun. 1978</td>
<td>1.1</td>
<td>8</td>
<td>0.06 ± 0.05 (7)</td>
</tr>
<tr>
<td>17 Jun. 1978</td>
<td>0.9</td>
<td>12</td>
<td>0.07 ± 0.05 (7)</td>
</tr>
<tr>
<td>19 Jun. 1978</td>
<td>0.7</td>
<td>5</td>
<td>0.05 ± 0.03 (7)</td>
</tr>
<tr>
<td>10 Jul. 1978</td>
<td>–</td>
<td>–</td>
<td>0.05 ± 0.02 (7)</td>
</tr>
</tbody>
</table>

* Number of sampling points in parentheses.

Figure 6 shows the vertical distribution of the mean concentration of tritium relative to the maximum concentration in each run, and the average thickness $H$ seems to be greater than 460 cm.

3.3. Variation of tritium contents in surface water after the termination of discharge

Fifteen runs were carried out for this purpose from January to June 1978. Analytical results are shown in Table IV. The detection method for the lower level of tritium concentration down to $3 \times 10^{-8}$ μCi/cm$^3$ was adopted for eight runs. The background level of tritium concentration in the coastal water of the
Tokai area has been measured since June 1971 as part of the pre-operational monitoring for the reprocessing plant. The mean value and one sigma standard deviation of tritium concentration thus obtained were $(8 \pm 8) \times 10^{-9} \mu\text{Ci/cm}^3$ [5].

In only one run was an appreciable increase in tritium concentration observed. This was obviously caused by the effluent released from the plant. The sampling of that run began 4.5 h after the discharge of two batches containing 190 Ci of tritium. The amount of tritium discharged daily and the concentrations in surface sea-water in each run plotted against time are shown in Fig. 7.

As seen in Fig. 7, the concentration of tritium in the surface sea-water increases temporarily and locally in the surveyed area after the discharge of more than several tens of curies, but returns to the background level within a day or so. It shows that tritium released into this area rapidly disperses and is transported from the area.
4. CONCLUSION

Using tritium as an indicator, the following conclusions are drawn concerning transport behaviour of the effluent released into the open sea:

(a) Dilution due to the mechanical mixing of buoyant jet fitted well the results of the flume experiments.
(b) A slightly larger dispersion was observed than that expected from the equation used in the pre-operational safety assessment.
(c) The liquid effluent released into the sea-water does not remain in the adjacent area for long.

REFERENCES


DISCUSSION

A.B. GUREGHIAN: Have your experiments enabled you to formulate a relationship for the hydrodynamic dispersion as a function of the water velocity?

T. NOMURA: Yes, the experiments have enabled us to establish a relationship between dilution ratio and water velocity in a uniform cross-stream with homogeneous density. We often use the dilution factor or dilution ratio since this is a help in determining the concentrations of radionuclides such as $^{137}$Cs in sea-water.

A.B. GUREGHIAN: Have you, on the other hand, been able to determine a functional relationship between the dilution factor and the density ratio of the buoyant jet and the ambient temperature of the water?

T. NOMURA: No, because the vertical and horizontal distribution of seawater density in the Tokai area was too complicated.

L. FARGES: The dispersion law put forward by the authors of the paper seems to have an applicability which goes beyond Tokai-mura, since at the La Hague facility in France a dilution coefficient of the order of $10^5$ has been established in respect of the mechanical mixing process, and the $1/x$ law ($x$ being the distance to the point of release) has been confirmed as valid over at least 100 km.
TRITIUM IN THE
SAVANNAH RIVER ESTUARY AND
ADJACENT MARINE WATERS*

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Abstract

TRITIUM IN THE SAVANNAH RIVER ESTUARY AND ADJACENT MARINE WATERS.

The tritium distribution in the Savannah River estuary and adjacent marine waters was measured to provide information on the dilution, mixing and movement of Savannah River water in this region. The Savannah River marine region was chosen because the average tritium concentration in this river is ~5 pCi/ml, whereas other rivers in the southeastern United States of America average less than 0.5 pCi/ml. The increased tritium concentration in the Savannah River is due to releases from the Savannah River Plant of the Department of Energy. Tritium measurements have proved particularly effective in estimating the flushing time of the Savannah River estuary (2.4 days) and in delineating the relative contribution to the water masses in Ossabaw and Port Royal Sounds from the river and from sea-water. Ossabaw and Port Royal Sounds are located approximately 20 km south and north of the Savannah River estuary respectively.

INTRODUCTION

The emphasis of many oceanographic studies has shifted to areas closer to the shore because of its importance for recreation, resource development and marine life. An understanding of coastal currents is a first step toward evaluating the influence on biological, chemical and geological processes occurring in the near shore region. Elevated tritium concentrations in the Savannah River provide a unique opportunity to determine the movement of Savannah River water in the estuary and contiguous marine waters (Fig.1). Tritium in the Savannah River comes from fall-out from nuclear weapons tests, natural production by cosmic rays and releases from the Savannah River Plant (SRP), a Department of Energy facility operated by the E.I. du Pont de Nemours and Company.

* Paper prepared in connection with work under Contract No. AT(07-2)-1 with the U.S. Department of Energy.
FIG. 1. Savannah River, estuary and adjacent marine waters.
SRP has three production nuclear reactors, two nuclear fuel separations plants, a heavy water plant, and a fuel fabrication facility. Most of the tritium released from SRP comes from: (1) the neutron activation of the heavy water moderator, deuterium oxide, used not only to moderate the neutrons in the reactor, but also to remove the heat produced by the nuclear reactions occurring within the reactor; (2) the irradiation of lithium in the reactors to produce tritium; and (3) the tritium resulting from uranium fission [1]. Tritium is released to surface streams from the reactor area fuel and target storage basins, and indirectly by discharge from the separations area to seepage basins.

The average tritium concentration for 1977 at the below plant monitoring site, Highway 301 (Fig.1), was 4.8 pCi/ml [2]. This would result in an annual dose commitment of \( \sim 0.5 \) mrem, if 1.2 ltr of the water is consumed daily. This is \( \sim 0.5\% \) of the natural radiation dose.

Although the tritium content has been used to determine the travel time and dispersion coefficients for the Savannah River [3], it has not been used to study the movement of Savannah River water in the marine region. The purpose of this study is to report the results of studies: (1) tritium-salinity relationships in the Savannah River estuary, (2) the flushing of the estuary, and (3) the dilution and movement of tritium in the estuary and inland marine waters.

**ANALYTICAL METHODS**

Samples were distilled before analysis to avoid counting and/or enrichment problems. Tritium concentrations greater than 1 pCi/ml were determined by liquid scintillation counting. When samples were less than 1 pCi/ml, gas proportional counting and/or enrichment schemes were used. The two enrichment methods were gas chromatography [4] and electrolysis [5, 6]. Analytical measurement errors for all samples averaged \( \pm 15\% \).

**SAVANNAH RIVER ESTUARY**

The Savannah River estuary has a main (north) and a secondary channel (Fig.2). The main channel is a harbour of nearly constant cross-sectional shape, and is an example of a moderately stratified estuary. The main channel is maintained by dredging to a minimum width of \( \sim 120 \) m at a mean low water, a length of \( \sim 35 \) km, and a depth of \( \sim 11 \) m [7]. The South Channel is relatively shallow, 2.5 m, is 16 km long, and is of minor importance in the transport of tritium. Studies conducted in the estuary determined the tritium-salinity relationship for dilution studies and estimated an approximate flushing time for the estuary. The flushing time of an estuary is a crude estimate of how long a conservative pollutant will remain in an estuary.
FIG. 2. Tritium concentration in the Savannah River estuary (pCi/ml).

FIG. 3. Tritium-salinity relationship for Savannah River water.
To determine the tritium-salinity relationship, tritium concentrations and the salinity of surface water samples were measured at several locations in the Savannah River estuary (Fig. 2). The samples were collected during a period of relatively constant tritium concentrations in the Savannah River. The tritium concentration (Fig. 3) decreased toward the mouth of the estuary, because the tritium content of Gulf Stream sea-water (~0.05 pCi/ml) is less than that of Savannah River water. The observed linear relationship is analogous to the temperature-salinity analysis of water masses [8]. The linear plot obtained from the data indicates that only two water types are present in the estuary: Savannah River and South Atlantic Bight water. If the tritium-salinity curve is extrapolated to 0 salinity, the intercept is 4.4 pCi/ml, which compares with the 4.4 pCi/ml measured at the freshwater station.

The flushing time for the Savannah River estuary was calculated from tritium and hydraulic data by three different methods: (1) the tidal prism method; (2) the fraction fresh-water method; and (3) tritium data using the fraction fresh-water method. These methods usually underestimate the flushing times and also assume that the estuary is not stratified [9].

Tidal prism method

The tidal prism method assumes that the water entering on flood tide is fully mixed with that in the estuary, and that the volumes of sea-water and river water added together equal the volume of the tidal prism. The tidal prism is that volume of water between the high- and low-water marks. On the ebb tide, the same volume of water is removed, and the fresh-water content must equal the increment of river flow. The water volume estimates were obtained from the Savannah Harbor Investigation and Model Study [7].

For the Savannah River estuary:

\[
T \text{ (flushing time)} = \frac{\text{Volume at low water} + \text{tidal prism}}{\text{tidal prism}}
\]

\[
= \frac{5.39 \times 10^7 \text{ m}^3 + 1.29 \times 10^7 \text{ m}^3}{1.29 \times 10^7 \text{ m}^3}
\]

\[
= 4.2 \text{ tidal cycles or } \sim 2.1 \text{ days}
\]

Fraction fresh water

The fraction fresh-water method uses the residence time formula [10]:

\[
T \text{ (flushing time)} = \frac{\text{Total amount of fresh water in estuary}}{\text{River flow rate}}
\]
FIG. 4. Tritium distribution in the Savannah River estuary.

The total amount of fresh water in the Savannah River estuary was calculated from estuary water volume and fraction fresh water present at both high and low water using data from the Savannah Harbor Investigation and Model Study [7]. These data represent an average for all hydraulic conditions of the Savanna River estuary.

Low water:

\[
T = \frac{5.39 \times 10^7 \text{ m}^3 \times 0.73}{1.98 \times 10^3 \text{ m}^3/\text{s} \times 86400 \text{s/day}} = 2.3 \text{ days}
\]

High water:

\[
T = \frac{6.68 \times 10^7 \times 0.26}{1.98 \times 10^2 \text{ m}^3/\text{s} \times 86400 \text{s/day}} = 1.1 \text{ days}
\]

Average flushing time = 1.7 days.

Tritium method

Samples were collected in the Savannah River estuary following a tritium release from the SRP. The results shown in Fig. 4 were used to estimate a flushing
time. The tritium analogue of the fraction fresh-water calculation was used, but only surface concentrations were taken. The average tritium content in the estuary was calculated to be 727 Ci. The input rate, 2166 Ci/week, was calculated by averaging the tritium input at a monitoring location below the plant for three weeks in May. Tritium flushing time was calculated to be:

\[ T = \frac{727 \text{ Ci}}{2166 \text{ Ci/week}} \times \frac{7 \text{ days}}{\text{week}} \]

\[ = 2.4 \text{ days} \] (2)

These three flushing times are in good agreement, but represent the minimum time a conservative pollutant would reside in the Savannah River estuary.

SAVANNAH RIVER WATER IN THE INLAND MARINE WATERS

Water movement is governed by wind, tidal action, interconnecting creeks, coastal currents and fresh-water flow. With the aid of the U.S. Coast Guard, samples of surface water were collected from the inland marine area (Fig.5). In general, lower salinities were found south of the Savannah River as a result of a general southward current, which is substantiated by the higher tritium concentrations.

To evaluate the mixing of water in this area, salinity versus tritium plots for the Coosawhatchie and Ogeechee River water were added to Fig.3 to yield Fig.6. The source of the sea-water is considered to be the Gulf Stream with a 0.05 pCi/ml tritium concentration and 36.1 parts per thousand (%) salinity. Points that lie on or within ~15% of the lines are considered to be dilution of that river source with sea-water; points between the two lines represent water mixes from the Savannah River, nearby rivers and sea-water. Since the Ogeechee and Coosawhatchie Rivers are nearly identical in tritium concentration, they are lumped as one fresh-water source for this analysis. A method for estimating the relative fresh sea-water contributions from the different sources is as follows:

\[ T = \frac{(T1 \cdot M1) + (T2 \cdot M2) + (TS \cdot MS)}{M1 + M2 + MS} \] (3)

where tritium concentration (T) at a location fed by three sources is the sum of the products of tritium measured at the Savannah River (T1), a nearby river (T2) and sea-water (TS), and the respective contributing water volumes M1, M2 and MS.
FIG. 5. Tritium concentration in tidal waters (pCi/ml).

FIG. 6. Tritium-salinity relationship of inland waters in the Savannah area.
<table>
<thead>
<tr>
<th></th>
<th>Salinity (%o)</th>
<th>Tritium (pCi/ml)</th>
<th>Relative volume (%)</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>AN</td>
<td>BN</td>
</tr>
<tr>
<td>Port Royal Sound</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sample AN</td>
<td>25.8</td>
<td>0.8</td>
<td>71</td>
<td>77</td>
</tr>
<tr>
<td>Sample BN</td>
<td>27.9</td>
<td>0.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Source water:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gulf Stream</td>
<td>36.2</td>
<td>0</td>
<td>14</td>
<td>11</td>
</tr>
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<td>Savannah River</td>
<td>0</td>
<td>4.4</td>
<td>15</td>
<td>12</td>
</tr>
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<td>Coosawhatchie River</td>
<td>0</td>
<td>1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>AS</td>
<td></td>
<td></td>
<td>BS</td>
</tr>
<tr>
<td>Ossabaw Sound</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sample AS</td>
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<td>1.0</td>
<td>29</td>
<td>64</td>
</tr>
<tr>
<td>Sample BS</td>
<td>23.4</td>
<td>1.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Source water:</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Gulf Stream</td>
<td>36.2</td>
<td>0</td>
<td>14</td>
<td>26</td>
</tr>
<tr>
<td>Savannah River</td>
<td>0</td>
<td>4.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Little Ogeechee River</td>
<td>0</td>
<td>0.7</td>
<td>57</td>
<td>10</td>
</tr>
</tbody>
</table>

The salinity at the same location is

\[
S = \frac{(S1 \cdot M1) + (S2 \cdot M2) + (SS \cdot MS)}{M1 + M2 + MS}
\]

(4)

where S1, S2 and SS are the salinities for each source of water. The water volumes can be normalized:

\[
I = \frac{M1}{M} + \frac{M2}{M} + \frac{MS}{M}
\]

(5)

where M is the total water volume and the following assumptions are made:

1. The salinities of the rivers are essentially 0 when compared with the Gulf Stream (0.02%o compared with 36.2%o)
2. The tritium content of the Gulf Stream is negligible when compared with the rivers (0.05 pCi/ml to ~1 pCi/ml).
Substituting and simplifying:

\[
\frac{MS}{M} = \frac{SS}{36.2} \quad \text{(sea-water relative volume)} \quad (6)
\]

\[
\frac{M2}{M} = \frac{(T1 \cdot SS) - (T \cdot SS) - (S \cdot T1)}{SS \cdot (T1 - T2)} \quad \text{(one river's relative volume)} \quad (7)
\]

\[
\frac{M1}{M} = 1 - \frac{M2}{M} - \frac{MS}{M} \quad \text{(other river's relative volume)} \quad (8)
\]

The relative river-water and sea-water volume contributions for samples AS, BS, AN and BN (Fig.6), were calculated by using Equations (6), (7) and (8) (Table I). The samples from the Port Royal Sound region (AN, BN) had almost equal mixtures of Savannah and Coosawhatchie River water, whereas one Ossabaw Sound sample (AS) showed an abundance of Ogeechee River water. A second Ossabaw Sound region sample (BS) contained more Savannah River water than Ogeechee River water. Based on these results, the transport route of Savannah River water is extremely complex and not easily delineated, but in the upper inland marine waters, interconnecting waterways may be important.

CONCLUSION

The relatively high concentration of tritium in the Savannah River provides a useful tool to study mixing processes in this coastal environment. In the Savannah River estuary, the tritium concentration decreased from the upper to the lower estuary because of its dilution with sea-water. The tritium dilution in the estuary was inversely proportional to the salinity, and could be used as an indication of the amount of tritium dilution that can be expected for steady river tritium concentrations. The estuarine flushing time for tritium was estimated to be about 2.4 days, which compared favourably with tidal prism (2.1 days) and fraction fresh-water (1.7 days) estimate methods. The inland marine water dilution of tritium could be described by a three source box model to show the relative water contributions from the Savannah River, nearby rivers and sea-water.

REFERENCES


**DISCUSSION**

A.B. GUREGHIAN: Can the flow regime in the Savannah River be described as turbulent?

D.W. HAYES: Yes.

A.B. GUREGHIAN: I should now like to ask a question similar to one which I recently put to Mr. Nomura: have you been able to formulate a relationship for the hydrodynamic dispersion as a function of the shear velocity?

D.W. HAYES: No, the hydrodynamic dispersion coefficients were obtained from dye and tritium tracer studies in the river. The shear velocity approach probably could be used, although a large river-velocity data-base would be required.

F.A. PRANTL: It was interesting to see your results on tritium dilution in a large river system. In an earlier study we used the seasonal variations of natural tritium in the tributaries of a river draining a large glaciated basin in the Canadian Rockies to determine the contributions of glacial run-off. This study also provided us with useful information on seasonal changes of groundwater contributions, which still represent a difficult problem for the hydrologist. I feel that your study may also yield valuable information of this type, and I would like to encourage you to look at your data from this point of view as well.

D.W. HAYES: Thank you.
AN ASSESSMENT OF THE RADIOLOGICAL IMPACT OF TRITIUM RELEASED TO SEA FROM THE WINDSCALE FUEL ELEMENT REPROCESSING PLANT

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Ministry of Agriculture, Fisheries
and Food,
Lowestoft,
Suffolk,
United Kingdom

Abstract

AN ASSESSMENT OF THE RADIOLOGICAL IMPACT OF TRITIUM RELEASED TO SEA FROM THE WINDSCALE FUEL ELEMENT REPROCESSING PLANT.

Tritium is discharged routinely to the Irish Sea in low-level liquid wastes from the nuclear fuel reprocessing plant at Windscale, Cumbria. In the course of a programme of environmental research utilizing the tritium discharges, samples of sea-water have been collected regularly from the northern area of the Irish Sea during the period 1968–74. Electrolysis has been used to concentrate the tritium to achieve a limit of detection in sea-water of about 10 pCi/l. Results have been corrected for the background concentrations of tritium from both naturally occurring sources and nuclear weapons' testing. The corrected results show that under conditions of constant discharge there is a fairly uniform concentration of tritium in the vicinity of the discharge point which in numerical terms corresponds to about 5 pCi/l per Ci/d. The shape of the contours of concentration both close to Windscale and over the wider area of the northern area of the Irish Sea is discussed in relation to dispersal mechanisms and water turn-over times. Evidence is presented of marked reversals in the paths taken by activity after discharge from Windscale. Finally the radiological safety aspects of tritium discharges are discussed in terms of both dose to critical groups and collective dose. It is shown that the limiting discharge rate in terms of the need to keep individual doses within ICRP limits is something like \( 9 \times 10^6 \) Ci yearly. A simple, although conservative, assessment of collective dose shows that this is likely to be less restrictive than the dose to individuals.

1. INTRODUCTION

Tritium is produced during the operation of all nuclear reactors but in the case of the gas-cooled reactors in use in the United Kingdom it arises chiefly from tertiary fission of uranium-235. The rate of production of tritium depends on several factors,

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but from the operation of all the magnox reactors of the first phase of the UK nuclear power programme the total annual rate of tritium production was about 30 000 Ci/a. Almost all the tritium is retained within the fuel cladding until the reprocessing stage which is undertaken for the United Kingdom power programme by British Nuclear Fuels Limited at their plant at Windscale in Cumbria. At reprocessing the bulk of the tritium appears as tritiated water in the plant liquor streams, only a small fraction of the total being lost in gaseous form. By the nature of the process, practically all the tritium appears eventually in the low specific activity aqueous effluent which is discharged to sea. The location of Windscale is shown in Fig. 1 together with the principal features of the Irish Sea.
Tritium is of very low radiotoxicity, preliminary estimates having put the environmental capacity of the north-eastern Irish Sea for receipt of tritium as HTO in the region of $10^3$ Ci/a. Thus the tritium which has arisen during the reprocessing of all the fuel produced to date from the UK power programme has been trivial by comparison.

Because these discharges have been of such low radiological significance, no routine monitoring has been required to confirm their radiological safety. Nevertheless, a programme of research utilizing the discharges was initiated in 1967 in recognition of the great value of tritium as a water tracer. As such it provides not only useful information of a hydrographic nature but also a base line against which to compare the behaviour of less conservative components of the Windscale discharge, offering an opportunity to follow dispersion processes free from complications related to loss on to sediments which affect all the other radionuclides to some extent or another.

2. SAMPLING AND ANALYSIS

During the period 1968-75 samples of sea water for tritium analysis have been collected from the northern Irish Sea. The sampling has been in three parts. Each month a sample has been collected at the eleven shore-line stations from Maryport to the north of Windscale to Walney Island to the south (see Fig. 2). Secondly, at
quarterly intervals, samples of surface, mid depth and bottom water have been collected from the grid of offshore stations shown also in Fig. 2. Thirdly, samples have been obtained over a wider sea area during the course of three major research vessel cruises by RV Cirolana in October 1971 (8a/71), by RV Corella in July 1973 (11/73) and by RV Cirolana again in July 1974 (6b/74). In addition, seawater samples have been collected at a number of stations in the Atlantic to the south-west of Ireland to provide information on the tritium concentration in areas remote from Windscale which are unaffected by discharges from this or any other nuclear facility.

After return to the laboratory, shore-line samples were bulked to produce half-yearly average samples for each station. From the offshore samples collected quarterly, a composite sample was prepared of the surface, mid depth and bottom water from each station. The samples collected during the major cruises were in general surface samples only, although at a few stations, especially in the vicinity of Windscale, bottom samples were also collected and analysed separately to test for stratification of activity.

For analysis, 0.5 l of each sample was double distilled in the presence of carriers to prevent carry over of unwanted elements. An accurate 100 cm$^3$ of the final distillate was transferred to an electrolytic cell and was reduced to 5 cm$^3$ by electrolysis in the presence of sodium peroxide according to the method of Östlund and Werner [1]. Six cells were used, the current being passed through them in series. The current was 6 A initially but was reduced linearly during the electrolysis and was switched off after a total of 250 A-h. A recovery fraction for tritium in the range 0.75–0.9 was achieved, which with the volume reduction of 20 to 1 gave an enrichment of between 15 and 18 times in tritium concentration. The enriched solution was distilled under vacuum after adding lead nitrate in order to release as tritiated water any tritium contained in hydroxyl radicals. The final distillate was analysed by liquid scintillation counting to give a limit of detection for tritium in sea-water of about 10 pCi/l.

3. THE BACKGROUND CONCENTRATION OF TRITIUM IN SEA-WATER

Before attempting to describe the distribution of tritium in the north-east Irish Sea in relation to Windscale discharges, it is necessary to take account of the other sources of this nuclide in sea-water apart from Windscale, and to estimate the contribution of these sources to measured concentrations. There are essentially two sources producing the background against which the Windscale discharges must be assessed. First, there is the tritium present as a result of naturally occurring processes such as the interaction of cosmic rays with atmospheric nitrogen and oxygen, and second there is the tritium present in the environment as a result of the atmospheric testing of nuclear devices since 1954. Both these processes produce tritium in the atmosphere whence it is transported onto the earth's surface via rain.

Because of the run-off into coastal waters of the precipitation over adjacent land areas, the concentrations of tritium in the surface layers of coastal waters tend to be higher than those in similar samples from open ocean locations. Thus the tritium present in an area such as the northern Irish Sea may be regarded as originating from two sources. There is the tritium present in the Atlantic Ocean
TABLE I. CONCENTRATIONS OF TRITIUM AND CAESIUM-137 (pCi/l) AND THE SALINITIES (‰) OF SEA-WATER SAMPLES FROM AREASREMOTE FROM WINDSCALE, NOVEMBER 1974

<table>
<thead>
<tr>
<th>Station number</th>
<th>Location</th>
<th>$^3$H</th>
<th>$^{137}$Cs</th>
<th>Salinity</th>
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<tr>
<td>9A/80</td>
<td>52°00'N</td>
<td>05°28'W</td>
<td>46</td>
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<tr>
<td>9A/81</td>
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<td>05 54</td>
<td>52</td>
<td>0.4</td>
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<td>05 35</td>
<td>50</td>
<td>0.5</td>
</tr>
<tr>
<td>9A/88</td>
<td>51 15</td>
<td>04 34.5</td>
<td>53</td>
<td>0.4</td>
</tr>
<tr>
<td>9B/5</td>
<td>51 30</td>
<td>09 59</td>
<td>22</td>
<td>0.2</td>
</tr>
<tr>
<td>9B/6</td>
<td>51 59</td>
<td>10 59</td>
<td>18</td>
<td>0.2</td>
</tr>
<tr>
<td>9B/17</td>
<td>51 29</td>
<td>11 36</td>
<td>23</td>
<td>NA$^a$</td>
</tr>
<tr>
<td>9B/19</td>
<td>49 60</td>
<td>11 00</td>
<td>11</td>
<td>NA</td>
</tr>
</tbody>
</table>

$^a$ NA - not analysed.

Water which forms a baseline concentration of the nuclide entering the coastal circulation. Then, after entering the coastal system, this water receives the tritium from the second source, namely land run-off.

The relative contributions of these sources to sea-water as it enters and moves through the southern Irish Sea have been investigated by measuring the tritium concentrations at a series of stations to the south-west of Ireland and at the southern entrance to the St. George's Channel. The results together with caesium-137 concentrations and salinities in surface water samples are shown in Table I. The salinities and the caesium-137 concentrations of the samples collected to the south-west of Ireland indicate that these were representative of the North Atlantic water which forms the source of water entering the Irish Sea. The tritium concentrations in this area are in the range 11-23 pCi/l (mean 18.5); these values may be compared with the concentrations of tritium of 0.7 to 5.4 pCi/l in samples of open ocean surface water (area unspecified) collected during the days before nuclear weapons' testing[2]. It is seen that the present-day ocean water concentrations are due almost entirely to weapons' test fallout rather than to natural sources. As land is approached, the influence of run-off becomes apparent in both salinity and tritium concentration. For samples from the Bristol Channel and the southern entrance to St. George's Channel the mean salinity has decreased to 34.91 compared with values in excess of 35.20 in the Atlantic while the tritium concentrations averaged 53 pCi/l, some 32 pCi/l higher than in the Atlantic.

Under the most commonly prevailing conditions, water in the northern area of the Irish Sea has entered from the south via St. George's Channel, and has come under the influence of land run-off throughout this passage. The concentration of tritium in the Windscale area due to non-Windscale sources depends therefore on the effect
TABLE II. CONCENTRATIONS OF TRITIUM (pCi/l) IN RAIN WATER AT ESKDALEMUIR AND MILFORD HAVEN (DERIVED FROM CAMBRAY et al. [4])

<table>
<thead>
<tr>
<th>Year</th>
<th>Eskdalemuir</th>
<th>Milford Haven</th>
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<tbody>
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<td>720</td>
</tr>
<tr>
<td>1967</td>
<td>515</td>
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</tr>
<tr>
<td>1968</td>
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<td>1970</td>
<td>301</td>
<td>293</td>
</tr>
<tr>
<td>1971</td>
<td>429</td>
<td>328</td>
</tr>
<tr>
<td>1972</td>
<td>211</td>
<td>156</td>
</tr>
<tr>
<td>1973</td>
<td>170</td>
<td>131</td>
</tr>
<tr>
<td>1974</td>
<td>216</td>
<td>145</td>
</tr>
<tr>
<td>1975</td>
<td>159</td>
<td>102</td>
</tr>
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</table>

TABLE III. CALCULATED ANNUAL INPUT OF TRITIUM (Ci) INTO THE IRISH SEA FROM RIVERS AND DIRECT PRECIPITATION

<table>
<thead>
<tr>
<th>Year</th>
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<th>II</th>
<th>III</th>
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<td></td>
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<td>1.59 E (4)</td>
<td>3.37 E (4)</td>
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<td></td>
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<tr>
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<td></td>
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<tr>
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</table>
of the input from direct precipitation and run-off on the one hand and the loss by evaporation on the other. Information on the rates of freshwater input and evaporation for each of these sea areas is available from hydrographic studies [3], and may be combined with records of concentrations of tritium in rain water, river water and sea-water to calculate rates of input and loss of tritium during the period of interest.

Monthly concentrations of tritium in rain water from Eskdalemuir and Milford Haven have been reported annually by Cambray et al. [4], and the mean values for the period 1966-75 are in Table II. Unfortunately no such comprehensive measurements have been made on any of the major rivers which flow into the Irish Sea, and it is possible only to assume that the concentrations in river water have been the same as those in rain. Because most of the rivers in question are sustained primarily by direct land run-off rather than from underground sources, this assumption would seem reasonable. Some measurements made on samples collected during 1975 from the River Severn at Ashleworth and Tilley gave tritium concentrations of 1.56 pCi/l which were very similar to those in rain at Milford Haven during that year and give support to the assumption. Using this assumption and freshwater input data from Bowden [3], the amounts of tritium entering each sea area have been calculated and the results are shown in Table III.

For each sea area, the total amount of tritium, A, in the area may be expressed by an equation

\[ \frac{dA}{dt} = C - (\lambda_1 + \lambda_2)A \]

where C is the rate of input, \( \lambda_1 \) is the fractional loss rate from the area due to evaporation, and \( \lambda_2 \) is the fractional loss rate from the area due to advective transport. Although readily obtainable, the general solution to the equation is not useful because the data needed to establish the constant of integration are not available. It is however possible to use the near constancy of the rate of input of tritium from freshwater sources during the period 1968-74 to permit the assumption of equilibrium between input and loss and on this basis the equation reduces to

\[ A = \frac{C}{(\lambda_1 + \lambda_2)} \]

For the southern area from Bowden’s data \( \lambda_1 = 0.15 \text{ a}^{-1} \) and \( \lambda_2 = 0.006 \text{ a}^{-1} \), and the tritium inventory expected therefore at equilibrium with a total annual input of some \( 1.5 \times 10^3 \text{ Ci/a} \) from the Atlantic plus \( 1.7 \times 10^4 \text{ Ci/a} \) from direct precipitation and land run-off is \( 2.1 \times 10^5 \text{ Ci} \). This corresponds to a concentration of 40 pCi/l which is very similar to the values measured in samples from the northern boundary of the area during November 1974.

Similar calculations may be performed for the central and northern sections and the increments in seawater concentrations of tritium as the water moves through each area may be estimated. Taking input rates representative of conditions prevailing in 1972 of \( 5 \times 10^3 \) and \( 10^4 \text{ Ci/a} \) into the central and northern areas, the increments are approximately 5 and 10 pCi/l respectively, with values of up to 50% higher than this in the four years preceding 1972 and up to 50% less by 1975. It is concluded therefore that if during the preceding, say, eight years no tritium had been released from Windscale the concentration of the nuclide in sea-water in the
TABLE IV. QUARTERLY SURVEYS: CONCENTRATIONS OF TRITIUM IN SEA-WATER (pCl/l), CORRECTED FOR THE NON-WINDSCALE BACKGROUND AND NORMALIZED TO A DISCHARGE RATE OF 1 CI/d

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northernmost area would have been something like 55 ± 10 pCi/l. This is the correction which should be applied to the results of the surveys carried out in this area so as to take account of non-Windscale sources. The uncertainties associated with background correction are rather greater than the limit of detection of the measurement technique, and the limit of confidence for tritium concentrations after correction for background is about 30 pCi/l.

4. RESULTS

4.1. Quarterly surveys in the Windscale area

The results of the quarterly surveys at the grid stations shown in Fig. 2 are shown in Table IV. The values given are the concentrations obtained after first correcting for a background of 55 pCi/l as described above and then normalizing to a release rate of 1 Ci/d by dividing by the daily discharge rate averaged over the 13 weeks preceding the date of sampling. Some normalization is essential because, although the average discharge has remained fairly constant in the longer term, there are substantial variations in the shorter term according to the pattern of operation of the Windscale factory. Surveys conducted within a few kilometres of the discharge point tend to reflect the discharges during the weeks immediately prior to sampling. An averaging period of 13 weeks was chosen after examination of the correlation between the net tritium concentrations in sea-water averaged over all the stations sampled at each quarterly survey and the daily rate of discharge averaged over a selection of periods ranging from 1 to 6 months preceding the survey. The highest correlation (0.9) was obtained with a 13 week averaging period. However, the choice is not critical because the correlation was not less than 0.75 for any of the periods used.

The mean normalized seawater concentrations are presented diagrammatically in Fig. 3. For the purpose of understanding the dispersal processes operating in the vicinity of the discharge it would have been valuable to have had more detailed and comprehensive information. Unfortunately the limited data available together with their related uncertainties makes impossible anything more than the semi-qualitative treatment of Fig. 3. The position of the 4 pCi/l boundary especially involves a degree of interpolation although the 5 pCi/l one is better in that it is correct in relation to five points none of whose individual standard errors exceeds 22%. Despite the limitations of the data it is possible to draw several conclusions from them.

None of the points within the 5 pCi/l boundary exceeds this value by more than about 40% and it would appear that there is a fairly uniform concentration of radioactivity per unit discharge rate over an area of radius several kilometres about the point of discharge. In numerical terms this concentration is about 5 pCi/l per Ci/d. Although there is a degree of variation in the result from one survey to another, which reflects presumably changes in the rate of turnover of the water within the area, this average dilution factor of 5 pCi/l per Ci/d is adequate for radiological assessment purposes. The results of the shore-line surveys when corrected and normalized in the same way support the same conclusion. These data have not been presented here because the results fluctuate to a much greater extent than those offshore under the influence of the changes in the rate of freshwater run-off from the
FIG. 3. Tritium in sea-water — average concentrations in Windscale vicinity from December 1971 to February 1974 (pCi/l per Ci/d).

land. As described earlier this run-off is relatively rich in tritium and when there is a significant freshwater input to the coastal sea-water as during periods of rain or snow-melt the tritium concentrations of the shore-line waters increase substantially.

The figure of 5 pCi/l per Ci/d release rate represents the concentration which is in equilibrium with a constant rate of input, and it may be used to deduce the rate of loss of tritium from the area because at equilibrium this must be equal to the rate of input. By taking the mean depth of water as 10 m in the semi-circle of radius 10 km about the point of discharge the total tritium content of this volume may be calculated to be about 8 Ci. Thus on average this volume contains the equivalent of about 8 days input of tritium and the fractional rate of loss is 0.125 d⁻¹ corresponding to a half-time of between 5 and 6 days. Comparison of this result with the value of 13 weeks used earlier as the averaging period for normalizing seawater concentrations to unit discharge rate shows that although the activity is being dispersed within a matter of days from the immediate area of the discharge, it nonetheless persists in the area generally for much longer periods, producing a continuum against which day-to-day operations are seen.
Despite the uncertainties discussed above regarding the contours in Fig. 3, their general shape is regarded as sufficiently reliable to support a tentative conclusion concerning the mechanism by which tritium is being dispersed from the discharge vicinity. The direction of the total ebb and flow in the area is more or less parallel to the coast and the magnitude of the tidal excursion is about 5 km. This excursion is marked to scale on Fig. 4, and it may be seen that after taking account of the smearing action of the tide on the discharge, the contours show no signs of elongation in any particular direction. In other words, it would seem that the activity is spreading isotropically from the point of release, suggesting that the predominant mechanism of dispersion in the immediate vicinity of the outfall is diffusive rather than advective. It has been pointed out by Shepherd (personal communication) that the equilibrium concentration of 5 pCi/l per Ci/d is equivalent to a flow rate of 0.2 km$^3$/d. By considering the flow from the semi-cylindrical volume radius 10 km about the point of discharge, with assumed water depth of 10 m, this corresponds to an effective dispersion velocity for non-Fickian diffusion of rather less than 1 km/d ($7 \times 10^{-2}$ m/s).
Despite their approximately circular shape, the contours close to the point of discharge show some southward displacement. The precise reason for this is not known. The displacement may be more apparent than real being due to the existence of deeper water to the north of Windscale as compared with the south. Although diffusive processes will tend to spread the tritium uniformly, movement of the activity into the large volumes associated with the deeper water to the north will produce relatively lower concentrations in that direction than to the south. Another explanation of the displacement may be in terms of the mechanism which causes the eventual removal of the activity from the locality of Windscale altogether. Studies with other nuclides discharged from Windscale [5, 6] have shown that beyond the area radius about 10 km around the outfall advection assumes greater importance in the transport of the waste. In general but with some occasional but very marked exceptions it is found that the activity moves southwards, following the Cumbrian coast into the general area of Liverpool Bay. The displacement of the contours of Fig. 3 may be further evidence of the action of the prevailing southerly residual current.
4.2. Surveys of north-east Irish Sea

The results of the more extensive cruises are shown in contour form in Figs 4, 5 and 6. The data for these cruises are interesting especially in the context of Windscale plant operations. Two of the cruises (Cirolana 8a/71 in October 1971 and Corella 11/73 in July 1973) coincided with the end of two periods when discharges of tritium from Windscale had remained fairly steady for upwards of a year. The third cruise (Cirolana 6b/74 in July 1974) occurred within a few weeks of the start up of the chemical plant after an eight month shut down during which tritium discharges were virtually nil.

The measured concentrations have been corrected for background and normalized to unit discharge rate as before, except that the average discharge rate used has been chosen to take account of both the greater areas covered by the cruises and the pattern of discharges which preceded each cruise. For the two cruises (8a/71 and 11/73) when the rates had remained fairly constant, the normalizing factor taken was the mean daily discharge over the preceding 12 months. For cruise 6b/74, the
average was taken over the 10 weeks which had elapsed between the cruise and the
recommencement of discharges from Windscale. In considering the result it should
be noted that the limit of detection of 30 pCi/l on the net tritium concentrations (see
Section 3 above) corresponds to a lower limit of significance in the normalized con-
centrations of about 0.3 pCi/l per Ci/d with the rates of discharge concerned.

Two points of interest emerge from cruise 6b/74. After correcting for back-
ground tritium concentrations were virtually all everywhere except in the vicinity of
Windscale and within the confines of the Morecambe and Liverpool Bay areas. In the
immediate vicinity of Windscale, the significant levels of activity were clearly due to
the recently renewed discharges and the observed concentration of 8 pCi/l per Ci/d
is reasonably consistent with the value derived earlier from the local survey work.
The explanation of the activity detected in Liverpool Bay is uncertain. It may be
further evidence of the activity discharged during the preceding 10 weeks following
the southerly movement of water along the Cumbrian coast or it may be the local
influence of the large freshwater discharges from the rivers in this area. Salinity
data show that the latter explanation is probably more significant within Liverpool
Bay itself but further north the increased tritium concentrations were more probably
due to the front moving from Windscale. This is consistent with the observed south-
ward displacement of activity from Windscale seen with other radionuclides [3] and
suggests a rate of movement of about 1 km/d. It is notable that the tritium con-
centrations throughout the northern area of the Irish Sea generally were background at
this time. That this should be so at the end of a period of 9 months of minimal dis-
charges from Windscale suggests a flushing time for the water from the area of the
order of a year which is consistent with the estimates based on both classical hydro-
graphic observations [3] and radiocaesium studies [5].

As mentioned earlier, the other two cruises took place after long periods of fairly
steady discharges from Windscale and the results will be closely representative of
those expected of equilibrium. The normalized water concentrations in the imme-
diate vicinity of the discharge point of about 4 pCi/l per Ci/d in October 1971 and
5 pCi/l per Ci/d in July 1973 are very similar and agree very closely with the value
derived earlier from the quarterly surveys. In contrast with this similarity local to
the discharge point, the distribution of activity at greater distances in October 1971
was markedly different from that seen in the other cruises. In October 1971 the
contours show that the activity was being removed directly from the discharge area
in a north-westerly direction towards the Scottish shore-line and suggest that this
had been happening for several weeks beforehand. This pattern of behaviour is
regarded as important from the hydrographic viewpoint and will be discussed in that
context elsewhere.

5. RADIOLOGICAL ASSESSMENT OF TRITIUM DISCHARGES

For this purpose it is necessary to consider two things. The first is the dose to
members of the group (or groups) of the population who are likely to be most highly
exposed as a result of the discharge. This may be done by the usual critical path
procedure. However, in view of the conservative nature of tritium in sea-water and
the possibility therefore of large numbers of people being exposed as the activity
disperses over a large area, it is necessary also to consider the collective dose.
5.1. Critical exposure pathways

In order to identify the most highly exposed groups of people, we consider the internal exposure pathways by which the activity may return to man. There are two pathways to be considered. The first is the ingestion of tritium associated with marine foodstuffs such as fish, shellfish and edible seaweed, and the second is the inhalation of tritium in association with the water vapour present in the air. In both instances the assessment may be done quite simply by making the reasonable assumption that the tritium is present as tritiated water and that this equilibrates immediately with the ordinary water in the sea, the marine materials which exist in the sea, and in the atmosphere immediately above the sea surface. Although equilibration within biological systems will not be instantaneous, the uptake is sufficiently rapid for the assumption that it is so to be adequate in the circumstances.

5.1.1. Ingestion of foodstuffs containing tritium

It turns out that the most limiting ingestion pathway is via the consumption of fish. The water content of the species of fish which are caught regularly in the Windscale vicinity is about 80% of their net weight. If it is assumed that the concentration of tritium in this water is the same as in the sea-water and that there is no significant loss of water during the processing or cooking of fish, then it may be calculated that, for a constant discharge from Windscale of 1 Ci/d (producing, as shown earlier, an equilibrium concentration of 5 pCi/l in sea-water), the daily intake of tritium by a person eating 265 g (wet weight) of fish per day would be about 1.1 pCi/d. The annual limit of intake by ingestion for members of the general public is about \(2.6 \times 10^3\) µCi/a \([7]\). The above rate of intake from fish consumption is equivalent to approximately \(1.5 \times 10^{-5}\)% of this, giving therefore a limiting environmental capacity of \(7 \times 10^6\) Ci/d. All other ingestion pathways (e.g. consumption of shellfish and laverbread) are less restrictive than this by an order of magnitude at least.

5.1.2. Inhalation of airborne activity

For this purpose it is necessary to take account of the interchange of the tritium between the sea-water into which it is discharged and the water vapour in the atmosphere over the sea-water and the coastal zone. The water content of air at 15°C and a relative humidity of 70% is about 9 g/m³. Assuming complete exchangeability of water between the sea-water and the overlying airborne vapour, the airborne concentration of tritium in equilibrium with a seawater concentration of 5 pCi/l (corresponding to a discharge rate of 1 Ci/d) will be \(4.5 \times 10^{-5}\) µCi/m³. This figure may be compared directly with the maximum airborne concentration of tritium corresponding to the ICRP whole body dose limit of members of the public of 0.2 µCi/m³ \([7]\). By so doing we obtain the limiting environmental capacity for this route of exposure of \(4 \times 10^6\) Ci/d, a figure which is very similar to that for ingestion of contaminated foodstuff. However, it is undoubtedly pessimistic. It assumes that there is perfect mixing between sea-water and the atmospheric water vapour, and also that the air in the coastal region is always representative of that over the sea, which will not be so during, for example, periods of offshore winds. Nevertheless, these assumptions are regarded as adequate for radiological control purposes, and a limiting environmental capacity of \(2.5 \times 10^5\) Ci/d (9 \times 10^5 Si/a) is recommended to take account of all significant routes of exposure.
5.2. Collective dose

The collective dose cannot be dealt with quite so easily because of the lack of data on the fate in the longer term of the tritium released from Windscale. Detail is not available about either the eventual dispersion in sea-water or the true position regarding the degree of exchange of the tritium between sea-water and airborne water vapour. For the present purposes, however, it is sufficient to make some simplifying assumptions designed to give an upper limit for the collective dose from both routes of exposure. The collective dose from ingestion, for example, may be estimated by assuming that all the tritium discharged remains within the shelf waters of north-west Europe and that it affects only those populations who consume fish from those areas. In these circumstances it may be shown that for tritium being discharged at a constant rate into a water mass with mean depth, \( d \), and from which the rate of extraction of edible fish is \( F \) units of mass per unit area, the collective dose per unit input of activity is given by

\[
J = \frac{K \cdot F \cdot (1 - D)}{d \cdot \lambda \cdot \rho}
\]

where \( K \) is the factor relating dose to activity ingested, \( \lambda \) the radioactive decay constant for tritium, \( D \) is the dry/wet ratio of the edible fish flesh, and \( \rho \) is the density of water (Shepherd, personal communication). Solution of this equation with data appropriate to the shelf fisheries of north-west Europe gives a value for \( J \) of 2 \( \times \) \( 10^{-5} \) man-rem/Cl of tritium discharged. This is the dose integrated over all the time from the discharge of 1 Cl and represents the upper limit of collective dose from fish consumption. It is of course a very small dose, as may be expected for this nuclide which has very low radiotoxicity (cf. the corresponding figure for caesium-137 calculated by the same method of 0.6 man-rem/Cl discharged).

A value may be obtained by a similar procedure for the collective dose from the inhalation pathway. We assume that all the tritium discharged from Windscale spreads throughout the continental shelf waters around the United Kingdom, which are taken to have an area of \( 10^6 \) km\(^2\) and a mean depth of 0.1 km. If the only mechanism of loss of activity from the water is radioactive decay, it may be shown that with a constant rate of input of 1 Cl/d the equilibrium concentration (assuming uniform mixing) would be 6.4 \( \times \) \( 10^{-2} \) pCi/l. Assuming again that the water vapour present in the air above the sea contains tritium at the same concentration as the sea-water, then the tritium concentration of air at 15°C and 70% relative humidity may be calculated to be 6 \( \times \) \( 10^{-10} \) μCi/m\(^3\).

For the purpose of obtaining a first approximation to the collective dose via inhalation, it may be assumed that this is the concentration of tritium in the air which is breathed by the north-west European population as previously defined for the ingestion calculation. By this means the total rate of intake of tritium by the population may be related to dose by use of ICRP data. The report of Committee II [7] implies that continuous exposure to air containing 2 μCi/m\(^3\) leads to a whole body dose of 5 rem/a. The dose corresponding to the airborne concentration of 6 \( \times \) \( 10^{-10} \) μCi/m\(^3\) will be therefore 1.5 \( \times \) \( 10^{-9} \) rem/a. Thus under these conditions the annual collective dose to the population of north-west Europe will be 3 \( \times \) \( 10^{-1} \) man-rem/a. (The collective dose to the population of the UK only, calculated by the same processes, will be 8 \( \times \) \( 10^{-2} \) man-rem/a.) These figures relate to
the situation which would prevail under equilibrium conditions with a rate of discharge of 1 Ci/d. The total collective doses per curie discharged to the populations of north-west Europe and the UK integrated over all time are, therefore, $10^{-3}$ man-rem/Ci and $2 \times 10^{-3}$ man-rem/Ci respectively.

These estimates of collective dose for both ingestion and inhalation are without doubt conservative. They assume no loss of tritium other than by radioactive decay from the continental shelf waters into which the tritium is discharged, and they refer to the situation which would prevail under conditions of equilibrium. In view of the half-life of tritium ($12.28 \text{ a}$) there will be a significant loss from these coastal waters by dispersion well before equilibrium would be reached under the influence of radioactive decay alone. Nonetheless, even these simple pessimistic calculations of collective dose show that this factor is not likely to be limiting in the control of discharges of tritium to sea. The estimation may be used as a basis for guiding policy on the management of tritium in effluent. It may for example be used to calculate the detrimental costs of discharge to sea, a figure which may then be used as a basis for deciding how much it will be worth while spending to reduce sea discharges. Using a detrimental cost of £50 per man-rem, as derived previously in relation to the control of radioactive wastes by Shepherd and Hetherington [8], the detrimental cost per curie of tritium discharged will be £$10^{-3}$ and £$5 \times 10^{-2}$ via ingestion and inhalation respectively. Thus even with discharges of the order of $10^6$ Ci/a, the detrimental cost would not exceed £$5 \times 10^3$/a. There is no doubt that the magnitude of the reduction which could be achieved in discharges to sea by spending amounts of this order would be very small indeed.

REFERENCES

H. BONKA: As I showed in my own paper also, the best procedure at the moment is to discharge all $^3$H from reprocessing plants into the sea. If the processing plant is located inland it may not be possible to release all the $^3$H to the surface waters without exceeding the dose limits. Whether it is permissible to discharge all the $^3$H into the sea near the coast is a question of the mixing volume. How large is the water flow through the North Channel of the Irish Sea?

J.A. HETHERINGTON: Yes, the rate of dispersion of the radioactivity following its discharge into the sea is a crucial factor in determining the maximum acceptable discharge rate, and that is why it is important to establish the water concentration in equilibrium with unit discharge rate. The flow pattern of water through the North Channel is rather complicated and varies from time to time. Although I do not have the exact figure to hand, the long-term average flow rate is something like 2 to 3 km$^3$ daily, and the flushing time for the northern area (area III) of the Irish Sea is about one year.

J.A. GARLAND: I have two comments on your very interesting paper. First, I agree with you that the estimates of air concentration used in assessing the inhalation dose are probably very conservative. It may well require some hundreds of kilometres of fetch for the tritium concentration in the air to adjust to that in the water surface, and the fraction of the equilibrium value reached in a fetch of about 10 km should be rather small.

Secondly, the tritium carried inland by the wind is expected to deposit within some tens of kilometres and must enter surface run-off. A certain concentration must therefore be expected in drinking water, and also in the rivers, and this activity in rivers would have contributed to the apparent background subtracted from the field data. You appear to have ignored these effects, though I concede that they are probably very small.

J.A. HETHERINGTON: I take your point about drinking water, and it will be interesting to quantify the effect of tritium transfer from sea-water via evaporation into water used for that purpose; such calculations ought indeed to be done. But I should say at once that the (admittedly) limited amount of surface fresh water monitoring undertaken in the Windscale area tends to confirm my guess that the process, if it is occurring at all, is unlikely to produce a measurable effect on the existing concentrations due to weapons-test fall-out.

K.J. VOGLT: While the dose received by inhalation or from drinking water may be significant, I wonder whether the ingestion of food of vegetable origin upon which HTO has been deposited may not turn out to be the critical exposure pathway. Perhaps this should be investigated.

J.A. HETHERINGTON: In due course, if discharges increase substantially, it may be worth while measuring tritium in vegetation growing in the area.
However, on the basis of the discharge rate to the sea as practised up to now, it is doubtful whether tritium recycled from sea-water would be detectable in vegetation against the relatively high background from direct precipitation. In any event I do not expect that tritium in vegetation will ever be a critical exposure pathway.

P. BOVARD: Do you believe that good mixing conditions in the sea are achieved rapidly both in the northerly and the southerly directions?

J.A. HETHERINGTON: The degree of mixing seems to be about the same whether the general movement of the water is to the north or to the south, and there is no evidence of stratification.
OCEANIC DISTRIBUTION AND TRANSPORT OF TRITIUM

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Abstract

OCEANIC DISTRIBUTION AND TRANSPORT OF TRITIUM.
The tritium inventory (decay corrected to 1 Jan. 1972) tabulated from more than 70 stations in the Pacific Ocean shows approximately 59 kg of tritium in the North Pacific, 14 kg in the South Pacific (0° to 40°S), and a total oceanic inventory of 164 kg. This is expected from the estimated yield of atmospheric testing. The asymmetric distribution about the equator reflects the well-known fall-out pattern, and a source function has been formulated as an empirical equation representing the history of surface water tritium in the Pacific Ocean from 1963 to 1974. Unlike the Atlantic, in the Pacific there are no major areas of deep water formation. Thus, most of the bomb tritium is found shallower than 1000 m as it spreads from the source areas in the clockwise North Pacific gyre circulation, with the deepest penetration in the northwest Pacific. Tritium data used to model the exchange across the southern boundary of the North Pacific gyre are consistent with an upwelling rate for the equatorial region of 50 m yearly. The deep penetration of bomb tritium in some regions can be used to show that the Pacific Ocean is an important sink for excess atmospheric CO2.

GENERAL OCEAN CIRCULATION

Fifteen years have now passed since the major injection into the atmosphere of about 400 kg of tritium as water (HTO). During this time, half of it has decayed; and of the remainder, only about one fiftieth remains in the atmosphere [1], primarily in the upper stratosphere. The rest resides in the oceans and, to some extent, in larger groundwater aquifers.

The general circulation of the oceans is as follows: in the North Atlantic, surface waters drift northward, cooling and reaching the Greenland and Norwegian seas north of Iceland. There the surface density becomes equal to the underlying strata, and, especially in winter, very thick layers of deep water formation in the oceans. The newly formed water mass flows out around Iceland into the abyssal Atlantic Ocean. It mainly moves southward at great depths, and part of it eventually rounds the South
FIG. 1. Maps of the GEOSECS Atlantic and Pacific expeditions.
American continent, where it loses more of its identity and moves north in the Pacific Ocean. Another area where deep waters are formed is in the Antarctic Circumpolar Current. The time scale of this global circulation is on the order of hundreds of years, but the formation process and the first 5000 km, especially in the North Atlantic, can be followed with the tritium tag.

ATLANTIC TRITIUM TRANSPORT

A worldwide survey of the Atlantic and Pacific oceans as part of the GEOSECS (Geochromical Ocean Sections) program (see Figure 1) has resulted in profiles of tritium. These very clearly show the pattern of the newly formed deep water masses as they begin their travel into the bulk of the oceans. Figure 2 is a composite, north-south-depth section of the western Atlantic Ocean tritium data in 1972. It is perhaps the most striking illustration of the value of this bomb-produced tracer for studies of transport processes in the oceans on decadal time scales. The waters recently formed in the Norwegian and Greenland seas are seen flowing over the sill of the Denmark Strait toward the abyss; mixing into it are some shallower waters south of Iceland. The plume of tritium water (greater than 0.2 TU) marks the course of new deep water from its origin since 1957-1958, when the first substantial amounts of bomb tritium fell on the surface of the oceans. Incidentally, Figure 2 also illustrates the distribution pattern in the Atlantic of any other airborne, water-soluble pollutant produced in the northern hemisphere by the technologies of western civilization.

PACIFIC TRITIUM SECTIONS

A tritium section north-south of the western Pacific Ocean contrasts the difference between the Atlantic and Pacific oceans. Unlike the Atlantic, the Pacific section from 1973 to 1974, Figure 3, shows that there is no plume of high tritium sinking in the north and spreading toward the abyss. On the contrary, most of the bomb tritium is still found in the upper waters of the Pacific, which circulate in a clockwise gyre in the north and counterclockwise gyre in the south. The North and South Equatorial currents form boundaries of the two gyres, and between them is the eastward-flowing North Equatorial Counter Current and at the equator the Equatorial Undercurrent.

Figure 3 shows that in the northern part of the North Pacific gyre there are the highest values and the deepest penetration of bomb tritium (to 1000 m). In the center of the northern gyre (20° to 40°N), the waters are fairly well mixed, especially above 500 m, with very uniform tritium distribution. The southern boundary of the northern gyre is the North Equatorial Current (NEC), located from about 8° to 20°N; it shows a subsurface tritium maximum, which was also shown in earlier data by Michel and Suess [2]. The maximum is a result of waters sinking at higher latitudes, and finding their way south under less dense water. In over-simplified terms, the ocean waters move horizontally along surfaces of constant density, and superimposed on this is upwelling and downward diffusion. The signs of upwelling are particularly strong at the equator, where the constant tritium, or 'isotrit', surfaces slope upwards. This occurs partially because they follow the constant density surfaces which slope up, and partially due to mixing with upwelled water of lower tritium. Figure 3 also shows the considerably lower tritium in the equatorial regions and the South Pacific.
FIG. 2. Tritium section of the western Atlantic in 1972 from 70° N to 50° S versus depth (km). Vertical exaggeration is 2000:1. Horizontal scale is proportional to cruise track.
Figure 4 is a tritium section north-south of the eastern Pacific Ocean along 125°W in 1974. It has the same general features as the section of the western Pacific: a subsurface tongue of higher tritium, 'isotrit' surfaces sloping upwards at the equator, and lower tritium in the south (less than 3 TU). The difference between the eastern and western sections is particularly apparent in the North Pacific. Figure 5 shows the longitudinal tritium variation in a section, east-west at about 30°N in 1974. In winter in the northwest Pacific there is deep convection and mixing of shallow higher tritium waters. The uppermost waters in the eastern Pacific are low in salinity, thus considerably less dense than the deeper waters. This causes the water column in the east to be more stable and thus much more resistant to vertical mixing. The result is deep penetration of tritium in the west and shallower penetration in the east, but no discharge into the abyss.

HISTORY OF PACIFIC SURFACE TRITIUM

The asymmetric distribution of tritium about the equator is a consequence of the well-known fallout pattern. From GEOSECS data and tritium data collected starting in 1959 by Suess and Dockins and published by Dockins et al. [5] and Michel and Suess [2], Fine and Östlund [4] formulated an empirical equation to describe the time history of bomb tritium in the surface waters of the Pacific Ocean. The collection of data showed an exponential variation of tritium with latitude increasing south to north. Figure 6 shows tritium versus time (1963 to 1974) for Pacific Ocean surface water as predicted by the source function at 50°N, 30°N, and the equator. The maximum occurs in late 1965, two to three years later than the maxima in the rain data. The lag in the oceanic tritium maximum can be attributed to the length of time it takes tritium to build up in the mixed layer. Thus, since there is a two to three year stratospheric residence time for tritium, the decrease in ocean concentration after 1966 occurred because of the rapidly decreasing input. For many oceanographic purposes one can assume that all HTO was added in 1962-1963.

USE OF TRITIUM TO MODEL EQUATORIAL EXCHANGE

The tagging of the higher northern latitude waters with tritium have made it a unique oceanographic tool with which to examine short time scale mixing. For example, Figure 4 reveals a tongue of northern latitude higher tritium extending from the region of the northern gyre (NEG) into the North Equatorial Counter Current (NECC) to around 6°N. This suggests considerable exchange between the two regions. A two-box model was used by Fine and Östlund [5] to estimate the exchange between the NEC and NECC. The model results suggested that the exchange time is consistent with an equatorial upwelling rate of 50 m/yr, which is a meridional transport of water from the northern gyre into the equatorial region. This transport is balanced by a return surface flow to the north, manifested by the lower tritium at shallower depths. Thus, the equatorial region restricts the exchange of tritium water between the northern and southern oceans.

OCEANIC TRITIUM INVENTORY

A tritium inventory (decay corrected to 1Jan.1972) has been tabulated from more than 70 stations in the Pacific Ocean, which were occupied as part of GEOSECS. Most of the tritium was found in the North Pacific, approximately 39 kg. Only 14 kg of tritium were found in the
FIG. 3. Tritium section of the western Pacific, 1973 to 1974, from 50°N to 65°S versus depth (m). Vertical exaggeration is 2500:1.

FIG. 4. Tritium section of the eastern Pacific in 1974 at 125°W from 25°N to 40°S versus depth (m). Vertical exaggeration is 2500:1.

FIG. 5. Tritium section of the North Pacific in 1973 at 30°N from 130°W to 145°E. Vertical exaggeration is 2500:1.
South Pacific (0° to 40°S). The estimate of 73 kg of tritium in the Pacific Ocean as of 1 Jan. 1972 is 34 kg lower than the estimate by Michel [6] for the year 1970. The discrepancy is probably due to the availability of many more vertical profiles from GEOSECS.

Figure 7 is a map of the tritium inventory in units of kTUs, tritium units times kilometer depth. The northwest Pacific has the highest vertical column inventories, up to 5.5 kTUs at a station near 35°N, 150°E. Values greater than 5 kTUs are commonly found at stations in this region because of the deep penetration of tritium. As shown by Figure 5, the winter convection in the northwest has helped to mix tritium to the greatest depths found in the entire Pacific (north of 40°S), thus measurable tritium (greater than 0.2 TU) is found to extend down below 1000 m in this region. Horizontal exchange and vertical diffusion have spread the deep tritium into the central gyre region where bomb tritium is found to 800 m. In general, north of 10°N tritium is found to at least 400 m, giving inventories on the order of 2 to 3 kTUs. As a consequence of exchange with the higher northern latitudes, the region just north of the equator has received considerable bomb tritium, resulting in at least 1 kTUs. On the time scales being examined, the South Pacific has a low inventory of generally 0.6 to 0.8 kTUs, because of the source function and little exchange of water across the equator. Low values are also found in the eastern tropical Pacific, because the northern gyre circulation carrying the high tritium bypasses this region.
FIG. 7. Contours of kTUm (decay corrected to 1 Jan. 1972) superimposed on the GEOSECS Pacific expedition map.
TABLE I. OCEANIC TRITIUM INVENTORY 1972

<table>
<thead>
<tr>
<th>Ocean</th>
<th>kg T</th>
</tr>
</thead>
<tbody>
<tr>
<td>North Atlantic</td>
<td>66</td>
</tr>
<tr>
<td>South Atlantic</td>
<td>7</td>
</tr>
<tr>
<td>Arctic</td>
<td>6</td>
</tr>
<tr>
<td>North Pacific</td>
<td>59</td>
</tr>
<tr>
<td>South Pacific</td>
<td>14</td>
</tr>
<tr>
<td>Antarctic</td>
<td>6</td>
</tr>
<tr>
<td>Indian</td>
<td>6</td>
</tr>
<tr>
<td>Total Oceanic</td>
<td>164</td>
</tr>
</tbody>
</table>

Tritium data from the GROSECS Atlantic expedition, from other expeditions in the North Atlantic, and recent data from the two polar regions and the Indian Ocean have also been examined. The best estimate from the available data of an oceanic tritium inventory is presented in Table I. The total of 164 kg as of 1 Jan. 1972 is much as expected from the estimated yield of atmospheric testing. The inventory of the North Atlantic exceeds that of the North Pacific by 7 kg, even though the volume of the North Pacific is considerably greater. Tritium from continental runoff has added a considerable amount to the North Atlantic giving the total of 66 kg.

CONCLUSION

Tritium is a unique tracer, because it is in a transient state. It is clearly predicting where and how fast manmade pollution will travel in the oceans. From the tritium profiles, we have an estimate of the water that has been in direct contact with the atmosphere in the period 1963 to 1972, and thus, potentially contains 'excess CO₂' [7]. For this reason, tritium is an important tool with which to study the upper layers of the ocean, which have such a large impact on the world climate. In conclusion, a plea is made for continued measurements of tritium in the ocean on a worldwide basis. Furthermore, it is hoped that the potential refinement in measurement technique offered by the mass-spectrometric ³He/³H method will mean an extension of this tool to more areas and longer time scales in the oceans.

DATA BASE

The Miami Tritium Laboratory uses the combination of electrolysis and gas counting, described by Östlund and Dorsey [8]. Our oceanographic data are available from the senior author in tabular form in reports [9 to 12].
ACKNOWLEDGEMENTS

This work is supported by grants from DOE (Office for the International Decade of Ocean Exploration) of the U.S. National Science Foundation. The GEOSECS Operations Group in La Jolla, California, prepared the maps and material in forms suitable for making the sections.

REFERENCES


DISCUSSION

R.V. OSBORNE: In your conclusions you made a passing reference to the mass spectrometric method. Are you proposing to use this method in the future?

H.G. ÖSTLUND: Yes, we would like to do so and hope to be able to set up the necessary apparatus in the not too distant future; I may mention that the
mass-spectrometric method is being developed by B. Clarke, McMaster University, Hamilton, Ontario, and by W. Jenkins, WHOI, Woods Hole, Massachusetts. It involves the measurement of the rate of regrowth of $^3$He in a de-gassed water sample. The method is potentially superior to electrolysis/gas counting in sensitivity and accuracy.

L. FARGES (Scientific Secretary): As you will be aware, the IAEA is involved in the definitions of the rules to be observed within the framework of the London Dumping Convention. An important aspect is the turnover of water from the bottom (i.e. from depths of 4000–5000 m) to the surface. Do you think that tritium could be used to define the transit time for this process? If so, what would this transit time be?

R. KIRCHMANN: As a supplement to Mr. Farges’ question, I should like to ask whether there might not be a shorter pathway of transfer of radioactive material from the deep waters of the North Atlantic to the surface waters of the continental shelf, by upwelling due for example to barometric variations.

H.G. ÖSTLUND: The tritium at present in the oceans may not be the ideal tracer for this purpose and combined tracer studies ($^{14}$C, tritium, $^{85}$Kr) are more likely to yield an answer. There is, however, no simple answer to the question, since upwelling is a local phenomenon, occurring along the Equator and along the western edge of the oceans. Moreover, it seems likely that even in upwelling areas one would have to wait 50–100 years for ‘new’ top water to appear. By far the larger part of the deep waters would reside below the euphotic zone for 200 years or more.

W. ROETHER: As Mr. Östlund has just pointed out, tritium is not really the right tracer for the purpose in view, but rather $^{14}$C, and also perhaps $^{226}$Ra and radon-222. I may add that we are preparing some work in the northeast Atlantic, using the above-mentioned tracers, in order to study the movement and mixing of the deep and near-bottom waters in that area.

J.K. MIETTIEN: According to your data there are now about 164 kg of tritium in the oceans. On this basis it should be possible to calculate the total amount of tritium that has been produced. Can you give an estimate in MCI?

H.G. ÖSTLUND: We have not made this calculation to the desirable degree of accuracy. We do not have the continental inventory, and furthermore the upper stratosphere may still contain 20–40 kg of tritium, as Mr. Mason indicated at the beginning of Session I of this Symposium. Within these limits, however, our estimate would not contradict that of Miskel made at the 1971 Symposium at Las Vegas, Nevada.

J.A. HETHERINGTON: In your surveys of the North Atlantic, how far westwards did you sample? Did you sample in the Norwegian Sea, and if so, did you discover any anomalous results in this area?
H.G. ÖSTLUND: Both the Norwegian and Greenland seas are source areas for new water. They are separated at a certain depth by about a 1000-metre ridge as I recall, but they function with different residence times as new water areas. We have not observed anything out of the ordinary; for example we have not seen any Windscale tritium up there, and I don't expect that we shall.

L. MERLIVAT: Regarding the inventory of the tritium released by nuclear weapons tests, I believe that, besides the main part which is taken up by the oceans, an important fraction is retained in the ice caps, mainly the Antarctic cap. They constitute a vast reservoir of water, and also show large tritium concentrations. This fraction may possibly be more important than that stored in groundwater and in the stratosphere.

H.G. ÖSTLUND: Yes, I believe that this is substantially correct.
TRITIUM IN THE NORTH ATLANTIC OCEAN

Inventory, input and transfer into deep water

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Heidelberg,
Federal Republic of Germany

Abstract

TRITIUM IN THE NORTH ATLANTIC OCEAN: INVENTORY, INPUT AND TRANSFER INTO DEEP WATER.

The tritium inventory of the North Atlantic was found to be 665 MCI (± 8%) at the end of 1972, on the basis of the available North Atlantic tritium observations (depth profiles). Very nearly the same amount of tritium is calculated to have been delivered to the North Atlantic. Delivery by water vapour exchange is almost four times that by precipitation, and of the total delivered about 10% is by river run-off and 15% by net tropospheric tritium inflow from the continents. The calculation uses the hydrological data of Baumgartner and Reichel, the distribution of tritium in precipitation taken from the IAEA compilations, and the fact (for which evidence is presented) that tritium in water vapour at ship's height is near isotopic equilibrium with that of precipitation. The tritium time lag in river run-off is also accounted for. The data on which the calculation is based are presented in tables. The delivery calculation scheme can be used to predict future tritium uptake by the North Atlantic. A strontium-90 delivery of 2.8 MCI is estimated from the tritium delivery. The tritium deposition data are used to determine the volumes of the North Atlantic two-box mixing model of Dreisigacker and Roether, which has been devised to simulate the response of the near-surface ocean to the tritium and strontium-90 input. The result is an average depth of the two boxes combined of 720 m (20°–60°N), and a rate of water transfer into the deep ocean of 10.6 × 10^8 m³/s. This rate is compatible with oceanographic estimates. The volume-calibrated model can be used to predict the response of the North Atlantic surface water to input from the atmosphere on a year-to-decade time scale. As examples of this, the response to an addition in a single spike as well as to a constituent having an exponential concentration increase in the atmosphere (e.g. CO₂) is presented.

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1. INTRODUCTION

A quantitative assessment of the uptake of tropospheric tritium into the ocean, and of its transfer from the ocean's surface into the interior, for the North Atlantic, is presented in this report. Knowledge on these processes is necessary for estimating future environmental burdens of tritium, because the ocean, as has long since been recognized [1], is the predominant sink for atmospheric tritium. Furthermore, knowledge of the transfer of tritium within the ocean can be applied to predict the dispersion of other oceanic pollutants added to the ocean on a years-to-decades time scale. A third aspect is that the temporal and areal pattern of the oceanic tritium uptake must be known if the dynamics of the internal oceanic transport processes are to be studied on the basis of the observed oceanic tritium distribution.

The subject is approached by presenting an inventory of the North Atlantic, based on observed tritium versus depth profiles. In a second step, the total tritium delivery to the N. Atlantic is calculated by means of a hydrological model. These parts of the paper are based on unpublished work of one of the authors [2]. From the fact that the inventory and the delivered amount of tritium agree, we conclude that we understand in a quantitative way the oceanic tritium uptake. Lastly, this information is used to determine the transfer of tritium, which at the same time is that of N. Atlantic near-surface water, into deeper layers of the N. Atlantic. This is done by an extension of a previously formulated N. Atlantic mixing model [3]. In setting the borders of the N. Atlantic we follow Baumgartner and Reichel [4] (i.e. inclusive marginal seas, borders to the Arctic Ocean at about 80°N between 90°W and 60°E), except that we exclude the Mediterranean.

2. THE NORTH ATLANTIC TRITIUM INVENTORY

The tritium inventory is calculated on the basis of measured tritium versus depth profiles in the N. Atlantic. About 180 such profiles, comprising a total of 2700 tritium measurements, are available [5—15]. The tritium concentrations of all these profiles have individually been integrated vertically, to give local inventories (in T·U·m or C/m³). Vertical resolution and particularly deep water coverage varies greatly among the stations. Non-linear interpolations between data points, on the basis of general oceanographic knowledge and in analogy with more detailed profiles, were used for several of the profiles. The local inventories were normalized to a common reference time, i.e. end of 1972. The normalization accounts for local tritium input from the atmosphere and in-situ tritium decay, between reference time and time of sampling, but neglects changes due to horizontal tritium transport. The procedure was to multiply the inventories by the
TABLE I. YEARLY AVERAGES OF TRITIUM CONCENTRATIONS AND
TIME-INTEGRATED TRITIUM SUPPLIES, 1952 to 1972

Tritium concentrations \( c_p \) (precipitation), \( c_r \) (river-run-off) both in 50°N, \( c_b \) North
Atlantic surface water 20°–60°N (concentrations 0°–20°N are about half these
values [3]). Tritium supplies, \( S \), are defined as
\[
S(t) = \int_{1952}^{t} c(t') \exp \lambda (t' - t) \, dt',
\]
with \( \lambda = \) tritium decay constant = 0.05635/year. For explanations see text.

<table>
<thead>
<tr>
<th>Year</th>
<th>( c_p )</th>
<th>( c_r )</th>
<th>( c_b )</th>
<th>( S_p )</th>
<th>( S_r )</th>
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ratio of the time-integrated tritium supplies (Table 1, column 5; see text below) for the reference time to that for the time of sampling. The corrections amount to no more than 16% for any of the profiles.

Figure 1 shows the normalized local inventories, plotted as a function of latitude, the station positions being shown in the inset chart. The inventories exhibit a pronounced latitudinal variation, with values of about 8000 TU·m⁻¹ (26 μCi/m²) north of 35°N and values up to a factor of 20 lower in equatorial latitudes. Much of the northward increase is concentrated near 15°N, i.e. in the frontal zone between the N. Atlantic Central Water and equatorial waters [16]. The western and eastern N. Atlantic basins are equally well documented. Whereas the inventories in general appear to be somewhat larger for the western basin stations, the differences are comparable with the data scatter in Fig. 1 and are not considered further.
The total N. Atlantic tritium inventory has been calculated by computing
the average (and standard deviation\(^1\)) of the local inventories in 5 deg latitude
bands (10 deg bands for 50\(^0\)–80\(^0\)N), and by multiplying these figures by the
N. Atlantic surface areas in the respective bands. Summing up the band contribu-
tions, and using error propagation for the uncertainties, the total N. Atlantic
inventory becomes 722 MCI ± 4%. A correction is needed, because in the tritium
profiles on which Fig. 1 is based, the depth below which still half the tritium
is found is between 200 m in equatorial latitudes and 1000 m in the northern
N.W. Atlantic, and on the other hand the profiles tend to be from deep ocean
areas, so that the calculation above over-estimates the inventories in shallow water
areas. Shelf (0 to 200 m) and continental slope (200 to 1000 m) areas for the
N. Atlantic, with the borders at present used, are about 15% and 9% [17]. It can
be assumed that the Baltic and part of the Canadian Archipelago sea area and
perhaps also of the Barents Sea are sufficiently separated so that they still contain
most of the tritium they have received. Therefore, for these areas the open-ocean
figures should not be too much of an over-estimate. Taking this into consideration,
we correct the inventory down by 8 ± 5%. This correction brings the largest
uncertainty into the total inventory estimate. Furthermore, we account for the
uncertainty brought about by tritium measurement blanks. For the Heidelberg
tritium data, the 1-sigma blank uncertainty, \(\delta_\text{b}\), is no more than ± 0.5% of the
surface water concentrations, \(c_\text{b}\), north of 20\(^0\)N [18]. The resulting error for the
inventories can be calculated for an exponential concentration decrease with depth
to be \(\sim \delta_\text{b}/c_\text{b} \cdot \ln (c_\text{b}/\delta_\text{b})\) or about 3%. It is assumed that this is a fair approximation,
and that the data of the only other reporting laboratory (Miami, see Ref. [5]) have
a similar blank uncertainty. Accounting for the fact that at lower latitudes the
blank makes a larger effect, the resulting error for the total inventory is taken to
be ± 4%. Intercalibrations between the Miami and Heidelberg laboratories [19, 20]
have shown that the tritium scales agree to better than ± 3%.

Applying the correction, and making allowance for the additional uncertainties,
the total North Atlantic tritium inventory is found to be 665 MCI ± 8%. A similar
value has been deduced by Östlund and Fine [21] who give 640 MCI for the
N. Atlantic (= 66 kg tritium; borders of the N. Atlantic and uncertainty of the
estimate not specified), whereas a previous estimate for the total Atlantic [39]
is certainly too high.

3. THE NORTH ATLANTIC TRITIUM DELIVERY

The delivery calculation starts with the tritium pattern in precipitation,
which is presented in a factorized form of a temporal and a geographical variation.

\(^1\) Standard errors are given throughout this work.
FIG. 2. Tritium concentrations in northern hemisphere precipitation relative to a reference station (long-term mean ratios, reference station is Valentina, 51°N, 10°W, see text) plotted versus latitude. ■ oceanic weather ships, □ island stations, ○ coastal stations, ○ N. American and W. African continental stations. Solid line is a best fit of the data (squares); dashed line is the linear extrapolation of the best straight line towards latitudes which have not been included in the fitting procedure. Inset chart gives station locations.

This is followed by proof that the tritium concentration of water vapour at ship’s height is in near isotropic equilibrium with that of falling precipitation. The tritium concentration pattern is then converted into delivery rates (via rain-out and vapour exchange) by means of a hydrological model, using the hydrologic-cycle data of Baumgartner and Reichel [4]. Tritium input from the continents by river run-off and net tropospheric inflow is added to this. The final evaluation yields 693 MCi total delivery.

3.1. The tritium pattern in North Atlantic precipitation and tritium in marine water vapour

About 2000 monthly composite precipitation data have been available since the late 1950s, collected at up to 40 monitoring stations on oceanic weather ships
FIG. 3. Tritium concentrations (in TU) of samples, simultaneously collected approximately weekly, of marine water vapour $c_v$ and precipitation, $c_p$, taken aboard F.S. "Meteor" during 1965 to 1973. $\alpha = \text{tritium separation factor, see text, } c_s \text{ is simultaneously measured surface-water concentration.}$

(OWS), small islands and at coastal sites [22, 23]. The general pattern of tritium in N. Atlantic precipitation was deduced from these data, in a manner described previously [2, 24, 25]. It is based on the fact that tritium concentrations at any two monitoring stations have varied in near-constant proportions through the years, so that the tritium pattern can be factorized into temporal and areal distribution functions (see also Ref. [3]). Detailed evidence of this for the restricted region of western and central Europe has been presented in another paper in this Symposium [26], and the finding apparently also holds over much larger distances: Yearly mean ratios of tritium concentrations at the stations Ponta Delgada (~38°N, 27°W), OWS E (35°N, 48°W), San Juan (19°N, 66°W), and even in the Pacific at OWS V (31°N, 164°E), to those at the station Valentia (51°N, 10°W; Valentia is used as the reference station in Refs [3, 26], show a year-to-year scatter of no more than ± 20% with no apparent time trend, compared with a variation of a factor of 20 in the yearly mean concentrations for each individual station.
The areal and temporal tritium distributions in precipitation, as used in the present work, are summarized in Fig. 2 and Table I. Figure 2 shows mean ratios of tritium concentrations at the monitoring stations mentioned, and at additional continental stations, to those at Valencia, plotted as a function of latitude. The straight line, which has been fitted to the data points which supposedly represent open-ocean conditions (squares), is taken for the average open-ocean tritium-versus-latitude relationship. The apparent single-station scatter is no larger than \( \pm 15\% \) north of 20\(^\circ\)N. The temporal distribution for the reference station Valencia (taken from Refs [3, 26]) is given in the form of annual means in Table I, column 2. The product of the ratio read from Fig. 2 for the specific latitude and the annual mean concentration taken from Table I, column 2, thus is our tritium concentration estimate for any open-ocean location and year. The continental stations in Fig. 2 typically plot a factor of 4 or so higher than the oceanic ones. The effect of this on the oceanic delivery is considered in Section 3.3.

Figure 3 presents a correlation of tritium concentrations in marine water vapour with that of simultaneously fallen precipitation for composite samples of typically one-week sampling periods. The samples were collected during several cruises of the F.S. "Meteor", 1965–73. Precipitation was sampled by conventional gear, and vapour (intake \(~ 15\) m above the sea surface) by means of a collecting unit, designed by one of the authors (W.R., see also Ref. [27]), which is based on quantitative absorption/desorption on activated charcoal in a periodically reversing arrangement. Whereas the individual data points show a considerable scatter — as expected, because the vapour was sampled continuously whereas the precipitation was naturally quite intermittent — the vapour on the average is near to isotopic equilibrium with precipitation. In quantitative terms, one may regard the vapour as consisting of two fractions, derived from precipitation and surface water, i.e.

\[
c_v \cdot \alpha = f \cdot c_p + (1 - f) \cdot c_s
\]  

where \( c_v, c_p, c_s \) = tritium concentration (TU) of vapour, precipitation, and surface water; \( \alpha \) = tritium separation factor between liquid water and water vapour (\( \alpha = 1.1 \) [1]).

Figure 3 shows that, on the average, \( f \), the precipitation-derived fraction, is very nearly unity, i.e. the marine water vapour at ship's height is practically in isotopic tritium equilibrium with the precipitation.

3.2. The hydrological model

Deposition of tritium from the atmosphere on to the ocean surface occurs by both rain-out and water vapour exchange [28–30]. The latter process arises
from water molecules crossing the air-sea interface in both directions. Whereas the humidity gradient above the water surface is such that a net upward flux of vapour occurs, i.e. the evaporation E, the tritium gradient is opposite so that there is a net transport into the ocean, i.e. the vapour exchange tritium transport.

The tritium deposition rate, d (in TU·m/a), can be written as

\[ d = P \cdot c_p + E \cdot \frac{h}{1-h} \cdot c_v - E \cdot \frac{1}{\alpha(1-h)} \cdot c_s \]  

(2)

where \( P, E = \) precipitation, evaporation (m/a); \( h = \) humidity relative to phase equilibrium at the air-water interface.

The second and third term on the right hand side are the in- and outgoing fractions of the vapour exchange [2, 28, 29]. Inserting Eq. (1) and neglecting higher-order terms in \( c_s \), one obtains

\[ d = \left[ P + E \cdot \frac{f \cdot h}{\alpha} \cdot 1-h \right] \cdot c_p - E \cdot \frac{1}{\alpha(1-h)} \cdot c_s \]  

(3)

Since \( f, \alpha \approx 1, \bar{P} \approx \bar{E}, h \approx 0.75, \) and \( \bar{c}_s \ll \bar{c}_p, \) it follows that the second term dominates over the first, i.e. that vapour exchange is much larger than rain-out, and that the last term is a smaller correction only.

The total tritium deposition, \( D \) (in TU·m), at a set location up to the time of observation, \( t_0, \) corrected for the tritium decay before observation, is

\[ D(t_0) = \int_{-\infty}^{t_0} d(t) e^{-\lambda(t_0-t)} dt \]  

(4)

The meteorological data (\( E, P, h \)) being taken as constant in time, the integration only refers to the tritium concentrations. These integrals, termed time-integrated tritium supply, \( S, \) are given in Table I, columns 5 and 7. The total deposition, therefore, is

\[ D(t_0) = \left[ P + E \cdot \frac{f \cdot h}{\alpha} \cdot 1-h \right] \cdot S_p(t_0) - E \cdot \frac{h}{\alpha(1-h)} \cdot S_s(t_0) \]  

(5)

The tritium supplies of Table I refer to 50°N. For other latitudes the correction read from Fig. 2 is applied.
TABLE II. SUMMARY OF QUANTITIES USED TO CALCULATE THE NORTH ATLANTIC TRITIUM DELIVERY

E = evaporation, P = precipitation, \( R_t \) = continental run-off, from Ref. [4], \( R_t \) = tropospheric water vapour inflow, 
\( c/c_{Vd} \) = tritium concentration ratio (Fig. 2), \( D_{E,P} \) = tritium deposition (Eq. (5)), \( A \) = North Atlantic area of 5 deg latitude bands [4], \( I \) = tritium input in latitude bands: for explanations see text

<table>
<thead>
<tr>
<th>Latitude</th>
<th>Band °N</th>
<th>E (m/s)</th>
<th>P (m/s)</th>
<th>( R_t ) (km²/s)</th>
<th>( R_v ) (km²/s)</th>
<th>( c/c_{Vd} )</th>
<th>( D_{E,P} ) (TU m)</th>
<th>A (10^4 km²)</th>
<th>( I_{E,P} ) (MCi)</th>
<th>( I_r ) (MCi)</th>
<th>( I_r ) (MCi)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>0–5</td>
<td>1.20</td>
<td>1.45</td>
<td>2345</td>
<td>0.10</td>
<td>653.9</td>
<td>3595</td>
<td>7.5</td>
<td>2.5</td>
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<td>5–10</td>
<td>1.33</td>
<td>1.69</td>
<td>2130</td>
<td>0.13</td>
<td>1039.6</td>
<td>2796</td>
<td>9.3</td>
<td>3.2</td>
<td>28.5</td>
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<tr>
<td>10–15</td>
<td>1.46</td>
<td>1.01</td>
<td>654</td>
<td>0.17</td>
<td>1157.2</td>
<td>3857</td>
<td>14.3</td>
<td>1.3</td>
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<tr>
<td>15–20</td>
<td>1.53</td>
<td>0.68</td>
<td>336</td>
<td>0.22</td>
<td>1535.5</td>
<td>4176</td>
<td>20.5</td>
<td>0.9</td>
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<td>20–25</td>
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<td>0.52</td>
<td>100</td>
<td>0.29</td>
<td>1925.0</td>
<td>4547</td>
<td>28.0</td>
<td>0.4</td>
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<tr>
<td>25–30</td>
<td>1.53</td>
<td>0.64</td>
<td>640</td>
<td>0.36</td>
<td>2615.5</td>
<td>4522</td>
<td>37.8</td>
<td>5.9</td>
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<tr>
<td>30–35</td>
<td>1.62</td>
<td>0.63</td>
<td>176</td>
<td>0.46</td>
<td>3636.8</td>
<td>3629^a</td>
<td>42.2</td>
<td>1.0</td>
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<tr>
<td>35–40</td>
<td>1.53</td>
<td>0.82</td>
<td>150</td>
<td>0.57</td>
<td>4645.9</td>
<td>3119^a</td>
<td>46.4</td>
<td>1.2</td>
<td>43.1</td>
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<tr>
<td>40–45</td>
<td>1.19</td>
<td>1.00</td>
<td>180</td>
<td>0.71</td>
<td>5049.0</td>
<td>3415^a</td>
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<tr>
<td>45–50</td>
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<td>810</td>
<td>0.86</td>
<td>5646.6</td>
<td>2562</td>
<td>46.3</td>
<td>7.8</td>
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<td></td>
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<tr>
<td>50–55</td>
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<td>1.18</td>
<td>917</td>
<td>1.02</td>
<td>6504.8</td>
<td>2303</td>
<td>47.9</td>
<td>12.6</td>
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<tr>
<td>55–60</td>
<td>0.77</td>
<td>1.02</td>
<td>518</td>
<td>1.19</td>
<td>6430.1</td>
<td>2588</td>
<td>53.3</td>
<td>8.4</td>
<td>25.7</td>
<td></td>
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<tr>
<td>60–65</td>
<td>0.59</td>
<td>0.97</td>
<td>566</td>
<td>1.36</td>
<td>6163.3</td>
<td>2230</td>
<td>44.4</td>
<td>10.7</td>
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<tr>
<td>65–70</td>
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<td>637</td>
<td>1.52</td>
<td>4580.1</td>
<td>2100</td>
<td>32.0</td>
<td>13.6</td>
<td></td>
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<tr>
<td>70–75</td>
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<td>0.34</td>
<td>143</td>
<td>1.67</td>
<td>3673.0</td>
<td>2116</td>
<td>24.9</td>
<td>3.4</td>
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<td></td>
</tr>
<tr>
<td>75–80</td>
<td>0.18</td>
<td>0.26</td>
<td>83</td>
<td>1.79</td>
<td>2369.5</td>
<td>1253</td>
<td>9.5</td>
<td>2.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0–80</td>
<td>1.21</td>
<td>0.87</td>
<td>10385</td>
<td>7800</td>
<td>48908</td>
<td>519.5</td>
<td>76.6</td>
<td>97.3</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\[ \sum = 693.4 \text{MCi} \]

^a Excludes the Mediterranean.
3.3. Tritium addition from the continents

The two processes considered are river run-off and the addition by tritium-rich continental vapour that is advected into ocean areas. The run-off contribution is estimated as follows. The relationship of tritium concentrations in river run-off to that of precipitation in the catchment area is taken to be that observed for the Rhine (Western Europe [31]). This relationship is the result of delay and dispersion of precipitation in the ground before its reappearance in the river, and of a general reduction in tritium concentration owing to seasonal selectivity of continental evapotranspiration. River concentrations according to this concept typical for a catchment area receiving Valencia precipitation and the corresponding time-integrated tritium supplies, are listed in Table I, columns 3 and 6. The geographical variation in tritium concentrations was accounted for by applying the general latitudinal variation given in Fig. 2 (straight line), corrected upwards by a factor of 4 for the general marine-continental difference in tritium concentrations (see Fig. 2 and Ref. [26]). The tritium run-off is then the product of the thus determined time-integrated tritium supply for continental river run-off for any latitude (the latitude of the river mouth being taken as the relevant latitude), and the run-off (in m$^3$/a). The procedure has been checked for several rivers where measured run-off tritium concentrations are available, and satisfactory agreement has been found (within $\pm 20\%$ generally, for details see Ref. [2]). The net tropospheric tritium inflow was calculated from published transport rates of vapour across the continental borders [32–35], and the excess of continental over marine tritium concentrations in precipitation (i.e. latitudinal variation of Fig. 2 and a factor of 3 for excess continental concentration), details are again found elsewhere [2].

3.4. Evaluation

The evaluation is summarized in Table II. Long-term averages of evaporation, precipitation and continental run-off, in 5 deg latitude bands, as well as the net tropospheric inflow of water vapour, are listed in columns 2 to 5. The deposition D by rain-out and vapour exchange in column 7 was calculated from Eq. (5) and the relative tritium concentration in column 6. The value for h (Eq. (2)) was chosen as 0.755, which is the average of published values for the N. Atlantic [36–38]. Multiplying by the areas in column 8 gives the inputs of column 9. The inputs by river run-off and vapour inflow in columns 10 and 11 were obtained by multiplying the amounts in columns 4 and 5 with the respective tritium supplies (Table I, columns 5 and 6), and the tritium concentration ratios in column 6 raised by factors of 4 and 3.
A total tritium delivery to the N. Atlantic of 693 MCI results, of which 75\% is by open-ocean rain-out/vapour exchange, and the remainder about equally contributed by river run-off and net tropospheric tritium inflow from the continents. The tritium deposition by vapour exchange is almost four times that by rain-out. The uncertainties in these inputs are difficult to assess. As discussed in detail elsewhere [2], the error in the rain-out/vapour exchange deposition should be $\lesssim \pm 20\%$, the largest source of uncertainty being the humidity $h$ via Eq. (3). The relative errors of the other two delivery mechanisms are certainly larger than this figure, but because their contributions are small, and presumably the errors for the various contributions largely independent, the error of the total delivery should not exceed $\pm 20\%$. It should be mentioned that the quantities in Eq. (3) all show a seasonal variation, the correlation of which has to be considered. It turns out, however, that seasonal effects very nearly cancel out [2], so that the use of yearly mean values is justified.

4. COMPARISON OF INVENTORY AND DELIVERY

The 5 deg latitude band values of the total tritium deliveries (Table II) are entered as the histogram into Fig. 1. The agreement with the observed local inventories is good, but somewhat accidental because the tritium certainly has undergone latitudinal displacement during the period between input and observation. The explanation offered for the apparent agreement is that the northward increase of the deposition (histogram) is largely paralleled by an increase in the depths down to which tritium has been carried by oceanic mixing. A result of such near correspondence of deposition and mixing depths is that the surface water tritium concentrations are rather uniform areae in the N. Atlantic north of $20^\circ$N [3]. It also follows that other industrial pollutants should show a similar distribution in the N. Atlantic, because they should also have maximal input in mid to higher latitudes ($\sim 40^\circ$ to $60^\circ$N).

Because of the latitudinal tritium displacement, a comparison of inventory and delivery, in order to be rigorous, must be made on the N. Atlantic totals. Such a comparison only requires a minor correction for net tritium transport across the N. Atlantic marine boundaries. Neglecting transport at the northern boundary, there remains inflow of South Atlantic surface water ($\sim 6 \cdot 10^6$ m$^3$/s [39], yielding $\sim 20$ MCI) and surface water outflow into the Mediterranean ($\sim 1 \cdot 10^6$ m$^3$/s [39], yielding 10 MCI). The additions are obtained by multiplying the flow rates by the time-integrated tritium supply (Table I, column 7) reduced by a factor of 3 for the South Atlantic inflow to account for the lower titanium concentration there [12].
TABLE III. TOTAL NORTH ATLANTIC INVENTORY AND DELIVERY
Values are for end of 1972. For inventory see Section 2; deliveries from Table II;
0°–80°N: 10 MCi added for net addition by ocean currents, error is estimated
error of delivery calculation [2]; 0°–60°N: 60 MCI (± 30%) added for addition
by currents, error is estimated error using the total inventory as additional
constraint for the total delivery, see text

<table>
<thead>
<tr>
<th></th>
<th>Inventory (MCI)</th>
<th>Delivery (MCI)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0°–80°N</td>
<td>665 ± 8%</td>
<td>703 ± 20%</td>
</tr>
<tr>
<td>0°–60°N</td>
<td>–</td>
<td>512 ± 10%</td>
</tr>
</tbody>
</table>

The final comparison is summarized in Table III. One finds that inventory
and delivery agree well within their errors. We conclude from this agreement that
our delivery calculation is an adequate account of the contributing mechanisms
without serious omissions or systematic errors. In particular, it offers quantitative
proof of the dominant role of water vapour exchange, which previously had only
been postulated [30], deduced by difference [40] or even had been questioned [41].
The present scheme of calculation, therefore, can be used to convert tropospheric
tritium data into atmosphere-ocean tritium transfer, with good accuracy. For a
situation where the tropospheric tritium pattern is similar to the situation at
present evaluated, the overall delivery is defined even better than to ± 20%,
because the total inventory, with only ± 8% uncertainty, is a constraint for the
total delivery.

For application in the following section, Table III also contains the delivery
for the N. Atlantic sub-area 20° to 60°N. The error here may be taken to be
± 10%, because the sub-area still receives the larger part of the total delivery.
The figure in Table III further includes tritium addition over the southern boundary
(20°N) and by the East Greenland current from the north. The former is taken
to be the same as the net inflow over the equator (20 MCI, see above), i.e. net
tritium gain or loss of the 0° to 20°N latitude belt is neglected. The East Greenland
current has been postulated to add between 50 and 100 MCI [42]; the net addition,
however, is only about half this amount, because nearly the same amount of water
with about half the tritium concentration enters the Norwegian Sea from the
Atlantic (see also next section), so a net gain of 40 MCI is adopted.

2 This result of a minor contribution was for a high-altitude continental lake (Crater Lake)
and as such is correct, resulting from relatively low evaporation in the lake relative to oceanic
conditions (cf. Eq. (3)).
FIG. 4. North Atlantic two-box mixing model 20° to 60°N, after Ref. [3]. See also Table V and text. \( Q \) = atmospheric and river input, Equ. Atl. = net addition over the southern marine boundary, EGC = net addition over northern boundary which is presumed to be affected primarily by the East Greenland current. In the model, the input is stored in a surface-water layer (surface to \( d_1 \)), and an adjoining mid-depths layer (\( d_1 \) to \( d_2 \)), before it enters the deep ocean. The transfer into the deep ocean is via the Norwegian Sea (NS) and the Mediterranean (Med.) overflows and the Labrador Water (LW) formation.

The tritium delivery can be converted into the N. Atlantic strontium-90 delivery, using the previously deduced [3] tritium-to-strontium-90 input ratio (both inputs non-decay corrected) of 310 Ci/Ci. Neglecting differences in the areal input pattern between the two nuclides and applying the respective decay corrections (cf. footnote next section), we convert the 703 MCi tritium (Table III) into 2.8 MCi of strontium-90. This figure refers to 1972 and the borders of the N. Atlantic assumed here, 0° to 80°N, and its uncertainty is estimated to be ± 10 to 20%. The figure is larger by about 50% than delivery estimates obtained by assuming the published depositions from the HASL fall-out sampling network [43, 44] to be valid also for the N. Atlantic.

The tritium inventory of the N. Atlantic apparently is quite similar to that of the North Pacific [21, 40], despite the difference in area. The contribution deduced above of 25% to the total N. Atlantic input by net tritium outflow from the continents (which is insignificant for the Pacific) is not large enough to account for the difference. However, a further argument for larger N. Atlantic deposition is that the N. Pacific does not extend northward of 60°N, so that in these latitudes — in view of the essentially zonal tropospheric circulation — the Atlantic (together with the Arctic Ocean) has little competition as a sink for tropospheric tritium. This situation may also explain the fact that the maximum of the N. Atlantic tritium deposition (histogram in Fig. 1) is found at higher
TABLE IV. OCEANOGRAPHIC ESTIMATES OF DEEP WATER FORMATION RATES

After Ref. [39]. The Atlantic surface water component (= loss from the surface-water box of the model) enters the overflows via the Norwegian Sea or the Mediterranean; the entrained component is supposed to be derived from the subsurface box of the model.

<table>
<thead>
<tr>
<th></th>
<th>Surface water component ((10^6 \text{ m}^3/\text{s}))</th>
<th>Entrained component ((10^6 \text{ m}^3/\text{s}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Norwegian Sea overflow</td>
<td>6</td>
<td>4</td>
</tr>
<tr>
<td>(total of Island-Scotland</td>
<td>(total of Island-Scotland</td>
<td></td>
</tr>
<tr>
<td>and Denmark Strait</td>
<td>overflows)</td>
<td></td>
</tr>
<tr>
<td>Mediterranean overflow</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Labrador Water</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

latitudes than that of the strontium-90 deposition [43, 44], for which, despite the statement in the previous paragraph, land or sea does not make so much of a difference.

5. A NORTH ATLANTIC MIXING MODEL: TRITIUM TRANSFER INTO THE DEEP WATER

The time history of tritium (as well as of strontium-90) in N. Atlantic surface water has, in a previous paper [3], been treated by means of a two-box mixing model. Figure 4 shows an adaption for the present purpose. The two boxes simulate the mixing down to intermediate depth strata in response to atmospheric input, covering the area 20° to 60° N. The boxes interchange water, and lose water to the deep ocean at equal rates. Back flow from the deep water is neglected, because the model is supposed to be valid only for time scales up to a few decades. The previous treatment yielded the exchange times and the box volume ratio. In the following we use the delivery as determined above to fix also the box volumes.

The volumes are obtained from the previously deduced surface-box-depth to tritium-input ratio of 22 m/100 MCi if the input is non-decay corrected [3], or
TABLE V. MODEL PARAMETERS
See also Ref. [3]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model area ($20^\circ$--$60^\circ$N)</td>
<td>$26.9 \times 10^6$ (km$^2$)</td>
</tr>
<tr>
<td>Depth of surface-water box ($d_1$)</td>
<td>$187 \pm 10%$ (m)</td>
</tr>
<tr>
<td>Depth of subsurface box ($d_2$)</td>
<td>$534 \pm 10%$ (m)</td>
</tr>
<tr>
<td>Volume of surface-water box</td>
<td>$5.03 \times 10^6$ (km$^3$)</td>
</tr>
<tr>
<td>Volume of subsurface box</td>
<td>$14.36 \times 10^6$ (km$^3$)</td>
</tr>
<tr>
<td>Rate constant for deep water formation ($k_1$)</td>
<td>$1/30 \pm 50%$ (year$^{-1}$)</td>
</tr>
<tr>
<td>Rate constant for internal exchange ($k_2$)</td>
<td>$1/2.5 \pm 30%$ (year$^{-1}$)</td>
</tr>
<tr>
<td>Box volume ratio ($\gamma$)</td>
<td>$0.35 \pm 30%$</td>
</tr>
</tbody>
</table>

36.5 m/100 MCi for input in the present notation.$^3$ The resulting box depths are 187 m and 534 m. The total transfer into the deep water becomes 78 MCi of tritium and $10.6 \times 10^6$ m$^3$/s of water. This latter figure can be interpreted as follows: Processes of deep-water formation in the N. Atlantic are the Norwegian Sea and Mediterranean overflows, and the Labrador Water formation [39]. All of these remove both surface water and (by entrainment) intermediate-depth Atlantic water, which latter can be identified with the subsurface box of the model (see arrows in Fig. 4). The Atlantic surface water components have considerable delay in the Norwegian Sea or the Mediterranean before they reappear in the respective overflows, delays which are, however, irrelevant in the present context. Table IV lists the oceanographic estimates for all these flows. The combined rates of surface water and of entrained water flow are about equal, and they total $15 \times 10^6$ m$^3$/s. This figure compares with the model result of $10.6 \times 10^6$ m$^3$/s total rate and equal surface water and intermediate-depth water contributions. In view of the uncertainties — including those of the oceanographic estimates — the agreement is regarded as very satisfactory, and is taken as evidence for the consistency of the model of Fig. 4. The model now has all its parameters fixed, and Table V gives a listing. As for the error margins, the volume error ($\pm 10\%$) is that of the uncertainty of the subareal delivery (Table III). The other parameters have larger errors, e.g. $\pm 50\%$ for the combined loss rate into the deep water. As pointed out previously [3], the loss rate value may be improved by continued N. Atlantic fall-out observations. If this value is changed,

$^3$ Non-decay corrected input corresponds to a time-integrated supply with the exponential left out; the factor 22/36.5 is 1959 TU-years (Table I, column 5) divided by the sum of column 2 in Table I.
FIG. 5. Concentration (in g/m³) in North Atlantic surface water, calculated from the model (Fig. 4, Table V) to result from a single instantaneous addition over the model area (N. Atlantic, 20° to 60°N) averaging 1 kg/m².

the box volumes, in order to give the same 1972 tritium inventory, need a slight adjustment (−8/+3% for a ±30% loss rate change).

In total, the model, with the parameters of Table V, correctly reproduces the surface water concentrations of tritium and strontium-90, takes up the correct amount of tritium, and is compatible with known deep-water formation rates. At this point, there is still some uncertainty in its response over a few decades, because the fall-out has not yet covered such a time span. The model is not valid for time scales smaller than about one year (because no seasonal effects are taken into account). The model should be applicable to other tracers (or pollutants) as well. Such applications will require consideration of differences in the time and areal input patterns relative to tritium, which have a bearing also on the inputs from the north and south (broken arrows in Fig. 4). Uncertainties arising from these differences presumably will introduce larger errors than the model parameter uncertainties.

6. MODEL APPLICATIONS

As applications of the model, Figs 5 and 6 give two examples of the response of the N. Atlantic to specific atmospheric inputs. Figure 5 shows the case of a single instantaneous addition. The resulting surface water concentrations, normalized to unit input, are plotted as a function of time. The dilution becomes five-fold after about 15 years. Figure 6 gives equivalent mixing depths (i.e. the depths which, if multiplied by the surface water concentration, give the correct local oceanic inventory) for a substance the concentration of which increases exponentially in the surface water (or in the atmosphere), as a function of the
characteristic time of the exponential. This curve can be used to calculate the amount of excess atmospheric CO$_2$ [46] which has been taken up by the N. Atlantic. Representing the growth of fossil fuel by an exponential with 30 years' characteristic time, which rather is an upper limit [46], the corresponding equivalent depth is 900 m. Because the air-sea exchange of atmospheric excess CO$_2$ is fast [47], the amount taken up by the North Atlantic, 20° to 60°N, up to time $t_0$, practically is that of a water column of average depth 900 m, equilibrated with the atmospheric excess CO$_2$ concentration at time $t_0$. A curve showing the response according to the global box-diffusion model of Oeschger and co-workers [45] is included in Fig. 6. Comparison of the two curves demonstrates the faster down-mixing of the N. Atlantic compared with global-average conditions, and furthermore, that, quite apart from the difference in ordinate conditions, the response of the box-diffusion model is not as unique as is sometimes assumed.

7. SUMMARY

We have calculated the amount of tritium delivered to the N. Atlantic. Notable features of the delivery are that deposition by water vapour exchange is almost four times that by precipitation, and that 25% of the delivery is contributed by net tritium outflow from the adjoining continents (river run-off and tropospheric flows). As for the tropospheric tritium pattern, we find that the tritium concentration of marine water vapour and precipitation at ship's height are in near
isotopic equilibrium, and that continental concentrations are about four times those of the marine troposphere at the same latitude. We have further calculated the N. Atlantic tritium inventory (665 MCi ± 8%), and find agreement with the total delivery. We conclude from this that our delivery calculation scheme can be used to calculate, with good accuracy, future removal of tritium from the troposphere by uptake into the North Atlantic.

From the tritium delivery and the previously derived [3] tritium-to-strontium-90 input ratio, we deduce that about 2.8 MCi of strontium-90 have been taken up by the N. Atlantic — an amount 50% in excess of what is calculated from the deposition data of the HASL precipitation network.

In an extension of previous work [3], we present a N. Atlantic mixing model, which can be used to relate N. Atlantic surface water concentrations and inventories to input from the atmosphere, for constituents that have input time scales between a year and a few decades.

ACKNOWLEDGEMENTS

Sample collection for and measurement of the Heidelberg tritium data were supported by the Deutsche Forschungsgemeinschaft. G. Bader was responsible for most of the tritium measurements. We are grateful to numerous helpers, and to the masters and crew of the F.S. “Meteor”, for assistance in the sample collection work. The name of K. Fugmann of the “Meteor”, who on various cruises collected precipitation samples and ran the water-vapour sampling apparatus, may stand for many.

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DISCUSSION

K.J. VOGT: As I understand it, the river run-off delay factor used in the model has been taken from measurements on the River Rhine. Therefore you
may not have a universally applicable parameter here. Would you comment
on the possible variations and errors that may result from this?

W. ROETHER: The factor has been checked against tritium concentrations
measured in various rivers — not only European ones — and reasonable agree-
ment has been found, agreement certainly good enough for the present purpose.

L. MERLIVAT: I do not see clearly what you mean by the 'net
tropospheric inflow of vapour'. I would have thought rather that this
contribution had already been taken into account when you calculated the
molecular exchange above the ocean surface, since you assumed that the tritium
content of the vapour was in equilibrium with the precipitation, whose tritium
content should itself be fixed, at least in part, by this 'net tropospheric inflow
of vapour' of yours. I scarcely think that the two phenomena can be treated
independently in the tritium balance calculations. What would you say to this?

W. ROETHER: We have split off the contribution by excess tritium
from the continents as follows: firstly, we select tritium monitoring stations
unaffected by continental tritium, and we assume the corresponding depositions
to be valid for the entire latitude band. In reality, of course, tritium concentra-
tions will increase when you approach the continental boundaries, although no
detailed observations on this are available. Thus we take the effect of the
continents into account by calculating the amount of excess tritium that is
advected over the continental boundaries and we assume that this tritium is taken
up quantitatively by the ocean.
TRITIUM IN THE TERRESTRIAL ENVIRONMENT

Session 5, Part 2, and Sessions 6 and 7
TRANSFER AND UPTAKE MECHANISM OF TRITIUM IN SOIL

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Abstract

TRANSFER AND UPTAKE MECHANISM OF TRITIUM IN SOIL.

Movement of tritiated water (HTO) in soil is determined by a number of factors such as evapotranspiration, soil-water diffusion, gravity flow of soil water, rainfall and mechanical characteristics of the soil. Elimination of HTO from a soil column usually is a semilogarithmic function of time. Residence half-times vary greatly, from hours to years, depending on the factors mentioned above and the depth in soil of the maximal HTO concentration. Short half-time values are observed when the HTO remains near the soil surface. In deeper strata of dense soils the HTO is effectively retained for years.

1. INTRODUCTION

Tritiated water, HTO, is being used extensively as a tracer to indicate the flow of water in porous media. Although many other radioisotopes (e.g. $^{82}$Br, $^{131}$I, $^{99}$Te$^{m}$) have been used as tracers for water in hydrology, tritiated water offers several advantages. The slightly larger atomic radius of tritium compared with protium causes only a small isotope effect, and chemically HTO behaves in soils virtually like ordinary water. Tritium is inexpensive, has a convenient half-life (12.3 years) and is easily measurable by liquid scintillation, thus permitting large quantities of groundwater to be labelled inexpensively.

Tritium is naturally present in small concentrations ($^3$H : H = $10^{-18}$) in the atmosphere, hydrosphere, biosphere and soils. Furthermore, in the thermonuclear bomb tests of the 1960s large quantities of man-made tritium were released to the environment. All this tritium will eventually be converted into tritiated water and mixed with water circulating in the hydrological cycle. The bomb test tritium has been successfully used as a tool to study groundwater movements. In future, thermonuclear devices may be used for peaceful purposes, e.g. for excavation and soil movement, and these could produce huge additional quantities
of tritium. Some tritium will also be produced by light-water power reactors, by breeders and, eventually, by fusion reactors.

Information on the movement of tritiated water in soils will be needed in all these connections.

2. THE ISOTOPE EFFECT

Several authors have reported an isotope effect in movement of tritiated water in soils. Stewart [1] observed that during the diffusion of tritiated water through a column of oven-dried clay the water near the wetting front becomes diluted with respect to tritium. Kaufman and Orlob [2] and Biggar and Nielsen [3] observed a slight retardation in the flow velocity of HTO compared with the flow of chloride ions. Corey and Horton [4], on the other hand, found no measurable difference in the relative flow rates of deuterium, tritium and $^{18}$O-labelled water molecules through acid kaolinitic soils. Stewart’s studies [5] also show that fractionation of tritium between tenaciously adsorbed water and bulk pore water is not measurable for the following clay minerals: Davison clay, kaolinite, montmorillonite, vermiculite, illite and halloysite. For most practical hydrologic investigations it is unlikely, therefore, that the isotope effect of HTO in soils would be great enough to require a special correction or even to be observable. Other factors affecting the flow of water in stratified soils are more significant. In some theoretical studies on the mechanism of interaction of water and soil, e.g. the exchange of TO in bulk pore water with hydroxyl groups of mineral lattices [1, 6], it may be possible to make use of the isotope effect of tritium.

TO probably exchanges slowly with hydroxyls in clay mycelles, perhaps with those attached to Al and Mg atoms. The TO groups thus bound may become available to plant roots only with difficulty, and the residence time of tritium in such soils may be relatively long. Soils rich in humus may also contain exchangeable hydroxyl groups. Compared with other variables — rainfall, gravity flow, diffusion, evapotranspiration — the exchange reactions have little effect, however.

3. WATER MOVEMENT IN SOILS UNDER VARIOUS CLIMATES

In the early 1970s Sasscer at the University of Puerto Rico and Jordan and Kline at the Argonne National Laboratory, Illinois, United States of America [7], as well as Koranda and Martin [8], carried out pioneering studies on water movement in soil under various climatic conditions. At the same time they presented dynamic models which predicted the observed data fairly well.
Sasscer and co-workers [7] applied their model to the soils of a tropical rain forest, an Illinois grassland and a Nevada desert site. The coefficients of their model were assumed to be functions of evapotranspiration, soil-water diffusion, gravity flow of soil water and the rainfall history of the previous 48 hours. After rainfall, soil water was found to move downward in a pulse due to gravity flow, to spread out due to diffusion, and to leave the system by evapotranspiration.

The results of Sasscer and co-workers for the Puerto Rico rain forest and their predictions are presented in Fig.1. Basic agreement on the location of the bulk of the tritium was obtained: e.g. after 77 days the peak concentration was at about 30 cm depth and had a value of about $5 \times 10^3$ dis/min per ml in both cases. In Fig.2 their values for the downward velocity of the peak concentration are illustrated as a function of time after application. In Puerto Rico, the peak velocity decreased linearly for one week but slowed down thereafter. The 'hump' of the Illinois grassland curve occurred during the spring when the soil was saturated. In the Nevada desert diffusion moved HTO deeper into the soil, whereas transpiration removed it from the upper layers. The total tritium per cm$^2$ of surface in the Illinois grassland is presented as a function of time in Fig.3. During the first two months a half-residence-time of 10 days prevailed, but then the loss became retarded and a half-time of 225 days soon became dominant. Two half-times were also observable in the Nevada desert: 30 days was evident for almost a year and then a half-time of 710 days became dominant about two years after the application. The initial value prevailed while the HTO was in
the region of the plant roots and plants controlled the transpiration, and the larger value prevailed when the HTO remained only in deeper layers, below the plant roots.

Koranda and Martin [8] studied a tritiated soil profile at the Sedan nuclear test crater in the Nevada desert over a period of five years. They observed, between the 4th and 9th year after the test, a residence half-time of $1.1 \pm 0.24$ years, i.e. a somewhat shorter value than the 710 days of Sasscer and co-workers [7] mentioned above. This value (1.1 years) was obtained by sampling the soil to a depth of 1.8 to 2.4 m, integrating the HTO in the soil column and expressing the data as activity per unit area (Fig.4). The soil at the Sedan crater is sandy and contains a small amount of clay; rainfall is 75 mm yearly.

Useful information on the behaviour of a pulse of water in soil is obtained from the experiments of Zimmerman and co-workers [9] carried out with HDO. Labelled water applied to the soil surface was eluted into the soil profile when
**FIG. 3.** Total tritium per square centimeter of surface in the Illinois grassland as a function of time after application [7].

**FIG. 4.** Environmental half-time of tritium in Sedan Crater ejecta [8].
TABLE I. SOME HALF-RESIDENCE TIMES OBSERVED FOR VARIOUS SOILS IN WIDELY DIFFERENT CLIMATES IN THE IAEA CO-ORDINATED PROGRAMME

<table>
<thead>
<tr>
<th>Location and reference</th>
<th>Region and biome</th>
<th>Half-residence time (d)</th>
<th>Notes on soil</th>
<th>Annual rainfall (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Finland [10]</td>
<td>Tundra</td>
<td>6-90-780</td>
<td>Fine sand-silt</td>
<td>450</td>
</tr>
<tr>
<td></td>
<td>Boreal</td>
<td>104-1280</td>
<td>Fine sand-silt-clay</td>
<td>650</td>
</tr>
<tr>
<td></td>
<td>Boreal</td>
<td>5-63-204</td>
<td>Fine sand-silt-clay</td>
<td>650</td>
</tr>
<tr>
<td>France [11]</td>
<td>Mediterranean</td>
<td>1 h 45 min to 2 h 15 min</td>
<td>Clay, Calcerous</td>
<td>700–800</td>
</tr>
<tr>
<td>Mexico [12]</td>
<td>Temperate</td>
<td>14-85</td>
<td>Sandy loam, crops</td>
<td>530</td>
</tr>
<tr>
<td></td>
<td>Agricultural</td>
<td>10-180</td>
<td>Clayey vertisol, crops (+irigation)</td>
<td>800 (+700)</td>
</tr>
</tbody>
</table>

successive additions of water were applied. The HDO moved sheet-like through the profile, slightly blurred by diffusion. The more water present in the soil, the slower was the downward movement of the labelled layer. Fine grain soil showed an average draining rate of less than one metre annually.

4. STUDIES BY THE IAEA CO-ORDINATED PROGRAMME DURING 1973–76

The terrestrial studies in the IAEA tritium programme were diverse and embraced widely different environmental conditions, from arctic to tropical. Some were carried out in well-irrigated agricultural plots, others in native fields or forests obtaining natural rainfall only. Some of the residence half-times measured are presented in Table I.

In northernmost Finland [10], where the duration of the soil frost is about 200 days, the half-residence times were initially six, then 90 days during the first year and thereafter 780 days. In subarctic southern Finland values of 5, 63 and 104 days were measured for the first year, and about 200 and 900–1300 days for the subsequent years in two fine-sand-silt-clay soils studied in two separate labelling experiments. In southern Finland, duration of soil frost is usually 120 days. Long half-residence times, up to 435 days, were also observed in the hot, dry desert soils of Nevada.

In the arctic region (Finland) the maximum was found after one year at a depth of 20–30 cm, after 2 years at about 60–80 cm and thereafter deeper than
1 m. In southern Finland the horizontal movement of groundwater contributed to the elimination of HTO during the long experimental period.

The extremely short half-times reported from France [11] were measured during a 24-hour period at a shallow depth (0–30 cm) after 1 mm water was sprayed on a porous agricultural soil of medium permeability, not covered with grass, on a warm day (max. temperature 28°C). The rapid loss is mainly due to evapotranspiration.

The Mexican [12] values shown in Table I were obtained over a longer period in deeper strata in agricultural soils. They are much greater than the French values observed during the first day, but shorter than the Finnish values owing to differences in soil and climate.

In the Philippines [13] HTO retention was studied in agricultural soils in the ten top cms over a 12-week period. Daytime temperature was 28.5°C. Half-times varied from 12 to 14 days in this top soil layer.

Many reported half-times of tritium in soil are based on measurements of the specific activity of the soil water only. These values are not comparable with the integrated values if the water content of soil is increased during the observation period through irrigation and/or rainfall which causes dilution. True residence times are obtained only if the total amount of tritium in the soil profile per unit area is calculated.

The effect of frost in the ground upon a tritiated water layer in groundwater was studied in Helsinki during the winter and spring of 1978 [14]. When the ground surface is frozen in mid-winter, tritiated water from the deeper layers is evaporated or transported by capillary action up to the lower surface of the frozen crust which acts as a condenser. When the soil surface melts in the spring, higher tritium values may be observed near the surface than during the previous autumn.

The behaviour of tritiated water in soil can be summarized as follows:

1. Tritiated water spread upon the soil surface moves into deeper strata at a rate proportional to the amount of water (irrigation, rainfall) applied after the HTO application.

2. The rate of movement of the labelled water layer also depends on the mechanical characteristics of the soil. In practice, fissures or holes in soils cause irregular 'injections' into deeper strata.

3. The deeper the layer of tritiated water has penetrated the longer its residence half-time. In clay-containing soils of subarctic regions a very rough 'rule of thumb' is valid: the residence half-time is of the same order of magnitude as the time passed from the surface application.

4. Short half-times observed when the water layer is still near the surface are caused by evapotranspiration, which is affected also by the vegetation. From deeper strata (below plant root level) removal takes place by diffusion and deep
drainage and, during frost, by capillary transfer and evaporation upwards to the frozen crust.

5. In deeper strata of dense soils tritiated water may be retained for years. A pulse of HTO rises from this depot to the soil surface in the summer, mainly determining the tritium content of the vegetation.

REFERENCES

J. DELMAS: I should first like to make two comments whose purpose is to indicate the work that still remains to be done in this field.

Firstly, your paper takes only total tritium into account, particularly free-water tritium, and ignores that bound to organic matter thanks to the activity of soil and plant micro-organisms. The cycle of this organic-matter tritium is not well known, and this is precisely one of the things about which we should like to discover more.

Secondly, with regard to soil-water bound $^3$H and its ascending and descending movements in the soil under various climatic effects, it would be a good idea to establish a mathematical model taking account of contributions and losses due to precipitation and evapotranspiration. One of the uses of such a model would be to yield an estimate of long-term contamination of groundwater. This is an important factor in evaluating the radiological component of a given site's environment.
TRANSFER OF TRITIATED WATER VAPOUR TO AND FROM LAND SURFACES

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Abstract

TRANSFER OF TRITIATED WATER VAPOUR TO AND FROM LAND SURFACES.

Deposition of tritiated water vapour may result in contamination of food supplies and reduction of air concentrations, but the process has received little attention. Field and laboratory experiments were undertaken to investigate the mechanisms involved. The exchange between air and ground involves turbulent transfer in the atmosphere near the surface and diffusion within the soil. The exchange velocity for moist soil was limited by atmospheric mixing to about 1 cm s$^{-1}$, and was only a little smaller at dry soil and grass surfaces. For exposure times exceeding a few minutes re-evaporation becomes significant, reducing the net rate of uptake, but the total amount deposited continues to increase as the tritiated water diffuses deeper into the soil. Diffusion in soil was investigated and a simple equation proposed to predict the effective diffusion coefficient. Tritiated water deposited during a brief exposure in the field evaporated from the surface during several weeks, and its behaviour can be described by the diffusion equation with a suitable boundary condition. Rain washes activity into the soil and impedes further evaporation. Most of the vapour will interact with the surface within two or three days of release, and transport over continental distances will be retarded as a result. The effect of surface exchange on the distribution of doses following a release of tritiated water vapour will be complicated by changes in the weather over a period of several weeks, and will be difficult to foresee.

1. INTRODUCTION

The prediction of radiological consequences of accidental or routine releases of tritium is an important part of the safety assessment and emergency planning for nuclear facilities. A large fraction is released as tritiated water (HTO) in vapour or liquid waste streams, and the ready access of this material to food chains and water supplies enhances the significance of this material. Safety assessments frequently neglect to account for the interaction between the vapour and the ground surface in predicting doses at a distance, because of inadequacies in the understanding of the processes involved. Here we describe measurements of the rate of uptake of HTO by land surfaces and the fate of the absorbed HTO, and indicate their significance in the environmental behaviour of the vapour.
2. EXCHANGE OF HTO AT THE ATMOSPHERE-SOIL SURFACE

HTO is frequently considered as a tracer for water, and often follows the flow of water faithfully. However HTO transport can occur by diffusion where there is no mass flow of water. In the vapour phase the diffusive flow of HTO may even be in the opposite direction to that of water, and the evaporation of HTO from a soil surface is not controlled by the availability of heat as is the evaporation of water. Here we consider exchange of HTO in the absence of a net flow of water.

Exchange of HTO between air and soil requires transport through the air to the surface and transport within the soil. Uptake is expected to be reversible and the deposition flux can be written

\[ F = (x(z) - x_s)v(z) \]  \hspace{1cm} (1)

where \( F \) is the downward flux of HTO, \( x_s \) and \( x(z) \) the concentration of HTO in air that would be in equilibrium with the absorbed HTO at the soil surface, and at height \( z \) respectively, \( v(z) \) a parameter with dimensions of velocity, which will be called the exchange velocity.

The exchange velocity is determined by the degree of turbulent mixing above the surface as well as the nature of the surface itself. It depends on wind speed, surface roughness and the thermal structure of the air, and in suitable circumstances it can be predicted from micrometeorological measurements.

Within the soil HTO vapour exchanges with the soil water, and locally there exists an equilibrium between the vapour density in the pore space and the concentration in the liquid phase. Let

\[ \gamma = \frac{\text{concentration of HTO vapour per unit volume of air (} x_s \text{)}}{\text{total concentration of HTO per unit volume of soil water (} x_l \text{)}} \]  \hspace{1cm} (2)

Ignoring a small isotope effect on vapour pressure, \( \gamma \) may be written

\[ \gamma = \frac{\rho_v}{\rho_w} \frac{H}{100} \]  \hspace{1cm} (3)

where \( \rho_v \) is the saturation vapour density of water at the temperature of the soil, \( H \) the relative humidity in the soil, practically 100 except in very dry soil, \( \rho_w \) the density of water.
Diffusion of HTO occurs in the liquid and vapour phases. Penman [1] measured the diffusion of vapour in soil and found that the ratio of effective diffusion coefficient $D_\text{a}$ to that in free air, $D_{0\text{a}}$, was 0.66 $S_\text{a}$ where $S_\text{a}$ is the fraction of the total volume occupied by air. The coefficient 0.66 reflects the tortuosity of the vapour paths through the soil pores and $S_\text{a}$ may be regarded as the fraction of the total cross-sectional area available for vapour diffusion.

Treating the liquid phase diffusion similarly, the total diffusion coefficient for HTO in soil is predicted as

$$D = 0.66 \frac{S_\text{a}D_{0\text{a}}}{S_1} + S_1 D_1$$

(4)

where $S_1$ and $D_1$ are the fractional soil volume occupied by water and the appropriate diffusion coefficient for HTO. Equation (4) was tested in laboratory experiments in which HTO was allowed to diffuse into soil columns under known conditions. The results showed agreement with Eq.(4) to within 10%. This formula also agreed well with the observations of diffusion in saturated sediment of White and co-workers [2].

In this work we deal with one-dimensional diffusion problems with the boundary condition specified by Eq.(1). Solutions are expressed in terms of

$$y_1 = 2(Dt)^{1/2}, \quad y_2 = (S_1 D)/(\gamma \nu) \quad \text{and} \quad t_1 = y_2^2/D$$

The total deposited per unit area of soil surface after time $t$ is

$$M = \frac{x S_1 y_2}{\gamma} \left[ \exp \left( \frac{1}{t_1} \right) \text{erfc} \left( \frac{1}{t_1} \right)^{1/2} - 1 + \frac{2}{\pi^{1/2}} \left( \frac{t}{t_1} \right)^{1/2} \right]$$

(5)

At large $t/t_1$, $\exp \left( \frac{1}{t_1} \right) \text{erfc} \left( \frac{1}{t_1} \right)^{1/2} \rightarrow 0$ so the asymptotic behaviour of $M$ is

$$M \rightarrow \frac{2x S_1}{\gamma} \left( \frac{Dt}{\pi} \right)^{1/2}$$

(6)

So a long exposure of sufficiently deep soil results in continued uptake, the total deposited increasing with the square root of exposure time. The retention and the concentration profile during evaporation after a brief exposure of duration, $\tau$, are also of interest. These are given by
\[ C_1 = \frac{\tau X S_1}{t_1 \gamma} \left( \frac{t_1}{\pi t} \right)^{1/2} \exp \left( -\frac{Y^2}{Y_1^2} \right) - \exp \left( \frac{Y}{y_2} + \frac{1}{t_1} \right) \text{erfc} \left[ \frac{Y}{Y_1} + \left( \frac{1}{t_1} \right)^{1/2} \right] \]  
(7)

\[ M_1 = \tau X \exp \left( \frac{-t}{t_1} \right) \text{erfc} \left( \frac{1}{t_1} \right)^{1/2} \]  
(8)

3. EXPERIMENTAL

3.1. The rate of sorption of HTO by soil surfaces

3.1.1. Method

Sieved soil, packed in trays of 1 cm depth, was exposed to HTO vapour in a wind tunnel. So that the exchange of HTO and of water vapour could be compared, the rate of evaporation was determined simultaneously by measuring the weight loss from small trays of soil, carefully levelled with the soil surface. The soil surface temperature, the air temperature and relative humidity were also measured. HTO vapour was introduced into the tunnel by passing air through a bubbler containing tritiated water. To determine the concentration of HTO 5 cm above the down-wind end of the soil surface, air was drawn at 5 ltr·min⁻¹ through bubblers containing distilled water. Uptake in the soil was determined by distilling water from the soil. Aliquots from the resulting samples were counted for tritium in a liquid scintillation counter.

The wind speed profile above the soil surface was measured with a Pitot tube.

Soil was exposed to HTO vapour for periods of 20 minutes to 1 hour in the wind tunnel. Longer exposures were performed in a perspex box, a cube of 50 cm side, in which the air was mixed vigorously by a fan below the roof of the box and directed upwards. Soil, packed in Petri dishes, was placed on the floor of the box. HTO vapour was supplied by passing air at 0.5 ltr·min⁻¹ through a bubbler and into the box and an additional flow of 0.5 ltr·min⁻¹ of dry air was supplied to prevent condensation on the sides of the box. The box atmosphere was sampled at 1 ltr·min⁻¹ through a bubbler containing distilled water, so that the concentration of HTO could be obtained.

3.1.2. Results

The results were expressed in terms of the exchange velocities

\[ v_{HTO} = \frac{\text{HTO deposit per unit area per unit time}}{\text{HTO concentration 5 cm above the soil}} \]
\[ \nu_{H_2O} = \frac{\text{Evaporation per unit area per unit time}}{\text{(saturation vapour density at soil surface temperature - vapour density in air at 5 cm)}} \]

Table I shows values of both parameters deduced from wind tunnel runs with air-dry soil (experiments 1 to 3) and moist soil. The values may be compared with an exchange velocity for a surface freely supplied with moisture as indicated by Chamberlain [3]. The resulting exchange velocity, \( \nu \), is an upper limit attainable when exchange is controlled by aerodynamic transport above the surface and exchange at the surface itself is unimpeded.

For moist surfaces in experiments 4 and 5 (Table I) and short exposures \( \nu_{HTO} \) and \( \nu_{H_2O} \) are similar and approach the upper limit. For dry soil the evaporation rate is some 40 times smaller but \( \nu_{HTO} \) is reduced only two or three times. Considerable variability results from inhomogeneity in the soil and the difficulty of measuring the surface temperature, but a decrease in \( \nu_{HTO} \) at longer exposure times is apparent.

Deposition was measured for a wider range of exposure times in the box experiments (Table II). The apparent deposition velocity, \( \nu_{HTO} \), compares reasonably well with the predicted value, \( M/(\text{xt}) \), calculated from Eq.(5).

3.2. Evaporation of HTO from contaminated soil in the field

3.2.1. Method

Prepared surfaces of bare soil and grass, cut to 5 cm, in an experimental field were exposed to HTO vapour for periods of 30 minutes, and profiles of HTO

**TABLE I. EXCHANGE VELOCITIES FOR HTO AND WATER VAPOUR AT A SOIL SURFACE**

\( u \) (5 cm) = 4.0 m\( \cdot \)s\(^{-1} \), \( u_* \) = 31 cm\( \cdot \)s\(^{-1} \), \( z_0 \) = 0.02 cm; hence \( v \) = 1.9 cm\( \cdot \)s\(^{-1} \)

<table>
<thead>
<tr>
<th>Expt. No.</th>
<th>Soil moisture content (per cent wet weight)</th>
<th>( \nu_{H_2O} ) (cm( \cdot )s(^{-1} ))</th>
<th>( \nu_{HTO} ) for exposure times of</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>20 min</td>
<td>40 min</td>
</tr>
<tr>
<td>1</td>
<td>4.3</td>
<td>0.05</td>
<td>0.66</td>
</tr>
<tr>
<td>2</td>
<td>4.3</td>
<td>0.05</td>
<td>0.51</td>
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<td>1.15</td>
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<td>21</td>
<td>1.1 \pm 0.5</td>
<td>1.42</td>
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<tr>
<td>5</td>
<td>14.5</td>
<td>1.9 \pm 0.7</td>
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</tbody>
</table>
TABLE II. UPTAKE OF HTO BY SOIL IN THE BOX EXPERIMENT

<table>
<thead>
<tr>
<th></th>
<th>Exposure time, t (seconds)</th>
<th>(v_{\text{HTO}}) (cm \cdot s(^{-1}))</th>
<th>(v_{\text{HTO predicted}}^a) (Eq.(5)) (cm \cdot s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Dry soil</strong>(^b^)</td>
<td>300</td>
<td>0.88</td>
<td>0.66</td>
</tr>
<tr>
<td></td>
<td>1800</td>
<td>0.31</td>
<td>0.43</td>
</tr>
<tr>
<td></td>
<td>3600</td>
<td>0.26</td>
<td>0.35</td>
</tr>
<tr>
<td></td>
<td>20600</td>
<td>0.09</td>
<td>0.11</td>
</tr>
<tr>
<td><strong>Wet soil</strong>(^b^)</td>
<td>300</td>
<td>0.91</td>
<td>0.85</td>
</tr>
<tr>
<td></td>
<td>1800</td>
<td>0.38</td>
<td>0.66</td>
</tr>
<tr>
<td></td>
<td>3600</td>
<td>0.38</td>
<td>0.58</td>
</tr>
</tbody>
</table>

\(^a\) Conditions: \(v = 1.0\) cm \cdot s\(^{-1}\)

\(^b\) Dry soil: \(S_1 = 0.08, S_a = 0.4,\) temperature 20°\(C\)

Wet soil: \(S_1 = 0.35, S_a = 0.13.\)

---

**FIG.1.** Results for bare soil. The fraction of the HTO, deposited during a thirty-minute exposure, that remained after various times. The curve was calculated using Eq.(8) with \(t_i = 2.64 \times 10^6\).
FIG. 2. The depth distribution of HTO in bare soil sheltered from rain (a) 1 day, (b) 2 days, (c) 8 days and (d) 28 days after a 30-minute exposure. The smooth curves show predicted distributions based on Eq. (7) with $D = 3 \times 10^{-6}$ cm$^2$·s$^{-1}$ and $y_2 = 0.28$ cm.

in soil measured subsequently for several weeks. Some plots were shielded from rain by perspex covers which were raised some 20 cm above the surface so that natural ventilation was not prevented.

The HTO exposures were achieved using a perspex box, a cube of 50 cm side with no base, in which the air was mixed with a fan. The box was set on the ground so that the soil surface formed the bottom of the box. Air was passed through a bubbler containing HTO in water at 1 ltr·min$^{-1}$ and into the box for 30 minutes.
FIG. 3. The depth distribution of HTO in soil exposed to rain (broken line) and sheltered from rain (solid line).
(a) 8 days after exposure to HTO vapour, when the accumulated rainfall was 1.8 cm.
(b) 13 days after exposure, when the accumulated rainfall was 6.5 cm.

Soil cores were extracted with a corer 4.3 cm in diameter, containing a closely fitting tube cut into 1 cm segments. This made it easy to separate the soil core into 1 cm layers, so that profiles of HTO concentration in soil could be obtained.

3.2.2. Results

The most coherent data were obtained from the bare soil plot that was sheltered from rain. The fraction of the deposited activity remaining at various times is plotted in Fig. 1.

It was found that a value of $2.64 \times 10^4$ s for $t_1$ in Eq. (8) gave a good fit to Fig. 1. With this value the best fit to the soil profiles, Fig. 2, required $D = 3 \times 10^{-6}$ cm$^2$·s$^{-1}$, whereas Eq. (5) predicts a diffusion coefficient of $1.03 \times 10^{-5}$ cm$^2$·s$^{-1}$. The value of the exchange velocity, $v$, implied by the
corresponding value of \( y_2 \) was 0.55 cm·s\(^{-1}\). This seems credible in view of the reduced ventilation under the rain cover. The discrepancy in the magnitude of D is unexplained but in the main the results are described by the theory. The results for grass plots are much more scattered, probably because of the irregular distribution of plants and roots, and are not amenable to mathematical analysis. However, the uptake by grass in this experiment was a few times less than uptake by bare soil, but the activity was retained rather longer.

In plots on which rain was allowed to fall the HTO was displaced downwards and diffused over a greater depth range (Fig.3). When sufficient rain had fallen, as in Fig.3(b), the HTO was completely displaced from the soil surface. Evaporation of HTO would then cease, at least for a while, and the tritium would join the groundwater drainage or be retained in the soil for an extended period. It was found that the depth of the peak of HTO concentration in plots exposed to rain was a little less than the depth given by dividing the rainfall by the soil porosity.

4. DISCUSSION

Experiments reported above show that HTO vapour is sorbed from the atmosphere by wet and dry soil and grass, and an appreciable fraction is retained for several days and even weeks.

It is not immediately apparent what the radiological consequences of these exchange processes are. Comparison with SO\(_2\) which has a similar deposition rate [4, 5] suggests that 50% or more of the HTO must be absorbed in a travel distance of 1000 km, and the exchange processes must significantly modify the air dose at distances exceeding 100 km. Contamination of crops and water supplies by the deposited HTO may be important close to the source.

It can be shown that the time integral of inhaled dose at a point downwind of a short release will be unchanged by deposition if the material all evaporates again provided the wind speed and direction are constant. However, large changes in the wind vector are expected during the several days that evaporation lasts, and the net effect may be to distribute the dose over a wider area. Some fraction of the evaporating material may be carried back to the source area, increasing the dose there.

Direct absorption of HTO vapour by water surfaces is also to be expected. HTO behaves like a very soluble gas and deposition velocities of the order of 1 cm·s\(^{-1}\) must be expected [6]. The rapid dilution in lakes, seas or rivers would impede re-evaporation. Uptake would therefore result in removal from the atmosphere and possibly significant contamination of water supplies.

Further work is necessary to quantify these processes and a method of calculating their influence on dose distribution following a release is required.
REFERENCES


DISCUSSION

P.J.C. DINNER: Is there any physical justification for assuming that the 'tortuosity coefficient' of 0.66 used in your Eq.(4) should be the same for both interstitial air and water diffusion paths?

J.A. GARLAND: Equality of the tortuosity coefficient in air and water paths implies a degree of similarity in the geometry of the two phases, and was indeed quite a gross assumption. However, experiments on diffusion of HTO in soil in the laboratory provided results which in fact agreed quite well with the equation for a wide range of soil water contents.

A.B. GUREGHIAN: You seem to have based your analysis solely on the diffusive and dispersive terms arising in the mass transport equation, while neglecting the convective term. However, during the initial stage of infiltration of water due, say, to rainfall in a quasi-unsaturated soil, the velocity factor would play a dominant role in the migration of tritium. Could you comment please?

J.A. GARLAND: The relative importance of diffusion and convection varies with time after exposure. At the start there are large concentration gradients, which generate correspondingly large diffusive fluxes, but later the gradients are small and convection will eventually become dominant. It seems probable that with typical soil conditions (mean evaporation or precipitation rate ~ 2 mm daily) diffusion will be dominant for some tens of days, but heavy precipitation could produce important convective flows at any time. The simple diffusion model should be adequate to describe tritium movement over such a period, during which most of the absorbed HTO will evaporate.

W. ROETHER: A few years ago our group at Heidelberg made extensive studies of the movement of layers of tritiated water in the soil, at depths larger than those you have been talking about (50 to 100 cm). We found essentially
advective movement, caused by gain or loss of soil moisture above the layer, and diffusion was superimposed on this.

J.A. HETHERINGTON: In relation to the last question, I should like to point out that convective mechanisms will always predominate in the longer term following a single release episode as the concentration gradients needed to sustain the diffusive process disappear.

Mr. Garland, although you use a composite term to cover diffusion in both the vapour and water phases of the soil, it will be interesting to know the relative contribution of diffusion in each phase in a typical soil (e.g. porosity about 0.2 and soil water content 40%).

J.A. GARLAND: The relative importance of water-phase and vapour-phase diffusion depends on the soil water content and temperature. In very wet soil there is little air space but ample water, and diffusion occurs almost exclusively in the water phase. Low temperatures also tend to suppress vapour phase diffusion. In rather dry soils and at temperatures exceeding 10°C–15°C the vapour phase diffusion is dominant.
ENVIRONMENTAL TRANSPORT AND CYCLING OF TRITIUM IN THE VICINITY OF ATMOSPHERIC RELEASES

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Abstract

ENVIRONMENTAL TRANSPORT AND CYCLING OF TRITIUM IN THE VICINITY OF ATMOSPHERIC RELEASES.

Routine releases of tritium to the atmosphere from the Savannah River Plant (SRP) have averaged $5.12 \times 10^8$ curies annually during the period 1974–77. Patterns of tritium dispersion in the environment surrounding the SRP indicate a power law relationship between air concentration and distance from the source. The relationship between the concentration of tritiated water in vegetation and distance is more complex. A model of the tritium dispersion and cycling in the environment explains the dispersion patterns in terms of the ratio of tritiated hydrogen to tritiated water, climate and vegetation characteristics. The annual variation in the environmental patterns around the SRP are almost completely determined by the fraction of tritiated hydrogen in the release.

INTRODUCTION

The Savannah River Plant (SRP) is a major producer of nuclear materials for the U.S. Government. Inevitably, some of these materials and their byproducts find their way into the environment around the Plant [1]. The SRP Health Physics Department and the Environmental Sciences Section of the Savannah River Laboratory (SRL) measure the distribution of these materials in the environment and assess the dose-to-man from the operation of the plant. We have collected data to demonstrate the patterns of distribution of tritium in the terrestrial environment. Experiments will be briefly described which have led to a better understanding of tritium movement in the environment. Simulations with a model of tritium movement were compared to the measured tritium distribution.

* This paper was prepared in connection with work under Contract No. AT(07-2)-1 with the U.S. Department of Energy.
THE SAVANNAH RIVER PLANT SITE

The production facilities at the SRL include three operating heavy-water reactors, a heavy-water separations facility, a fuel fabrication facility, a product separations facility and waste disposal facilities. The production areas are located in a 77 831 ha reservation in South Carolina, east of the Savannah River near Augusta, Georgia, in the southeastern United States of America.

The reservation is on the western edge of the coastal plain and has soils derived from coastal plain and piedmont parent materials, as well as extensive areas of alluvial soils near the Savannah River and sandhill soil away from the river. Much of the upland area that had been farmed before the establishment of SRP is now managed pine plantations. The river swamp and areas next to the streams are in mixed broadleaf forests. The climate of SRP is relatively temperate with mild winters and long, warm summers. The temperature averages 18°C in Augusta and the average relative humidity is 70%. Rainfall is evenly distributed throughout the year and averages 120 cm/a [2]. Dry periods of 8–10 days are encountered almost every year.

Tritium source characteristics

Tritium releases to the environment are about equally divided between releases from the heavy-water reactors and the tritium separations areas. Ninety per cent of the releases to the atmosphere are from 63-m-high stacks. The average annual release to the atmosphere during the existence of the Plant is $7.82 \times 10^5$ Ci/a. Releases from the separations area average 40% tritium hydrogen gas (HT) and 60% tritiated water (HTO). The releases from the reactor areas are primarily HTO under normal operating conditions. Accidental releases of $4.79 \times 10^5$ Ci on 4 May 1974, and $1.82 \times 10^5$ Ci on 31 December 1975, were greater than 99% HT.

Patterns of tritium distribution in the environment

The Health Physics Department routinely analyses precipitation, and moisture in air, vegetation and soils for HTO content at a number of collection locations at the plant boundaries and at radial distances of 40 km and 161 km [3]. In addition, special studies are made to determine HTO in vegetation along transects in four directions from the Plant centre. Results of the routine measurements of air water and vegetation water are summarized in Table I for 1974–77.

The air moisture samples are collected continuously on silica gel and are analysed once a week. The vegetation samples are grab samples made during collection runs. The water is removed from the vegetation by vacuum distillation and counted by liquid scintillation [4].
TABLE I. TRITIATED WATER CONCENTRATION IN AIR AND VEGETATION WATER (pCi/ml)

<table>
<thead>
<tr>
<th>Year</th>
<th>1974</th>
<th>1975</th>
<th>1976</th>
<th>1977</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Plant Perimeter</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(10 km)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td>11.8</td>
<td>8.8</td>
<td>5.0</td>
<td>6.5</td>
</tr>
<tr>
<td>Vegetation</td>
<td>15.2</td>
<td>14.0</td>
<td>7.1</td>
<td>2.9</td>
</tr>
<tr>
<td>(40 km)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td>4.9</td>
<td>4.5</td>
<td>1.8</td>
<td>1.8</td>
</tr>
<tr>
<td>Vegetation</td>
<td>12.3</td>
<td>6.0</td>
<td>2.6</td>
<td>0.6</td>
</tr>
<tr>
<td>(160 km)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Air</td>
<td></td>
<td></td>
<td>0.5</td>
<td>0.7</td>
</tr>
<tr>
<td>Vegetation</td>
<td>8.4</td>
<td>4.0</td>
<td>1.0</td>
<td>1.4</td>
</tr>
<tr>
<td><strong>Source strength</strong></td>
<td><strong>2.87 \times 10^{10}</strong></td>
<td><strong>1.55 \times 10^{10}</strong></td>
<td><strong>8.43 \times 10^{10}</strong></td>
<td><strong>1.24 \times 10^{10}</strong></td>
</tr>
</tbody>
</table>

A plot of the air concentrations against distance from the centre of the SRP on log-log paper is nearly a straight line. The slope of the line is a little less than the inverse of the distance that would result from radial advection. The vegetation samples are more scattered than the air concentration, and the ratio of air water concentration to vegetation water concentration varies between years. Results from transects of vegetation water taken in the four cardinal directions during June, July and August of 1973 have a logarithmic slope greater than the inverse of distance from the centre of the SRP. These transects were taken inside the boundary of the SRP from distances of 1.6 to 16 km.

Tritium releases over 99% HT in 1974 and 1975 were sampled to help understand tritium distribution in the environment. The environmental samples have already been presented and discussed [5, 6] so only a summary of main findings will be presented here.

The highest tritium concentration was found in the soil water (9000 pCi/ml) after the May, 1974, release. The highest vegetation concentration in the vegetation samples was in grass (1600 pCi/ml), and the concentration in pine trees had a maximum of 700 pCi/ml. Measurements made following the release indicated that a band of tritiated water was formed at the soil surface and then displaced downward by rain. The concentration in the trees decreased exponentially with time with half-lives of 2.45 and 22.7 days. The high concentration in the soil, compared with the vegetation, suggests that HT may be converted to HTO in
TABLE II. TRITIATED WATER DISTRIBUTION IN A FOREST ECOSYSTEM (pCi/ml)

<table>
<thead>
<tr>
<th>Ecosystem part</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pine needles</td>
<td>160</td>
</tr>
<tr>
<td>Pine branch</td>
<td>114</td>
</tr>
<tr>
<td>Pine trunk</td>
<td>127</td>
</tr>
<tr>
<td>Forest floor</td>
<td>166</td>
</tr>
<tr>
<td>Soil</td>
<td></td>
</tr>
<tr>
<td>0.0 – 8.0 cm</td>
<td>150</td>
</tr>
<tr>
<td>8.0 – 13.0</td>
<td>92</td>
</tr>
<tr>
<td>15.0 – 23.0</td>
<td>76</td>
</tr>
<tr>
<td>23.0 – 30.0</td>
<td>61</td>
</tr>
</tbody>
</table>

the soil. Conversion of HT to HTO at a slower rate in the vegetation is also possible; however, the concentrations in the vegetation can be explained by the vegetation coming into equilibrium with the small percentage of HTO in the release.

Another example of the spatial distribution of HTO is the concentration found in the needles, branch wood, stem and soil near the tritium separations facility (Table II). Sampling was made possible by pulpwod cutting in the area. These samples are likely to be characteristic of the terrestrial ecosystems under normal SRP operating conditions. The highest concentrations are found in the needles and the soil surface and are the likely points of entry from the atmosphere by diffusion. The lower concentrations in the deeper soil could be the result of water of lower concentration entering during rainfall. Rain gauge data indicate that tritium concentration in rain is usually lower than the weekly average concentration in the air. The concentration in the stem and branches should be a weighted average of the concentration in the soil. Since most of the tree roots are near the surface, the stem concentration should be nearer the surface concentration than the deeper soil concentration.

Tritium uptake and cycling experiments

The patterns of tritium distribution observed in the vicinity of the SRP suggest which experiments are necessary to understand how the patterns developed. The major mode of entry into terrestrial ecosystems seems to be from the atmosphere into the vegetation leaves and into the surface soil. The forms of tritium that are important are HT and HTO.
A number of investigations of the uptake of HTO by plant leaves have been conducted [7, 8]. The experiments all show that HTO enters and leaves the plant by the same paths as water. However, the gradient for HTO diffusion is from the atmosphere to the leaf water and thus HTO may diffuse at a rate very different from the diffusion of water vapour in the same leaf [9]. HT uptake by leaves has not been studied as much as HTO exchange [10]. Therefore, we conducted experiments to determine the rate of uptake of HT by pine needles. Groups of pine needles were exposed for one minute to different concentrations of HT [11]. The needles were slipped into a glass test tube while still attached to the tree. The base of the needles were sealed with a split test tube stopper and air was circulated through the tube at a rate of two test tube volumes a second. HT was added to the entrance air stream.

While there is a good deal of scatter, it appears that tritium uptake is rate limited at the higher concentrations (Fig.1). If we calculate diffusion resistance (assuming a zero internal HT concentration) we find an internal leaf resistance of 2.0 s/cm at the lower concentrations which gets larger as concentration increases. The low internal resistance at lower concentrations suggests an efficient internal uptake mechanism which is saturated at higher concentrations. Because of the low solubility of HT the uptake rate is not high compared with HTO even at the low concentrations.

\textit{FIG.1. Activity of tritium in water and dry matter after exposure to gaseous molecular tritium.}
HTO uptake by the soil is likely to be governed by the same principles as water exchange. However, the high values of HTO concentration found in the soil after releases which were largely HT indicated that the soil was a significant sink for HT. This has since been proven [12–14]. The action of micro-organisms has been implicated in the conversion of HT to HTO in soils. More work is necessary to make estimates of the conversion rate over a variety of conditions, but our calculations suggest a tritium deposition velocity (flux/concentration) of 0.4 cm/s which is equally divided between soil and vegetation uptake [15].

An understanding of tritium cycling in terrestrial ecosystems also requires an understanding of tritium movement in soils and plants. Tritium spiking of soil water has demonstrated that HTO movement in soils is dominated by mass flow with some hydrodynamic dispersion [16, 17]. Spiking experiments in trees also indicate that HTO moves up tree stems by mass flow [18, 19]. Dispersion in tree stems is greater than in most soils, because of the large amounts of exchangeable hydrogen in the tree stems.

The results of these experiments help explain the tritium distribution patterns we see in the environment of the SRP. However, they do not prove that the distribution patterns result from exactly these processes. A further, although not conclusive, test is to formulate the processes as a mathematical model and see if the model can simulate some of the patterns observed.

A steady-state model of tritium dispersion and cycling

Model structure and implementation

A mathematical model of tritium dispersion and cycling must contain mathematical statements which express the processes of (1) atmospheric dispersion of HT and HTO, (2) wet and dry deposition (uptake) of HT and HTO by vegetation and soil, (3) movement of HTO in soil, and (4) HTO absorption by vegetation roots from soil and flow through the vegetation. A steady-state model has been developed in this case because it seemed appropriate for comparing averages of environmental samples taken over a year’s time. Transient models have been developed to examine conditions after large accidental releases [20, 21]. However, transient models require environmental data at sampling rates of the duration of the release (15 min in the releases mentioned above).

Annual wind direction is nearly equally distributed at the SRP, although seasonal trends do exist. The annual dispersion by wind is nearly radial and the concentration should vary as the inverse of the radial distance from the plant. The expression used in our model is:

\[ X = \frac{Q}{\mu L 2\pi r} \]  

(1)
where
\[ X = \text{concentration in air (pCi/m}^3) \]
\[ Q = \text{source strength (pCi/s)} \]
\[ \mu = \text{wind speed (m/s)} \]
\[ L = \text{mixing depth of the atmosphere for HT and HTO (m)} \]
\[ r = \text{radial distance from the SRP (m)} \]

A correction for stack height and vertical diffusion is made to the air concentration at the ground using the equations of Novak and Turner [22]. This correction does not seem to be important at distances greater than 3 km.

Dry deposition is expressed by the equation:

\[ F = V_d (X - C) \]

where
\[ F = \text{flux density of material (pCi/m}^2\text{-d)} \]
\[ V_d = \text{deposition velocity (m/d)} \]
\[ C = \text{surface concentration (pCi/m}^3) \]

The classical use of deposition velocity assumes that the surface concentration is zero. This is appropriate when HT is converted to HTO at the surface. However, a zero surface concentration is not appropriate for HTO which may accumulate at the surface. Equations of the form of Eq.(2) are used for deposition of HTO and HT to both the soil and the vegetation.

Wet deposition of HTO is calculated by assuming that rainfall is in equilibrium with air HTO concentration. This probably overestimates wet deposition because much of the rainwater comes from heights in the atmosphere above the direct influence of SRP emissions. Wet deposition of HT is ignored because of the low solubility of hydrogen.

Soil movement is described by a mass balance equation for a surface soil layer and a deep soil layer. Water entering the soil is partitioned between vertical displacement to lower soil depths and absorption by vegetation on the basis of annual evaporation/rainfall ratios. The steady-state assumptions of the model smooths out the transient banded structure seen in field soils. The soil HTO concentrations calculated must be looked at as long-term averages. The balance equation for the surface soil is:

\[ V_{ds}(X_H - C_s H) + V_{dTs}X_T + R_R X_H/H - T C_s - (R_R - T) C_s = 0 \]  (3)
TABLE III. CONSTANT INPUT TO MODEL SIMULATIONS: DEPOSITION VELOCITIES (cm/s)

<table>
<thead>
<tr>
<th>Vegetation</th>
<th>Soil</th>
</tr>
</thead>
<tbody>
<tr>
<td>HT</td>
<td>HTO</td>
</tr>
<tr>
<td>0.2</td>
<td>0.6</td>
</tr>
<tr>
<td>0.2</td>
<td>0.1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Precipitation (cm)</th>
<th>Evaporation (cm)</th>
<th>Temperature (°K)</th>
<th>Humidity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>119</td>
<td>89</td>
<td>18.0</td>
<td>71.0</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Wind speed (cm/s)</th>
<th>Stack height (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.0</td>
<td>63.0</td>
</tr>
</tbody>
</table>

FIG. 2. Model and measured tritium concentrations in air moisture. Mixing layer is 700 m.
where

\[ \begin{align*}
V_{ds} &= \text{deposition velocity of HTO to soil} \\
X_H &= \text{concentration of HTO} \\
C_s &= \text{soil HTO concentration} \\
H &= \text{Henry's law constant for HTO} \\
V_{dTs} &= \text{deposition velocity of HT to soil} \\
X_T &= \text{concentration of HT} \\
R_R &= \text{rainfall rate} \\
T &= \text{transpiration rate}
\end{align*} \]

The equations were coded in the IBM simulation language CSMP and solved using annual average climatic and deposition parameters. Table III summarizes the inputs to the model.

**Model simulations**

Figure 2 shows the calculations of \( \frac{X_{HTO}}{Q_{HTO}} \) for a mixing depth of 700 m. The experimentally determined values of \( \frac{X_{HTO}}{Q_{HTO}} \) for the years 1974–77 are plotted on the same figure.

The plot is not an exactly straight line due to incomplete mixing through the surface layer near the stacks and the addition of HTO converted from HT which increases the amount of HTO as the air moves downwind. The effect of changing HT/HTO at the source on the ratio \( \frac{X_{HTO}}{Q_{HTO}} \) is shown as dashed lines in Fig. 2.

The percentage of HT in the releases is known for 1977 only (16% HT). However, an estimate of the HT percentage can be made for 1974 and 1975. In 1974, 479 000 Ci of the total release were more than 99% HT because of an incident at the tritium separations area. If we assume that the chronic releases contained 16% HT as in 1977, then 40% of the 1974 release was in the form HT. Similar calculation for the 182 000 Ci inadvertently released in 1975 indicate that 47% of the released tritium for the year was HT. We must assume 1976 was a normal year. The inclusion of the HT/HTO ratio brings the dispersion estimates into a better agreement with the measurements, but some unexplained variation is left. The variation is to be expected because the 1974 and 1975 releases were 15-min puffs which do not meet the long averaging period of the steady-state, radial diffusion model. A strategy for a best estimate seems to be selecting a mixing height of about 700 m. This is a little lower than expected from the climatic data which indicate a range of 350–1500 m. The low value of the mixing height may be explained by the fact that the higher values of mixing height occur for only short periods in the middle of the day.

It might be expected that the variation in vegetation HTO concentration is also related to the percentage of HT in the source release. This can be studied by
looking at the ratio of vegetation/air HTO versus percentage of HT in the source air. Figure 3 shows the model calculation and the average ratios from data for the years 1974–77. The ratios calculated above have been changed so that the 1975 inadvertent release will be credited to 1976 since the release took place late on 31 December 1975, and the vegetative effects would be expected over the first months of 1976. The results are surprisingly good. It seems that most of the variation in the ratio of leaf HTO to air HTO is due to the HT percentage.

SUMMARY

The results of analysing the tritium releases by simulation, based on a model constructed from controlled experiments, corroborate the hypothesis that the processes of dispersion, wet and dry deposition, and ecosystem water cycling are dominant in producing the tritium patterns seen in the vicinity of the SRP. The results also emphasize the importance of knowing the form of the tritium in the atmospheric releases.

The model used was developed to investigate chronic releases where the wind direction and speed were uniformly distributed. However, the dispersion is sufficiently linear that long-term averages of samples taken uniformly around
a point source should also show very similar radial distribution patterns. This may explain why the puff releases of HT in 1974 and 1975 do not seem to cause a large deviation from the radial pattern.

Dose estimates with radial population distributions are possible when the distribution of wind speed and direction are uniform. It is possible to make dose estimates for non-uniform distributions; however, the population must be weighted by the wind speed and direction.

ACKNOWLEDGEMENTS

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DISCUSSION

W. ROETHER: From your deposition velocities, and also from your Fig.3, it seems to me that in your analysis both soil and vegetation surfaces are regarded as nearly black, i.e. any TH molecule that hits the surface is converted to HTO; could you comment on this?

C.E. MURPHY: At the HT concentration we are dealing with, the soil is a very efficient sink for all cases we have studied. The soil uptake is, therefore, diffusion limited. Experiments we have done on pine needles indicate that at low concentrations the needles are fairly efficient sinks, but as the concentration increases the efficiency decreases until a constant uptake rate is reached at relatively low concentrations.

J.A. GARLAND: We have made some measurements at Harwell of tritium oxidation rates in various soils. We found deposition velocities up to 0.1 cm·s⁻¹ to soil and no uptake by vegetation, so our results suggest rather slower conversion than yours.

C.E. MURPHY: The deposition rates to the soil are extrapolated from laboratory data that have been corrected for field diffusion conditions. We have not looked at variation of uptake with the environment in the field; I suspect that there is in fact a great deal of variation. The deposition to the vegetation is higher than I would have expected but is based on chamber measurements made under field conditions.

J.P. CORLEY: Do I understand correctly that in your Fig.3 the ratio HT/(HT + HTO) is the source ratio and is not derived from field measurements at the HTO sampling locations? Also, is it correct that no allowance is made for conversion of HT to HTO with travel?

C.E. MURPHY: Yes, the figure refers to parts ratios at the source. The model does take into account the change in ratio with distance from the source. However, we have no data against which to check our tritium parts ratio.
INFLUENCE OF THE RATE OF
CONVERSION OF HT TO HTO ON
PROJECTED RADIATION DOSES FROM
RELEASE OF MOLECULAR TRITIUM*

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United States of America

Abstract

INFLUENCE OF THE RATE OF CONVERSION OF HT TO HTO ON PROJECTED RADIATION DOSES FROM RELEASE OF MOLECULAR TRITIUM.

Releases of tritium in the past have been largely in the form of tritiated water, and the projected radiation doses could be estimated by assuming tritium behaviour to parallel that of water. There is increasing interest in potential releases of tritium in the form of HT because of significant recent advances in fusion reactor research. Several recent studies have shown that bacteria containing the enzyme hydrogenase can catalyse the conversion of HT to HTO at rates several orders of magnitude faster than the rates measured in atmospheric systems. Rates of conversion in the soil have been combined with estimates of rates of permeation of HT into the soil and with global and local models depicting tritium transport and cycling. The results suggest that for the expected conversion rates, the impact on projected radiation doses should be relatively minor.

INTRODUCTION

It is generally assumed for estimates of environmental radiation exposure that tritium released into the environment is in the form of water vapor and would rapidly assume the same distribution as natural tritium. This has largely been true with the exception of several rather large releases of molecular tritium from the Savannah River Plant Production facility and the Lawrence Livermore Laboratory weapons program. Recent and anticipated advances in fusion research portend numerous future facilities each with inventories of tritium comparable to the natural inventory of 7 to 14 kg. We note that a fusion power plant may contain up to 10 kg (10^5 curies) of molecular tritium. Although limitation of release rates to ~1 to 10 Ci/day seems technically possible, there is not yet any definitive information

concerning the economic feasibility of such release rates. Hence it is appropriate to understand potential effects of chronic releases of tritium gas into the environment.

An adequate understanding of the potential radiological effects to man from a release of tritium involves several considerations. Not only must one understand the global movement and distribution of tritium depending on its form of release and the location of discharge, but knowledge is also required of the local effects as a function of form at release. Tritiated water is more readily incorporated into the human body than is molecular tritium [1], and the maximum permissible concentration of HTO in air for nonoccupational exposure is lower by a factor of 200 than for HT [2]. Thus, potential processes and rates of conversion should be considered in developing models for assessment of releases of tritium gas.

CONVERSION PROCESSES AND RATES

Physical Conversion

Eakins and Hutchinson [3] measured the conversion of HT to HTO at a concentration of $2 \times 10^{-2}$ mCi/cm$^3$ in the presence of various metal surfaces and estimated reaction half-times, extrapolated to a concentration of $3.4 \times 10^{-2}$ uCi/cm$^3$, ranging from 4 years in the presence of platinum to 1150 years for glass. Phillips and Easterly [4] reported a first order reaction rate constant of $4 \times 10^{-5}$ ml/mCi, which yields a reaction half-time of about 8 years; this is consistent with the range of 6-10 years residence time of HT in the atmosphere estimated by Mason [5].

Biological Conversion

The ability of enzyme systems of certain bacteria to catalyze the reduction of specific compounds to molecular hydrogen has been known since the 1930's [6] and metabolic oxidation of tritium gas was observed in the early 1950's [7,8]. However, not much attention has been paid to the role of metabolic conversion of tritium as it relates to a potential increase in radiation doses to humans. Rates of oxidation have been measured in various systems, from bacterial suspensions to whole mammals. Smith and Marshall [7] observed the conversion in 24 h of 41 of the 43 mCi of tritium introduced into a 100 ml suspension of Anotobacter vinelandii containing 4 mg cellular nitrogen per ml, demonstrating that bacteria containing hydrogenase are potential catalysts for tritium conversion.

Studies with several plants and soils have shown increasing evidence that soils, and in particular, the bacteria in soils, are important in catalyzing the oxidation process [9,10,11,12]. Liebl [13] reported rates of H$_2$ incorporation into soil of $2 \times 10^{-12}$ to $4 \times 10^{-12}$ g H$_2$/cm$^2$·s. Comparable rates of oxidation of HT in soil columns was found by McFarlane et al. [12]. If these rates are integrated over the land surface of the earth - excluding the polar regions - the conversion of H$_2$ would be about $114 \times 10^{12}$ g/a, which is about 71.5% of the tropospheric inventory [14]. Such a rapid conversion
TABLE I. GLOBAL MOLECULAR HYDROGEN BUDGET DATA

<table>
<thead>
<tr>
<th>Sources of $H_2$</th>
<th>$(10^{12} \text{ g } H_2/\text{a})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anthropogenic activity</td>
<td>13.0</td>
</tr>
<tr>
<td>Oceans</td>
<td>4.0</td>
</tr>
<tr>
<td>Photochemistry (atmosphere)</td>
<td>4.6 - 9.2</td>
</tr>
<tr>
<td>Volcanoes</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>21.7 - 26.3</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sinks for conversion of $H_2$ to $H_2O$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photochemistry (atmosphere)</td>
</tr>
<tr>
<td>Soils</td>
</tr>
<tr>
<td></td>
</tr>
</tbody>
</table>

* Adapted from Ref.[14].

rate is not consistent with the chemical decay constant of 10.2 to 15.5% per year for HT estimated on the basis of interhemispheric exchange [5].

McFarlane et al.[12] also have presented data indicating that HT in immediate contact with soil can be converted to HTO at a rate of about 20% per hour; their more recent data [15] shows a range in the conversion rates for various soils of 12 to 66% per hour or conversion half-times of 0.6 to 5.4 hours. Other laboratory data obtained with plants and soils suggest conversion half-times up to about 24 hours [9,12].

Diffusion of Tritium Gas into the Soil

Although these rates of conversion appear to be quite rapid, one must consider them in the context of the rate of penetration of HT into the soil from the atmosphere. The coefficient of diffusion of hydrogen gas in air is 0.611 cm$^2$/s(NTP) [16]. Heavier gases diffuse at rates inversely proportional to the square root of their density. Using the relationships presented in the Chemical Engineer's Handbook [17], it is estimated that the coefficient for diffusion of HT in air should be approximately 0.42 cm$^2$/s(NTP). In a porous medium, corrections must be made for porosity and for the tortuous path the molecule must follow to move around the solid particles. The effective diffusion coefficient is given by:

$$D_{\text{eff}} = \frac{D_0}{\tau}$$

where $D$ is the molecular diffusion coefficient in the gaseous phase, $\phi$ is the effective porosity of the porous medium, and $\tau$ is the
FIG. 1. Global hydrologic cycle with respect to man.
tortuosity factor. In our estimate we assume $D = 0.4 \text{ cm}^2/\text{s}$, $\varphi = 0.2$ and $\tau = 1.5$, yielding an effective soil diffision coefficient of $\approx 0.05 \text{ cm}^2/\text{s}$.

The mean penetration into the soil in a given time is given by:

$$\bar{x} = \sqrt{2D_{\text{eff}} t}$$

For a three-hour time period, the half-time for a conversion rate of 20% per hour, this corresponds to a mean penetration of about 30 cm. If this rate of penetration represents the average, it is estimated that the global rate of biological conversion in soil would be about 0.7% of the atmospheric inventory per year.

Conversion of HT in Vegetative Cover

Tritium gas can be converted to tritiated water in vegetative cover as well as in soils [9,10,11,12,18]. However, the rates of conversion appear to be significantly lower than in soil systems; McFarlane et al. [12] report conversion of about 1% in a 48 h exposure period. Even assuming a relative accessible surface area for vegetative cover over the earth's surface of ten times that of the land area occupied, it seems doubtful that conversion in the vegetative cover would be as great as in the underlying soil.

Conversion of HT in the Oceans

The solubility of hydrogen gas in water is fairly low, on the order of 2% of that in an equivalent volume of air in direct contact. Assuming rapid mixing and saturation of the surface 75 meters of the ocean, the inventory of HT in the ocean would be about $1.5 \times 10^{22}$% of that in the atmosphere. The mixing time of the ocean surface 75 meters is about 3.5 years [18], so that we can estimate an upper limit for the rate of conversion in the oceans of about $4 \times 10^{-3}$% of the atmospheric inventory per year, if all tritium entering the ocean were converted to HT0. Actually, the surface waters of the oceans are generally supersaturated with respect to H$_2$, indicating that they serve as a source rather than a sink for hydrogen gas [14].

Summary of Sources and Sinks for HT Conversion

Schmidt has attempted to estimate the magnitude of various sources and sinks for hydrogen conversion [14]. His values, based on measurements, are summarized in Table I. Based upon his estimates of sources and a tropospheric inventory of $160 \times 10^{12}$ g H$_2$, he estimates a tropospheric residence time for H$_2$ of 6.1 to 7.4 years. This agrees quite well with the range of 6 to 10 years estimated by Mason on the basis of measurements of interhemispheric exchange of HT [5] and with the rates of HT to HTO conversion measured by Phillips and Easterly [4]. However, Schmidt's estimated rate of conversion of H$_2$ to H$_2$O in the soil seems much too high, both when compared to the total sources of H$_2$ and when compared to residence time. He points out that field conversion rates are likely to be much lower than the rates measured in the laboratory by Liebl [13].
This is born out by the later studies of McFarlane et al. [12]. To balance the sources, the rate of conversion in the soil should be of the order of $15 \times 10^{12}$ to $20 \times 10^{12}$ g H$_2$/a. Estimates based on rates of conversion measured by McFarlane et al. [12, 15], limited by diffusion into the soil, are on the order of $1.1 \times 10^{12}$ g H$_2$/a.

MODELS FOR ENVIRONMENTAL CYCLING OF TRITIUM

Global Model

A generalized model for tritium cycling, based on the hydrological cycle, has been employed by Easterly and Jacobs [19] to estimate the effect on the collective radiation dose to the world population following discharge of tritium into various environmental compartments. A schematic of the cycle is presented in Figure 1. The water compartments are assumed to mix uniformly and instantaneously and to interact with each other in a linear manner. For a set of conditions (for example, a globally uniform input of tritiated water into one or more compartments), the equations can be solved and evaluated for the relative compartmental distributions at subsequent times. From this model, it is estimated that tritiated water releases to the atmosphere would give rise to collective radiation doses less by a factor of 10 than releases into surface waters (See Table II). The projected collective dose for atmospheric release of tritium gas is within a factor of two of that for tritiated water vapor, when atmospheric conversion half-times of up to 10 years are considered. If the primary compartment for conversion is the soil, the tritiated water formed may be in closer contact with man and the projected collective dose slightly higher, perhaps up to a factor of two. Both the preliminary data of Phillips and Easterly [4] and atmospheric residence time estimates of Mason [5] indicate a slow conversion in the atmosphere with half-times on the order of 5 to 10 years. Hence for the global scale, if releases are to the atmosphere, estimates of population dose are little affected by uncertainties in the conversion rate.

Local Model

Although our estimates indicate that the rate of conversion of HT to HTO does not have a major impact upon projected radiation doses from tritium releases when considered on a global scale, the same question needs to be addressed from a local perspective.

The calculations of radiation dose downwind from a molecular tritium release depends on numerous imprecisely known variables. The greatest uncertainties lie in the relative HTO/HT dose conversion factors and the rate of HT to HTO conversion. To test the sensitivity of the conversion rate, an atmospheric transport computer code, ACRA-II [20], was modified to include the conversion of HT to HTO. The code uses a three-dimensional normal distribution (Gaussian) model to simulate transport of material through the environment. The conversion rate is assumed to follow a first-order law; this is simulated by means of a pseudo-radioactive decay of HT with the daughter product being HTO. Sample calculations have
TABLE II. RELATIVE PERCENTAGE OF TRITIUM IN VARIOUS HYDROLOGIC COMPARTMENTS AFTER 100 YEARS' CONTINUOUS RELEASE OF TRITIATED WATER INTO THE ATMOSPHERE OR SURFACE WATERS

<table>
<thead>
<tr>
<th>Hydrologic compartment of accumulation</th>
<th>Compartment of release</th>
<th>Atmosphere</th>
<th>Surface water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmosphere</td>
<td></td>
<td>0.22</td>
<td>0.022</td>
</tr>
<tr>
<td>Vadose water</td>
<td></td>
<td>0.87</td>
<td>0.090</td>
</tr>
<tr>
<td>Deep groundwater</td>
<td></td>
<td>5.6</td>
<td>0.57</td>
</tr>
<tr>
<td>Surface water</td>
<td></td>
<td>0.017</td>
<td>0.19</td>
</tr>
<tr>
<td>Ocean surface</td>
<td></td>
<td>46.0</td>
<td>49.0</td>
</tr>
<tr>
<td>Deep ocean</td>
<td></td>
<td>47.0</td>
<td>50.0</td>
</tr>
<tr>
<td>Man</td>
<td></td>
<td>$2.5 \times 10^{-6}$</td>
<td>$1.5 \times 10^{-5}$</td>
</tr>
</tbody>
</table>

a Water in earth's crust above the permanent water table.

---

![Graph](image-url)

**FIG. 2.** Relative risk for HT and HTO versus distance for varying conversion rates for a release height of 0 m, a wind speed of 1.7 m/s and C-stability.
been made for various conditions of stability, height of release, wind-speed and rates of conversion.

The relative risks resulting from a release of HT from heights of 0 and 100 m are shown in Figures 2 and 3, respectively, for HT to HTO conversion rates of 0, 0.1, 1 and 10% per day. We feel that it is unlikely that more than 10% of the tritium gas released from an elevated source would come into immediate contact with the soil daily due to the limitations imposed by atmospheric mixing. The relative risk for 0% per day conversion is numerically equal to 1/10 in s/m³. Numerical values of relative risk are related to dose by a constant factor. The hazard from HTO is taken to be a factor of 200 times that for HT based upon ICRP recommendations [2].

Values along the conversion curves are found by multiplying the HTO concentration by 200 and adding the product to the remaining concentration of HT. Thus, each curve represents the total relative risk for that conversion rate.

Doses can be calculated by multiplying the relative risk index by the HT release rate (Ci/s), the exposure time (s), and the dose conversion factor for HT (rem Ci⁻¹ s⁻¹ m⁻³), which we have assumed to be a factor of 200 lower than that for HTO.
From Figures 2 and 3, it is seen that for distances up to 5 km from the point of release, the difference in risk at any point between 10% per day conversion and no conversion is less than a factor of two; at 100 km the difference is a factor of 14. Virtually the same results are obtained for both 0 and 100 m release heights and the difference caused by a deposition velocity for HTO of 1 cm/s compared to 0 cm/s is at most 15%. This occurs at a distance of 100 km where the concentration has been reduced about 9 orders of magnitude below the initial concentration.

If one further considers the land area affected, as an approximation of the number of people potentially exposed, the difference in risk between 0 and 10% conversion per day is less than a factor of 2 within 10 km and a factor of 5 within 100 km.

On the basis of our assumptions, we conclude that the rate of conversion of HT to HTO does not have a significant impact on projected radiation doses to the population near the point of release (within 10 km) nor on a global scale. The relative impact may be greatest in the region from 10 km up to the point where deposition into the ocean becomes significant.

SUMMARY

Recent studies have shown that biological systems, particularly the bacteria in soils, can catalyze the oxidation of HT to HTO at rates which are several orders of magnitude faster than the rates measured in atmospheric systems. However, the movement of HT from the atmosphere into contact with the soil is likely to be the rate limiting step. When the anticipated ranges of conversion rates are incorporated into models for global or local transport and cycling, it appears that the impact on projected radiation dose is relatively minor.

REFERENCES


DISCUSSION

A.A. MOGHISSI: Did you base your dosimetry on the specific activity concept or on 10-day turnover rate? This is an important point, because under environmental conditions the latter approach would be inappropriate because of the introduction of tritium into the organic fraction of the body. If you are making a comparison (HT versus HTS), this would not make any significant difference; however, the absolute dose values would not be free of error.

D.G. JACOBS: For global models of tritium circulation, we assumed that the collective radiation dose to man would be proportional to the quantity of tritium in the compartment designated man; this is equivalent to using the specific activity concept. For local models, we did not calculate radiation dose but computed a relative hazard index by multiplying the concentration of HTO in air by 200 and adding the product to the concentration of HT in air at the same point.

T. SHOKAWA: In soil, in such a condensed phase, I believe that the chemically active fragments originating from the beta decay of tritium also
play a very important role in the conversion not only of T₂ but also of HT to HTO. What is your opinion of this?

D.G. JACOBS: Many chemical reactions involving tritium can be postulated as occurring in soil, including conversion of tritium gas to tritiated water or tritiated hydrocarbons as well as the reverse reactions.

I doubt, however, whether the concentration of tritium in the soil environment would be high enough for the active fragments from beta decay of tritium to play an important role in the conversion reactions.
BEHAVIOUR OF TRITIUM IN THE PLANT DOMAIN*

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Abstract

BEHAVIOUR OF TRITIUM IN THE PLANT DOMAIN.

An attempt is made to review in three different ways the present status regarding the uptake, retention and release patterns of plants of different trophic levels exposed to a tritiated atmosphere: stem injection, soil application and foliar absorption, as a result of studies conducted in ten countries under the research programme co-ordinated by the IAEA. Three component release patterns for tissue-free water tritium (TFWT) are observed in many of the terrestrial vegetations. The mean residence time for the first major component is found to be within 1 – 12 days for all the terrestrial vegetations. The second and the third components form only a small fraction of the total tritium uptake and show significantly different mean residence times. The aquatic plant, *Hydrilla verticillata*, also shows very small residence time for the first component (0.4 h). An isotopic discrimination of about 30 – 40% against the incorporation of tissue-bound tritium (TBT) is found in some algal and aquatic species, and this is enhanced to as high as 90 – 99% for the terrestrial vegetations. Though the data indicate a regular gradation in the levels of fixation of TBT from aquatic plants to the terrestrial

* Part of the IAEA Co-ordinated Research Programme on the Behaviour of Tritium in the Environment.
vegetations, this seems to be the result of a single exposure or semi-chronic conditions used in the latter case. When conditions of constant specific activity are maintained, even for the terrestrial vegetations high specific activity ratios are obtained in the few cases cited. A synoptic view of the distribution data in different biochemical fractions indicates significant relative specific activity ratios in nucleic acids, protein and fats. The nucleic acid fraction has shown relative specific activity ratios of 0.37 and 1.0 for pea seedlings and algae respectively. In maize, though carbohydrate is a major fraction fat has shown maximum fixation of TBT. Mathematical resolution procedures to determine the mean residence times for TFWT/TBT fractions either from separated or composite sample fractions have been reviewed. TFWT time-activity profiles are built up for succulent plants from diffusion, xylem flow and the utilization of TFWT in the synthesis of TBT. A good agreement between the calculated and experimental values is obtained using molecular diffusion coefficients for succulent plants. The applicability of this concept for larger trees is discussed.

INTRODUCTION

The importance of the distribution of tritium in the ecosystems in general and in food chains in particular has received attention with the increased use of atomic energy for peaceful purposes [1]. Tritium enters the hydrosphere in the form of HTO and the atmosphere in the form of HTO vapour and HT. The elemental form is oxidized in the atmosphere and joins the overall water cycle [2]. Though the capacity of the hydrosphere would seem infinite for dilution, its interaction in the biological systems is of interest since hydrogen is one of the essential elements in all living systems. The two major pools of hydrogen in plants where tritium can be incorporated are tissue water and organic molecules, the former being the larger fraction. Its incorporation in the genetic constituents, i.e. DNA/RNA, has also been demonstrated [3, 4]. Generally tissue-free water tritium (TFWT) attains equilibration with the medium quickly, but the rate of incorporation of tissue bound tritium (TBT) and its turnover may vary in different systems to different extents depending upon the metabolic rates and biochemical functions.

The International Atomic Energy Agency has co-ordinated a research programme to study the behaviour of tritium in the environment. The principal aim of the programme is to obtain information on the residence times, pattern of movement and its distribution in the biosphere in order to understand the biological significance of this radionuclide under varying climatic conditions. The participant countries were Belgium, Finland, France, the Federal Republic of Germany, India, Mexico, the Netherlands, the Philippines, Thailand and the United States of America.

Data on the uptake, retention and release patterns of TFWT and TBT in different trophic levels of the plant domain under varied climatic conditions are discussed. The data represent the realistic behaviour of tritium under a wide range of environments. Mathematical models have been developed to understand the movement of tritium and its distribution in plants.
GENERAL METHODOLOGY

Field work

Plants and trees in Finland, Belgium, the Federal Republic of Germany and the Netherlands, tropical trees and vegetables in India, mediterranean trees in France, cereals and agricultural products in Mexico, vegetables and cereals in the Philippines and Thailand are the general species exposed to tritiated water in spraying and injection experiments for the study of mean residence times. Studies with foliar uptake, both under laboratory and field conditions, are also carried out in the Federal Republic of Germany (FRG) and India. Tritium is introduced into the experimental plot by a spraying device to create uniform deposition of activity per unit area. Small pressurized tanks, sprayers or manual sprinklers or irrigation type sprinklers are used depending on the areas of experimental plots used. With trees, tritium is injected into the stem/trunk or by the root soil irrigation method. Data obtained on the mean residence times for different vegetation are summarized in Table I.

Sample preparations

The tissue-free water tritium is generally collected by vacuum distillation followed by freeze-drying, though one participant has adopted the azeotropic distillation method. The organic bound tritium is analysed by a variety of combustion methods ranging from the conventional quartz tube furnace with flowing oxygen to the Schoniger closed flask method. Tritium in tissue water and combustion water is assayed by liquid scintillation counting using well-established quench correction methods.

BEHAVIOUR OF TRITIUM IN DIFFERENT PLANT SYSTEMS

Aquatic plants

Tritium uptake by aquatic plants such as algae and water grass are studied on laboratory cultures. The specific activity ratios TFWT and TBT to that in the medium are given in Table II. The tissue-bound tritium in the water grass H. verticellata and T. angustiphobla [5] show 1 – 2% discrimination against incorporation of tritium. The algal species Chlamydomonas and S. obliquus show a discrimination of 35 – 40%. Discrimination of similar orders are reported by Rosenthal and Stewart [6] for some algal species. Even the value of 50% discrimination observed by Weinberger and Porter [7] is not very far from the above values. Since the algal species form the source material in the food chain
<table>
<thead>
<tr>
<th>Country</th>
<th>Terrestrial species studied</th>
<th>Mean residence times, first component</th>
<th>Experimental conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Belgium</td>
<td>Pasture (1)</td>
<td>5 d</td>
<td>Temperate irrigation</td>
</tr>
<tr>
<td>Finland</td>
<td>Vegetable (1)</td>
<td>–</td>
<td>Arctic</td>
</tr>
<tr>
<td></td>
<td>Trees (5)</td>
<td>9 – 23 d</td>
<td>spray</td>
</tr>
<tr>
<td></td>
<td>Trees (3)</td>
<td>1 – 8 d</td>
<td>Mediterranean</td>
</tr>
<tr>
<td></td>
<td>Fruits (2)</td>
<td>30 – 120 d</td>
<td>spray</td>
</tr>
<tr>
<td>Federal Republic of Germany</td>
<td>Trees (3)</td>
<td>2 – 6 d</td>
<td>Temperate vapour exchange</td>
</tr>
<tr>
<td>India</td>
<td>Water grass (1)</td>
<td>0.4 h</td>
<td>Tropical immersion</td>
</tr>
<tr>
<td></td>
<td>Succulent (3)</td>
<td>72 – 115 d</td>
<td>Injection</td>
</tr>
<tr>
<td></td>
<td>Vegetables (6)</td>
<td>1 – 2 d</td>
<td>Spray and vapour</td>
</tr>
<tr>
<td></td>
<td>Trees (9)</td>
<td>0.3 – 3 d</td>
<td>exchange</td>
</tr>
<tr>
<td>Mexico</td>
<td>Vegetables (2)</td>
<td>4 – 12 d</td>
<td>Temperate, spray, residual tritium</td>
</tr>
<tr>
<td></td>
<td>Cereals (2)</td>
<td>7 – 15 d</td>
<td></td>
</tr>
<tr>
<td>Philippines&lt;sup&gt;b&lt;/sup&gt;</td>
<td>Vegetables (5)</td>
<td>4 – 18 d</td>
<td>Tropical</td>
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<tr>
<td></td>
<td>Cereals (2)</td>
<td>5 – 13 d</td>
<td>spray</td>
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<td>Thailand&lt;sup&gt;b&lt;/sup&gt;</td>
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<td>7 – 12 d</td>
<td>Tropical, spray</td>
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<td>United States of America</td>
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<td>Cereal (1)</td>
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<td></td>
<td>Weed (1)</td>
<td>0.9 h</td>
<td>vapour exchange</td>
</tr>
</tbody>
</table>

<sup>a</sup> Numbers in the brackets indicate number of species studied.

<sup>b</sup> Half-life values.
TABLE II. TRITIUM INCORPORATION IN AQUATIC PLANTS

<table>
<thead>
<tr>
<th>Species</th>
<th>Medium conc. and nature (μCi/ml)</th>
<th>Specific activity ratio</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>TFWT medium</td>
<td>TBT TFWT</td>
</tr>
<tr>
<td>Chlamydomonas</td>
<td>Sea-water 0.61 – 4.1</td>
<td>—</td>
<td>0.61 – 0.68</td>
</tr>
<tr>
<td>Scenedesmus obliquus</td>
<td>Fresh water 0.6 – 3.0</td>
<td>—</td>
<td>0.57 – 0.64</td>
</tr>
<tr>
<td>Hydrilla verticillata</td>
<td>Fresh water 0.50 – 50.0</td>
<td>0.85</td>
<td>0.98</td>
</tr>
<tr>
<td>Typha angustiphobia</td>
<td>Fresh water 0.58 – 0.70</td>
<td>0.58 – 0.70</td>
<td>0.60 – 0.99</td>
</tr>
<tr>
<td>Pithaphora</td>
<td>Fresh water</td>
<td>1.02</td>
<td>0.98</td>
</tr>
</tbody>
</table>

a Mean residence times of 0.016 and 16 days for TFWT and TBT respectively.

of the aquatic environment, the discrimination noted above could be expected to lead to lower specific activity ratios in the bound compartment in higher trophic level organisms. However, the high TBT incorporation levels shown by the water grass may be explained in terms of variation in the experimental exposure conditions and species specificity. The mean residence times for TFWT are not reported for algae whereas 0.4 h residence time is observed for H. verticillata.

Succulent plants

Succulent plants are of different class from aquatic plants in view of their adaptability when water is lacking. Experiments are conducted under water stress conditions to determine the mean residence times for TFWT and TBT and the results are presented in Table III. The stem of Opuntia sp. has shown significant incorporation of TBT, 10% of TFWT on a dry weight basis compared with the stem of the other two plants. The leaves of E. mili, however, have shown the same level of TBT as the stem of Opuntia. This indicates that the function of photosynthesis is carried out in such plants by the stem where the leaves are absent. The large residence times for TFWT (72 – 112 d) facilitate the synthesis of bound tritium to a greater extent. The residence times for TBT are also long,
TABLE III. TRITIUM PERSISTENCE IN SUCCULENT PLANTS

<table>
<thead>
<tr>
<th>Species</th>
<th>Tritium peak conc.</th>
<th>Mean residence times</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>TFWT (µCi/ml)</td>
<td>TBT (µCi/g)</td>
</tr>
<tr>
<td>Opuntia sp.</td>
<td>2.66</td>
<td>0.27</td>
</tr>
<tr>
<td>Euphorbia trigona</td>
<td>1.15</td>
<td>0.03</td>
</tr>
<tr>
<td>Euphorbia mili</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stem</td>
<td>1.55</td>
<td>0.03</td>
</tr>
<tr>
<td>Leaf</td>
<td>1.29</td>
<td>0.11</td>
</tr>
</tbody>
</table>

a Single injection of 2 mCi, peak arrival time 70 – 80 days.

ranging from six to twelve months. The transpiration rates calculated from the tritium tracer kinetics [8, 9] are of the order of 0.001 ml per gram wet weight per day for all the three plants, which is nearly hundred times less than that for flowering plants [10] and trees [11].

Vegetables

Soil spray

The mean residence times for radish, R. sativus, and the medicinal plant, P. fraternus, are about two days whereas the leafy vegetable A. viridis has considerably shorter residence time, 0.1 d (Table IV). The residence time for radish obtained by Yuthamanop [12] is somewhat higher (13.5 d). This may be due to the different exposure conditions and seasonal influences. Similarly tomato, L. esculentum, has been studied both under tropical and temperate environmental conditions. The residence times in the tropical climate range from 6 to 14 days. The large residence time of 60 days observed under the Mexican temperate climate is due to the organic bound tritium arising from the slow uptake of residual soil water activity (0.3 – 0.4 nCi/g soil) [5]. In general mean residence times for vegetables and fruit-yielding plants vary from 2 to 26 days. The mean residence time for sweet potato, E. batatas, (26 d) is found to be higher compared with all other species studied in the Philippines [13].
<table>
<thead>
<tr>
<th>Species</th>
<th>Mean residence times (d)</th>
<th>Remarks</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tomato, <em>Lycopersicum esculentum</em></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>–</td>
<td>60</td>
<td>Roots &amp;</td>
</tr>
<tr>
<td></td>
<td>–</td>
<td>25</td>
<td>stem</td>
</tr>
<tr>
<td></td>
<td>–</td>
<td>40</td>
<td>Leaf</td>
</tr>
<tr>
<td></td>
<td>3.9</td>
<td>–</td>
<td>Fruit</td>
</tr>
<tr>
<td></td>
<td>12.5</td>
<td>28</td>
<td>Leaf</td>
</tr>
<tr>
<td></td>
<td>14.0</td>
<td>31</td>
<td>Fruit</td>
</tr>
<tr>
<td>Radish, <em>Raphanus sativus</em></td>
<td>2.2</td>
<td>–</td>
<td>Stem</td>
</tr>
<tr>
<td></td>
<td>2.9</td>
<td>–</td>
<td>Roots</td>
</tr>
<tr>
<td></td>
<td>13.5</td>
<td>30</td>
<td>–</td>
</tr>
<tr>
<td>Maithuchi bhaji, <em>Amaranthus viridis</em></td>
<td>0.1</td>
<td>10</td>
<td>Whole</td>
</tr>
<tr>
<td>Medicinal plant, <em>Phyllanthus fraternus</em></td>
<td>1.7</td>
<td>14</td>
<td>Leaf</td>
</tr>
<tr>
<td></td>
<td>2.1</td>
<td>13</td>
<td>Stem</td>
</tr>
<tr>
<td></td>
<td>7.7</td>
<td>–</td>
<td>Roots</td>
</tr>
<tr>
<td>Beans, <em>Phaseolus vulgaris</em></td>
<td>3.0</td>
<td>17</td>
<td>23</td>
</tr>
<tr>
<td>Mung beans, <em>Phaseolus aureus roxb.</em></td>
<td>15.0</td>
<td>–</td>
<td>Roots &amp;</td>
</tr>
<tr>
<td></td>
<td></td>
<td>–</td>
<td>stem</td>
</tr>
<tr>
<td>Soyabean, <em>Glycine Max L.</em></td>
<td>10.0</td>
<td>–</td>
<td>Leaf</td>
</tr>
<tr>
<td></td>
<td>17.0</td>
<td>–</td>
<td>Fruit</td>
</tr>
<tr>
<td>Sweet potato, <em>Epomaea batatas</em> L.</td>
<td>7.3</td>
<td>–</td>
<td>Roots &amp;</td>
</tr>
<tr>
<td></td>
<td></td>
<td>–</td>
<td>stem</td>
</tr>
<tr>
<td>Chinese cabbage, <em>Brassica compestris</em></td>
<td>18.0</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Cowpea, <em>Vigna sinensis savi</em> L.</td>
<td>7.5</td>
<td>–</td>
<td>Leaf</td>
</tr>
<tr>
<td></td>
<td>8.0</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Egg plant, <em>Solanum melongena</em></td>
<td>6.6</td>
<td>50</td>
<td>–</td>
</tr>
</tbody>
</table>

* Values for the Philippines and Thailand are half-life values.
All the vegetables, tomato, radish, sweet potato and Chinese cabbage, studied under Thailand climatic conditions [12], are also of the order of 14 days. The fast component residence times of TFWT for most of the garden vegetations are seen to be of the order of 10 – 15 days.

**Foliar uptake**

Concentration factors\(^1\) of 0.10 (stem) and 0.45 (leaves) are obtained in the vapour phase exposures of mirchi, *C. frutescens* (Table V). The lower concentration factors of 0.04 obtained for lettuce and cabbage seedlings are probably due to the small pore openings of the shoots in contrast to the larger pore openings in the leaves of mirchi under active transpirational conditions. Koranda and Martin [14] reported concentration factors varying from 0.17 – 0.49 for eight different plants. McFarlane [15] observed a concentration factor of 0.34 for HTO absorption whereas HT absorption has been found to give 0.84 in the leaves of lettuce plants. The high value in the case of HT absorption is the possible result of a larger influx of the HT molecules into the leaves and their subsequent oxidation to HTO.

The mean residence times for the TFWT component in these experiments have been found to be much shorter, i.e. 0.4 days in comparison with the fast component residence times under soil irrigation conditions. Though there is a possibility of more than one component for TFWT, components of significantly different residence times could not be ascertained in the present experiments. Koranda and Martin [14] have observed a 270-h component which is attributed to the metabolism of TBT. In foliar absorptions, 3 – 4% TBT levels have been obtained for an exposure time of 2 hours. The behaviour of tritium is qualitatively similar for vapour or liquid phase exposures with the major part of tritium being associated with a short TFWT residence time and a small percentage with long residence time which might be attributed to organic fraction.

**Trees**

The uptake, retention and release patterns of tritium injected into the stem of large plants and trees have been investigated and the results are presented in Table VI. Parameters governing the biomass, peak arrival times and evapotranspiration rates have also been derived for some of these trees on the basis of tracer kinetics [16]. Under Indian tropical conditions, badam, mango, sapota, ashok, norfolk pine, banana, coconut, arecanut palm and casuarina show fast component turnover times of 0.3 – 3.0 days. Under Finland conditions [17],

---

\(^1\) The concentration factor is defined as the ratio of μCi/ml of TFWT to μCi/ml of water in air.
mean residence times of 9 – 23 days are reported for pines, blueberry, cowberry, birch and spruce. In France [18], during the warm dry season, fast component residence times of the order of 1 – 2 days are observed for apple tree and grape leaves. Citrus fruits and grapes indicate long components of the order of 1 – 4 months, and this suggests the water-retaining capacity of these fruits. The results presented above can be understood in terms of the different evapotranspiration conditions present in the various countries. The general conclusion that stems out of this study is that the mean residence times for the fast components of larger trees are of the order of a few days.

Food grains

Mexico [19], the Philippines [13] and the United States of America [14] have reported tritium turnover rates in food grain plants: maize, wheat and rice. The results tabulated in Table VII show the existence of multiple components. The mean residence times for the first component are found to be about 10 days, the same order as that for other terrestrial plants. The mean residence times for the third component of maize and wheat are representative of tissue-bound tritium.
<table>
<thead>
<tr>
<th>Species</th>
<th>Mean residence times (d)</th>
<th>Remarks</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>I</td>
<td>II</td>
<td>III</td>
</tr>
<tr>
<td>Badam, <em>Terminalia catappa</em></td>
<td>0.3</td>
<td>0.8</td>
<td>12–26</td>
</tr>
<tr>
<td>Mango, <em>Mangifera indica</em></td>
<td>0.6</td>
<td>1.7</td>
<td>5–7</td>
</tr>
<tr>
<td>Sapota, <em>Achras sapota</em></td>
<td>3.2</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Ashok, <em>Saraca indica</em></td>
<td>1.6</td>
<td>—</td>
<td>18</td>
</tr>
<tr>
<td>Norfolk pine, <em>Arctaria bidwilli</em></td>
<td>0.4</td>
<td>1.4</td>
<td>18</td>
</tr>
<tr>
<td>Banana, <em>Musa indica</em></td>
<td>0.4</td>
<td>—</td>
<td>14</td>
</tr>
<tr>
<td>Coconut palm, <em>Cocos nucifera</em></td>
<td>1.4</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Areca palm, <em>Areca catechu</em></td>
<td>1.3</td>
<td>—</td>
<td>14</td>
</tr>
<tr>
<td>Casuarina, <em>Casuarina equisetifolia</em></td>
<td>2.2</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Pine, <em>Pinus sylvestris</em></td>
<td>23.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Cowberry, <em>Vaccinium vitis idaea</em></td>
<td>10.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Blueberry, <em>Vaccinium myrtillus</em></td>
<td>9.0</td>
<td>300</td>
<td>—</td>
</tr>
<tr>
<td>Birch, <em>Betula verrucosa</em></td>
<td>9.0</td>
<td>40</td>
<td>—</td>
</tr>
<tr>
<td>Spruce, <em>Picea excelsa</em></td>
<td>17.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Orange, <em>Citrus sinensis</em></td>
<td>37.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Leaf</td>
<td>—</td>
<td>50</td>
<td>—</td>
</tr>
<tr>
<td>Fruit</td>
<td>—</td>
<td>120</td>
<td>—</td>
</tr>
</tbody>
</table>
### TABLE VI (cont.)

<table>
<thead>
<tr>
<th>Species</th>
<th>Mean residence times (d)</th>
<th>Remarks</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>I</td>
<td>II</td>
<td>III</td>
</tr>
<tr>
<td><em>Grape vine, Vitis vinifera</em></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Leaf</td>
<td>2.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Fruit</td>
<td>12.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Olive, Olea europaea</em></td>
<td>8.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Apple, Pyrus malus</em></td>
<td>1.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Spruce, Picea abies</em></td>
<td>4.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Hornbeam, Carpinus betulus</em></td>
<td>2.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Pine, Pinus sylvestris</em></td>
<td>4.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>6.0</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

### TABLE VII. TRITIUM RESIDENCE TIMES IN FOOD GRAINS USING SPRAY METHOD

<table>
<thead>
<tr>
<th>Species</th>
<th>Mean residence times (d)</th>
<th>Activity sprayed (mCi/m²)</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>I</td>
<td>II</td>
<td>III</td>
</tr>
<tr>
<td><em>Maize, Zea mays</em></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(H-309)</td>
<td>4.5</td>
<td>45</td>
<td>87</td>
</tr>
<tr>
<td><em>Maize, Zea mays</em></td>
<td>5.1</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Maize, Zea mays</em></td>
<td>7.4</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Rice, Oryza sativa</em></td>
<td>13.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td><em>Wheat, Triticum vulgare</em></td>
<td>6.0</td>
<td>31</td>
<td>40</td>
</tr>
</tbody>
</table>

*a Half-life values.*
### Table VIII. TBT Incorporation in Different Plants

<table>
<thead>
<tr>
<th>Species</th>
<th>Component</th>
<th>RSA&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Experimental conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Algae, <em>Chlamydomonas</em> sp.</td>
<td>Whole</td>
<td>0.62</td>
<td>Immersion</td>
</tr>
<tr>
<td>Algae, <em>Scenedesmus obliquus</em></td>
<td>Whole</td>
<td>0.60</td>
<td>Immersion</td>
</tr>
<tr>
<td>Cactus, <em>Opuntia sp.</em></td>
<td>Stem</td>
<td>0.16</td>
<td>Injection</td>
</tr>
<tr>
<td>Cactus, <em>Euphorbia trigona</em></td>
<td>Stem</td>
<td>0.04</td>
<td>Injection</td>
</tr>
<tr>
<td>Cactus, <em>Euphorbia mili</em></td>
<td>Stem</td>
<td>0.03</td>
<td>Injection</td>
</tr>
<tr>
<td>Cactus, <em>Euphorbia mili</em></td>
<td>Leaf</td>
<td>0.13</td>
<td>Injection</td>
</tr>
<tr>
<td>Flowering plant, <em>Tabernaemontana divaricata</em></td>
<td>Leaf</td>
<td>0.04</td>
<td>Irrigation</td>
</tr>
<tr>
<td>Radish, <em>Raphanus sativus</em></td>
<td>Root &amp; stem</td>
<td>0.06</td>
<td>Irrigation</td>
</tr>
<tr>
<td>Medicinal plant, <em>Phyllanthus fraternus</em></td>
<td>Whole</td>
<td>0.10&lt;sup&gt;b&lt;/sup&gt;</td>
<td>Irrigation</td>
</tr>
<tr>
<td>Badam, <em>Terminalia catappa</em></td>
<td>Leaf</td>
<td>0.001</td>
<td>Injection</td>
</tr>
<tr>
<td>Norfolk pine, <em>Araucaria bidwilli</em></td>
<td>Leaf</td>
<td>0.05</td>
<td>Injection</td>
</tr>
<tr>
<td>Mango, <em>Mangifera indica</em></td>
<td>Leaf</td>
<td>0.08</td>
<td>Injection</td>
</tr>
<tr>
<td>Mirchi, <em>Capsicum frutescens</em></td>
<td>Leaf</td>
<td>0.04</td>
<td>Vapour phase</td>
</tr>
<tr>
<td>Lettuce, <em>Lactuca sativa</em></td>
<td>Shoots</td>
<td>0.04</td>
<td>Vapour phase</td>
</tr>
<tr>
<td>Cabbage, <em>Brassica oleracea</em></td>
<td>Shoots</td>
<td>0.04</td>
<td>Vapour phase</td>
</tr>
</tbody>
</table>

<sup>a</sup> RSA, relative specific activity ratio, μCi/ml combustion water to that of μCi/ml TFVT (both peak concentrations).

<sup>b</sup> Four times irrigated.

### Tissue-Bound Tritium

#### Incorporation

The study of tritium turnover in the water compartment indicates the incorporation of a definite fraction in the organic constituents, and this varies in different plant systems. The levels of incorporation of TBT in plants are presented in Table VIII. A higher percentage fixation of TBT (60 – 98%) based
TABLE IX. DISTRIBUTION OF TBT IN DIFFERENT STRUCTURAL PARTS OF FOOD GRAINS

<table>
<thead>
<tr>
<th>Species</th>
<th>Parts</th>
<th>Activity (%)</th>
<th>Activity (pCi/g)</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maize, Zea mays</td>
<td>Endosperm</td>
<td>31.6</td>
<td>47.5</td>
<td>Mexico</td>
</tr>
<tr>
<td></td>
<td>Germ</td>
<td>39.5</td>
<td>59.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Seed coat</td>
<td>28.9</td>
<td>43.4</td>
<td></td>
</tr>
<tr>
<td>Wheat, Triticum vulgare</td>
<td>Flour</td>
<td>28.6</td>
<td>340</td>
<td>Mexico</td>
</tr>
<tr>
<td></td>
<td>Bran</td>
<td>40.3</td>
<td>480</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Germ</td>
<td>31.1</td>
<td>370</td>
<td></td>
</tr>
</tbody>
</table>

on RSA) is observed for aquatic plants such as algae and water grass, in comparison with the terrestrial vegetations (5%). The relatively lower percentage fixations in terrestrial plants is possibly due to the semi-chronic conditions or single exposure used. Higher specific activity ratios could be obtained by maintaining constant specific activity in the soil water [20].

Wheat and maize seeds have shown an even distribution of tritium in the structural parts [5] (Table IX), and 90% of their tritium content has been found to be organic. The remaining 10% (TFWT) is commensurate with the water content of these seeds.

Biochemical distribution

An attempt has been made to study the distribution of the organic bound tritium in the various biochemical constituents. Lipids and nucleic acid fractions show a higher level of tritium incorporation in algae. A similar study conducted on maize [19] indicates 68% fixation in carbohydrates compared with the total organic bound tritium (Table X). These results further indicate nonhomogeneity in the different biochemical constituents and preferential incorporation in fats and proteins. Kahma and co-workers [17] observed a low relative specific activity ratio (0.37) in the nucleic acid fraction of the pea seedlings. Most of the organic bound tritium incorporated in Acetabularia sp. is methanol-extractable and hence is probably the lipid fraction [5].
TABLE X. BIOCHEMICAL DISTRIBUTION OF TBT IN MAIZE – SINGLE EXPOSURE

<table>
<thead>
<tr>
<th>Fraction</th>
<th>Weight (%)</th>
<th>pCi/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry matter</td>
<td>89.5</td>
<td>64.3 ± 13.6</td>
</tr>
<tr>
<td>Tissue water</td>
<td>10.5</td>
<td>7.4 ± 0.9</td>
</tr>
<tr>
<td>Fat</td>
<td>6.1</td>
<td>87.2 ± 3.2</td>
</tr>
<tr>
<td>Proteins</td>
<td>9.3</td>
<td>63.2 ± 10.0</td>
</tr>
<tr>
<td>Carbohydrates</td>
<td>68.3</td>
<td>39.2 ± 10.7</td>
</tr>
<tr>
<td>Fibre</td>
<td>3.5</td>
<td>38.2 ± 8.7</td>
</tr>
<tr>
<td>Residue</td>
<td>2.3</td>
<td>–</td>
</tr>
</tbody>
</table>

MATHEMATICAL MODELS AND APPLICATIONS

Residence times

Tritiated water follows the path of water in general but for its isotopic effect [21], and hence these studies can be used to generate parameters indicative of water movement in the plant system. The occupancy principle of water by Orr and Guillesepie [22] has been adapted by Kline and co-workers [23] and Martin and co-workers [10] for the measurements of transpiration rates, mean residence times and biomass computations of large trees. In the study of uptake and releases of tritium from the different ecological systems, short- and long-term components of tritium releases have been resolved according to the procedure of Koranda and Martin [14] as given by the relation

\[ A_t = \sum_{i=1}^{n} (A_p)_i \exp(-\lambda_i t) \]

where \( A_p \) and \( A_t \) are tritium activity values at the peak time and 't' hours after peak, and \( \lambda (\text{h}^{-1}) \) is the rate constant governing the release process for a particular component. TFWT and TBT fractions are independently fitted into two or three components and the corresponding mean residence values (1/\( \lambda \)) are computed.
Iyengar and co-workers [24] have used an alternate resolution procedure for the measurements of TFWT, labile TBT and bound TBT release rate constants. The equation can be represented as

\[ A_t = A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t) + A_3 \exp(-\lambda_3 t) \]

where \( \lambda_1, \lambda_2 \) and \( \lambda_3 \) are the rate constants governing the release patterns of TFWT, labile TBT and bound TBT respectively, and \( A_1, A_2 \) and \( A_3 \) are the respective coefficients for the three terms. The advantage of this method is in its applicability for field use and all the three components can be resolved by the analyses of wet sample burning.

TFWT time profile

A mathematical model has been developed to predict the TFWT concentration at any time in the activity-time profile of succulent plants [25]. The model assumes that the tritiated water injected into the stem of the plant is subjected to diffusion, longitudinal flow in xylem vessels and biological conversion of TFWT into TBT,

\[ \frac{\partial C}{\partial t} = K \frac{\partial^2 C}{\partial X^2} - U \frac{\partial C}{\partial X} - rC \]

where \( K, U \) and \( r \) are the diffusion coefficient, flow velocity and conversion rate coefficient for TBT synthesis respectively. The diffusion in the water stream is considered as molecular diffusion for succulent plants, and a good agreement between the experimental and calculated values is obtained using diffusion coefficients of the order of \( 1 - 4 \text{ cm}^2/\text{day} \).

The basic assumption in the above model is that the flow rates and transpiration rates are more or less equal. While this may be justifiable in desert succulent plants, the same cannot be said to be true in large trees wherein it appears that there is considerable backflow/reverse flow as indicated by the appearance of multiple maxima and minima in the time activity profiles [10, 16]. The present model needs suitable modifications to represent these wave patterns of the activity profile.

DOSE COMMITMENTS

Data on the mean residence times for TFWT and TBT and their relative concentrations are useful in assessing the dose commitments to man via these food sources. An incorporation of 1 \( \mu \text{Ci}/\text{ml} \) HTO in the body water will give
a dose rate of 0.29 rad/d. Though drinking water forms the major source of tritium entry in man, contribution from other food sources cannot be neglected. Calculations have indicated that the tritium levels in man are as much contributed by the regular intake of vegetables as that of inhalation of air whereas the other two food sources, meat and milk, constitute only about 20% of the total [26]. This represents only the incorporation of tritium in the body water of man from air and water equivalents of food sources excluding the contribution from drinking water. However, the contribution from the vegetable intake will become significantly less in comparison with inhalation in tropical climates where the vapour pressure in air is higher.

**Steady-state concentrations**

The rate of change of concentration of TFWT in vegetables is represented by

\[
\frac{dC}{dt} = k_1 C_s - \lambda_v C_v = 0
\]

where \( C_s \) is the concentration in soil water and \( k_1 \) is the water intake rate per unit volume of the tissue water. Similarly the rate of change of TBT in the same is given by

\[
\frac{dC_{vo}}{dt} = k_1' C_v - \lambda_v C_{vo} = 0
\]

where \( C_v \) is the steady-state concentration of TFWT. The steady-state concentration of TBT is given by

\[
C_{vo} = \frac{k_1' k_1 C_s}{\lambda_v \lambda_v}
\]

Formulations for other food products such as meat and milk can be made similarly.

The rate of change of body water tritium in man consuming water, vegetables, meat, milk and cereals is given by

\[
\frac{dM}{dt} = \sum_{i=1}^{n} I_i C_i + \sum_{i=1}^{n} F_i \alpha_i C_i' + \frac{2BC_a}{W} - \lambda_v M = 0
\]
where \( C_i \) = steady-state concentration of tritium in the intake water systems (drinking water, water equivalents of vegetable, meat, milk, etc.)

\( C_i' \) = steady-state concentration of TBT in the intake food materials as above

\( I_i \) = intake rate of water equivalents from various sources per unit volume of body water

\( F_i \) = fraction of the particular organic constituents in the intake food

\( \alpha_i \) = metabolism coefficients of TBT into TFWT for different systems

\( B \) = inhalation rate of air

\( C_a \) = air concentration of tritium

\( W \) = body water content of man

\( \lambda_e \) = effective elimination rate coefficient for TFWT

\( M \) = body water concentration of tritium in man

The metabolism coefficients for organics can be obtained from the values of third component release rates.

The rate of change of TBT in man can be similarly represented by

\[
\frac{dM_0}{dt} = \sum_{i=1}^{n} F_i \ r_i \ C'_i + \beta \sum_{i=1}^{n} I_i \ C_i - \lambda_0 \ M_0 = 0
\]

where \( I_i \) and \( C_i / C'_i \) have the same significance as noted above, and \( \beta \) is the conversion rate coefficient of water into organics. \( r_i \) is the coefficient of utilization for organic synthesis from the particular constituent and \( \lambda_0 \) is the elimination rate coefficient for organic bound tritium in man.

The contribution of TBT concentrations will undoubtedly be less significant compared with the body water tritium levels in man because of its small concentration and the absence of specific enrichment in any biochemical constituents. However, the significant differences in the mean residence times between body water tritium and organic bound tritium might make the effect of the latter long lasting. Further the fixations of this component in the body of man or in any localized organs, its metabolism and subsequent release patterns are not yet properly understood.

ACKNOWLEDGEMENT

The authors are grateful to the International Atomic Energy Agency for providing financial assistance to carry out this work.
REFERENCES


ENVIRONMENTAL TRITIUM IN TREES

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Canada

Abstract

ENVIRONMENTAL TRITIUM IN TREES.

The distribution of environmental tritium in the free water and organically bound hydrogen of trees growing in the vicinity of the Chalk River Nuclear Laboratories (CRNL) has been studied. The regional dispersal of HTO in the atmosphere has been observed by surveying the tritium content of leaf moisture. Measurement of the distribution of organically bound tritium in the wood of tree ring sequences has given information on past concentrations of HTO taken up by trees growing in the CRNL Liquid Waste Disposal Area. For samples at background environmental levels, cellulose separation and analysis was done. The pattern of bomb tritium in precipitation of 1955–68 was observed to be preserved in the organically bound tritium of a tree ring sequence. Reactor tritium was discernible in a tree growing at a distance of 10 km from CRNL. These techniques provide convenient means of monitoring dispersal of HTO from nuclear facilities.

Trees present a convenient means of monitoring the dispersal of tritiated water in an area. Current levels of HTO in atmospheric and soil moisture are reflected in the tritium content of the leaf moisture. A dated record of the past concentrations of tritium in an area is preserved in the organically bound tritium of individual annual growth rings of the local trees.

Several workers have measured the tritium content of pine needles to study the dispersal of tritium in the environment. Stewart, Wyerman, Sherman and Schneider [1] found the tritium content of moisture extracted from pine needles collected at various sites across the United States correlated with the geographical distribution of tritium in precipitation resulting from thermonuclear tests or proximity to nuclear industrial facilities. Murphy and Corey [2] studied the uptake of HTO vapour by pine needles of branches enclosed in plastic bags. The appearance of HTO in pine needles, as well as other vegetation and soil, following passage of a cloud of molecular HT has been reported by Murphy, Watts and Corey [3].

Little work has been reported on the tritium content of individual tree rings. Kigoshi and Tomikura [4] measured the
concentration of tritium in dried wood of 1953–1960 rings and found a distribution corresponding to the known releases of thermonuclear tritium during the period.

We have been studying tritium distribution in tree components in the vicinity of Chalk River Nuclear Laboratories (CRNL). The CRNL Liquid Waste Disposal Area provides a useful site for studying tritium dispersal in the natural environment. In some areas, surface and ground waters have an elevated tritium content as the result of the controlled release of reactor-produced tritium. These concentrations are not high enough to constitute a health hazard, but provide convenient spikes which may be measured more easily than the tritium in the general environment. Studies have been extended to off-site locations to demonstrate potential applications of the techniques in the general environment.

EXPERIMENTAL PROCEDURES

This work has involved a wide range of tritium activity levels. To avoid contamination problems, low-level samples (<5000 TU; originally, <2000 TU more recently) have been analyzed in a separate low-level laboratory by personnel not involved with high-level samples.

Water Extraction

Water was recovered from soil, leaves, wood chips and cellulose by azeotropic distillation with toluene using a Dean-Stark receiver.

Ring Separation

(i) To obtain water from specific rings, separation was done manually with a sharp knife or chisel as soon as possible after the tree was cut.

(ii) When there was no requirement for isolation of free water from the wood, and cellulose separation was to be done, separation was greatly facilitated by extraction of an intact sector (2 cm thick) with refluxing monoethanolamine vapour for about three hours. This treatment softened the wood at ring boundaries making possible clean separations with a spatula blade.

\[1\text{ Tritium Unit (TU)} = 1 \text{ T atom per } 10^{18} \text{ H atoms}\]
\[= 0.12 \text{ Bq per litre water}\]
\[= 3.25 \text{ pCi per litre water.}\]
### Table 1. HTO Distribution in Maple Trees in Perch Lake Basin, CRNL (Concentration in Tritium Units)

<table>
<thead>
<tr>
<th>Free water from</th>
<th>Tree 1</th>
<th>Tree 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmosphere</td>
<td>~5000</td>
<td></td>
</tr>
<tr>
<td>Leaves</td>
<td>3700 - 4300</td>
<td>9200</td>
</tr>
<tr>
<td>Leaf stalks</td>
<td>1400 - 1500</td>
<td></td>
</tr>
<tr>
<td>Twig wood</td>
<td>1200 - 1350</td>
<td></td>
</tr>
<tr>
<td>Trunk wood</td>
<td>1200</td>
<td></td>
</tr>
<tr>
<td>Trunk bark</td>
<td>1175</td>
<td></td>
</tr>
<tr>
<td>Root wood</td>
<td>1400</td>
<td></td>
</tr>
<tr>
<td>Soil</td>
<td>770</td>
<td>75200</td>
</tr>
<tr>
<td>Ground water</td>
<td>750</td>
<td></td>
</tr>
</tbody>
</table>

**Tree 1.** Located on the east shore of Perch Lake where the atmospheric HTO level is raised by influx of reactor HTO evaporated from the lake, but the soil and ground waters are relatively low in tritium.

**Tree 2.** Located in a drainage path from the Liquid Waste Disposal Pit where the concentration of atmospheric HTO is low compared to the concentrations of soil and ground water HTO.

### Cellulose Separation

For low activity samples, 40 g wood chips were boiled in two litres monoethanolamine with vigorous stirring for about eight hours (Wise [5]). The pulp was filtered hot and washed free of solvent with hot water. Coniferous woods required additional treatment with hypochlorous acid and sodium sulphite to complete removal of lignin.

### Hydroxyl Equilibration

The cellulose pulp was boiled one hour in 0.4N HCl solution prepared with tritium-free water (<2 TU). The pulp was washed free of acid with tritium-free water and stored wet until combusted. This treatment was repeated just before drying for combustion.

### Drying

(i) Ground or chipped wood was dried overnight in an air oven at 130°C prior to combustion.

(ii) To avoid exchange with atmospheric HTO during and after drying, low activity cellulose was sealed in the combustion apparatus while still damp with tritium-free water. It was dried overnight by heating to 170°C in vacuo, the water being collected in liquid nitrogen cooled traps.
FIG. 1. HTO in free water of poplar leaves in the vicinity of Chalk River Nuclear Laboratories and Ontario Hydro NPD power station, July 1978.
Combustion

Wood or cellulose samples were burned completely in a flow of dried oxygen, and nitrogen and the water collected in a trap cooled to -80°C.

Tritium Measurement

Samples were measured by liquid scintillation counting or gas counting, in some cases after isotopic enrichment by electrolysis.

HTO IN THE FREE WATER OF TREES

Pickup of Atmospheric HTO by a Tree

The concentrations of HTO in various parts of two trees growing in the CRNL Liquid Waste Disposal Area are shown in Table I. Tree 1 was located on the east shore of a small lake where the soil and ground waters are relatively uncontaminated but the atmospheric HTO level is elevated due to the influx of evaporate from the lake where the tritium concentration is 40,000 TU. It is seen that the HTO concentration in the leaves of the tree is much closer to that of the atmosphere than to that of the soil and ground waters which contribute the transpirational stream of the tree. Evidently the leaf moisture equilibrates with the surrounding atmospheric moisture. HTO diffuses back into the tree and the water of the woody parts of the tree appears surprisingly well-mixed at a concentration intermediate between those of the soil and atmospheric moisture.

Tree 2 was growing in a drainage path from the Disposal Pit where the soil and ground waters HTO concentration was high compared to the HTO content of the atmospheric moisture. In this reverse situation, the leaf water HTO was found to be much lower than the soil water as a result of exchange loss to the atmosphere.

This evidence that the HTO content of tree leaf moisture is strongly affected by the HTO content of the surrounding atmospheric moisture suggests the use of tree leaves as a sampling device for monitoring of atmospheric HTO in the general environment.

Regional Pattern of Leaf HTO Around an Industrial Source

As a result of reactor operations, HTO vapour is released from CRNL and the nearby Nuclear Power Demonstration (NPD) plant. A survey of the HTO content of poplar (Populus L.) leaves has delineated the overlapping plumes resulting
from these releases as shown in Figure 1. Concentration contours are similar in geographic pattern and level of activity to those observed for HTO in snow cores (Brown and Fraser [6]). Reactor tritium is discernible above a background level of 150 TU (natural and bomb-test tritium) over an area 70 km long by 20 km wide oriented along the Ottawa River similar to the local wind pattern. The maximum concentration at the NPD site boundary was about 5000 TU, at the boundary of the larger CRNL site about 1000 TU. These concentrations were established by a release rate of about 7200 curies per year (270 TBq a⁻¹) from CRNL and roughly twice that from NPD. A concentration of 5000 TU in atmospheric moisture is 0.1% of MPCₐ for general population.

It should be pointed out that operations at these two stations are experimental in nature and releases per unit power at full-scale commercial power stations are much lower. Furthermore, methods are under development for the removal of tritium from reactor fluids to limit the exposure of station personnel as well as the general population.

**HTO Content of a Sequence of Tree Rings**

Only the outer few rings of a tree conduct its transpirational stream. Resins and lipids are deposited in the conducting passages of older rings which become heartwood and no longer take part in the active life of the tree. It is of interest to see how isolated the inner wood is in terms of water exchange.

The distribution of HTO in a sequence of growth rings of a tree grown in the CRNL Liquid Waste Disposal Area is shown in Figure 2. The rings of 1971 to 1977 show a uniform HTO concentration. Evidently these are the active rings conducting present-day water up the tree. In earlier rings the HTO concentrations are higher indicating that water of higher tritium content came into the tree at some time in the past. Resistance to water exchange is evident since there has not been equilibration with the water of the outer rings. However, there certainly is significant diffusion of HTO into older rings since tritium is observed right to the removal of the tree, ring year 1938, in spite of the fact that there was no reactor tritium in this area prior to 1948 and very little prior to 1958.

**ORGANICALLY BOUND TRITIUM IN TREE RINGS**

**Disposal Area Trees**

The concentration of tritium found in the combustion water of dried wood of the tree discussed in the previous section is also shown in Figure 2. It is of note that the wood of pre-1958 rings has less than 5% of the tritium content of
water permeating that wood. In another tree in which the concentrations were 100 times higher, this value was about 2%. This is rather surprising in view of the reported exchangeability of the hydroxyl hydrogen of cellulose (Lang and Mason [7], Sepall and Mason [8]). Presumably it indicates steric protection of these groups in the intact wood structure. In any case, it means that a reasonable estimate of the original tritium content of a tree ring can be obtained simply from the dried wood without serious interference by exchange of hydroxyl hydrogens or deposition of non-cellulosic materials after the original ring formation. This greatly facilitates the use of tree ring studies to learn the history of tritium dispersal within the Disposal Area.

The results from the analysis of several trees from the Disposal Area are shown in Figure 3 together with the concentration of tritium for recent years in the small stream draining the area. Concentrations are adjusted for radioactive decay back to the year of ring growth. The initial tree was cut and analyzed in 1964. It grew in the middle of the stream and had a tritium concentration in its outer ring similar to the
FIG. 3. Tritium concentrations in:
- Small stream draining area near the Liquid Waste Disposal Pit, CRNL; mean May to August values.
- ◦ Combustion water of dried wood from white spruce, cut 1964.
- ▲ Combustion water of dried wood from white spruce, cut 1974.
- + Combustion water of dried wood from balsam fir, cut 1978.

stream water. Subsequent concentrations in the stream look like a continuation of the pattern observed in the tree. The other trees were attempts to duplicate the original results at later dates and demonstrate preservation of the 1962 peak in trees that continued growing. The differences in concentration levels are attributed to siting differences. The area is close to the Disposal Pit and the appearance of tritium in specific trees is very dependent on the microhydrology of the area which is probably complex with narrow and variable subsurface drainage paths influenced by discharges to the Pit.
FIG. 4. Comparison of the distribution of tritium in tree ring cellulose from trees grown at Ottawa and near Chalk River Nuclear Laboratories with summer and winter mean concentrations in Ottawa precipitation.

In all trees, the pre-1957 values reflect the contamination level inherent in the whole dried wood samples. They are higher in Figure 3 than in Figure 2 because of the decay adjustment in the former. Pre-1955 cellulose separated from the 1964 tree and hydroxyl-equilibrated with tritium-free water in the low-level laboratory gave a value of 800 TU, <0.1% of the concentration of the water permeating this wood when it was cut. Thus, it is certain that there was no reactor tritium in the waters feeding these trees prior to 1957.

Concentration started to rise in the 1964 tree in 1958, one year ahead of the other trees. The 1974 tree seems to be one year out of phase relative to the other two trees, suggesting an error in ring year assignment. If this is the case, the 1962 peak is common to all three and the 1974 and 1978 trees have coincident peaks in 1959 and 1969 as well.

The patterns observed are consistent with the known history of tritium release in this area. NRU reactor, the chief source of tritium for liquid disposal, started operation in late 1957. It was plagued with heavy water leaks in its early years but these were remedied by 1962. Heavy water management was steadily improved through the mid-1960’s. The reactor was out of operation 1972-74 for vessel change and as shown by the recorded stream concentrations and the 1978 tree, tritium levels in the area dropped accordingly. The cause of the 1969 peak in the 1978 tree is unknown but it does reflect a rise in the stream concentration for 1968-69.
<table>
<thead>
<tr>
<th>Year</th>
<th>Concentration as of June 1969</th>
<th>Adjusted for Blank and OH</th>
<th>Concentration at Year of Growth</th>
<th>Concentration in Precipitation Average April-September</th>
<th>Tree/Precipitation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1968</td>
<td>201</td>
<td>230</td>
<td>243 ± 17</td>
<td>262</td>
<td>0.93</td>
</tr>
<tr>
<td>1967</td>
<td>272</td>
<td>331</td>
<td>371 ± 26</td>
<td>410</td>
<td>0.90</td>
</tr>
<tr>
<td>1966</td>
<td>438</td>
<td>568</td>
<td>672 ± 38</td>
<td>903</td>
<td>0.74</td>
</tr>
<tr>
<td>1965</td>
<td>605</td>
<td>807</td>
<td>1010 ± 54</td>
<td>1070</td>
<td>0.94</td>
</tr>
<tr>
<td>1964</td>
<td>1126</td>
<td>1551</td>
<td>2057 ± 106</td>
<td>1886</td>
<td>1.09</td>
</tr>
<tr>
<td>1963</td>
<td>2213</td>
<td>3104</td>
<td>4353 ± 316</td>
<td>4107</td>
<td>1.06</td>
</tr>
<tr>
<td>1962</td>
<td>516</td>
<td>680</td>
<td>1010 ± 55</td>
<td>1080</td>
<td>0.94</td>
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<tr>
<td>1961</td>
<td>139</td>
<td>141</td>
<td>222 ± 20</td>
<td>204</td>
<td>1.09</td>
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<tr>
<td>1960</td>
<td>156</td>
<td>165</td>
<td>275 ± 22</td>
<td>198</td>
<td>1.39</td>
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<tr>
<td>1959</td>
<td>315</td>
<td>393</td>
<td>692 ± 42</td>
<td>550</td>
<td>1.26</td>
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<tr>
<td>1958</td>
<td>246</td>
<td>291</td>
<td>542 ± 37</td>
<td>730</td>
<td>0.74</td>
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<tr>
<td>1957</td>
<td>133</td>
<td>133</td>
<td>262 ± 26</td>
<td>142</td>
<td>1.85</td>
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<tr>
<td>1956</td>
<td>92</td>
<td>74</td>
<td>154 ± 21</td>
<td>222</td>
<td>0.69</td>
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<tr>
<td>1955</td>
<td>47</td>
<td>10</td>
<td>22 ± 20</td>
<td>59</td>
<td>0.37</td>
</tr>
<tr>
<td>1954</td>
<td>90</td>
<td>71</td>
<td>166 ± 23</td>
<td>393</td>
<td>0.42</td>
</tr>
<tr>
<td>1953</td>
<td>35</td>
<td>-</td>
<td>-</td>
<td>20</td>
<td>-</td>
</tr>
<tr>
<td>1949-52</td>
<td>44</td>
<td>-</td>
<td>-</td>
<td>20</td>
<td>-</td>
</tr>
</tbody>
</table>

*±5% analytical uncertainty

b Adjusting for blank (40 ± 5 TU) and 30% loss of original tritium by hydroxyl group exchange:

\[
\text{Adjusted value} = \frac{\text{Measured concentration (±5%) - 40 (±5) TU}}{0.70}
\]
Off-Site Trees

On extending tree ring analysis for tritium to the level of bomb-test tritium in the general environment it was found advisable to prepare separated cellulose to reduce contamination problems. This eliminates material deposited subsequent to ring formation and provides a material of definite chemical composition from which all hydroxyl tritium can be eliminated in case it has been incorporated by exchange subsequent to ring formation. Various treatments were tried to achieve complete exchange of the hydroxyl groups of cellulose with water in a manner that would be practical with tritium-free reagents. Finally this was accomplished by boiling cellulose for one hour in 0.4N HCl prepared with tritium-free water (<2 TU). Experiments using tracer levels of tritium (10^5 TU) showed 28 to 32% exchange of cellulose hydrogens. After replacement of the original hydroxyl hydrogen with tritium-free hydrogen, the cellulose has 70% of its original tritium content and its measured T/H ratio needs to be corrected by the factor 10/7 to obtain the original tritium concentration.

This hydrolytic treatment did not eliminate contamination problems completely. No source of contamination could be identified but a consistent background level of about 50 TU was obtained on old cellulose. While this precluded any measurement of the pre-1953 tritium level, significant measurement of the distribution of thermonuclear tritium in post-1953 rings was possible. Values obtained for pre-1953 rings were accepted as a blank to be subtracted from all values for that tree.

In Figure 4, results obtained on poplar trees grown at well-drained sites at Ottawa and in the Chalk River vicinity are compared with the record of tritium concentration in Ottawa precipitation (Brown [9]). Data for the Ottawa tree is presented in Table II to show the levels of activity measured and the magnitude of the adjustments for blank, loss of hydroxyl tritium and radioactive decay. Concentrations observed in the Ottawa tree show good correlation with the mean concentrations of tritium in precipitation over the summer growth interval of each year back to 1960.

Reactor tritium is clearly evident in the rings of the trees growing 5 and 10 km northwest of the CRLN stack. Concentrations in the pre-1960 rings of the tree at 10 km correlate well with the record of thermonuclear tritium at Ottawa. This demonstrates the potential utility of such measurements for establishing environment levels before and after installation of a nuclear power plant.

CONCLUSIONS

Analysis of the tritium content of tree leaf moisture has been found to provide a convenient means of observing regional atmospheric dispersal of HTO from an industrial source.
Concentrations established in such vegetation by a given release are particularly relevant to population exposure considerations since they combine contributions from atmospheric moisture, precipitation and soil water averaged over a few weeks time.

Tree ring analysis has proved to be a practical means of investigating the history of tritium release in an area. Although recent water penetrates older rings, retention by the older wood components of tritium in the permeating water is <5%. When a contamination level of this order is acceptable, the tree ring tritium distribution can be obtained using dried whole wood samples. If HTO levels in the tree water of the wood are much higher than the organically bound tritium levels, e.g., in old wood, separated cellulose samples must be used. Purified cellulose containing <0.1% contribution from the water that permeated it in the tree can be prepared. This demonstrates that the original isotopic composition of the carbon-bonded hydrogen as laid down in ring formation is preserved in the cellulose and can be determined.

In general, the pattern of tritium concentrations in tree ring sequences has correlated with the known history of tritium dispersal at the elevated levels of the CRNL Liquid Waste Disposal Area and at the level of bomb-test tritium in the general environment. However, it must be borne in mind that a tree ring tritium record simply reflects past concentrations in a specific tree's water supply and must be interpreted in light of knowledge of the origin of that water.

ACKNOWLEDGEMENTS

Much of this work was done by a succession of university students during their summer employment periods. These include the former Misses M.L. Church and A.R. Robertson, Eric Lloyd and Joan Yaraslavitch. Mrs. E. Robertson performed the cellulose analyses on off-site trees in the low-level laboratory. The leaf sampling and analysis was done by C.D. Fraser.

The author acknowledges with gratitude the able technical assistance of all the foregoing in carefully performing the numerous analyses involved.

REFERENCES


DISCUSSION

A.S. MASON: Was the source of HTO to which the trees were exposed at the Chalk River site evaporation or direct input from the effluent pond?

R.M. BROWN: In the study of tree ring tritium in trees growing in the Liquid Waste Disposal Area, the source of tritium was the surface and groundwater draining from the Disposal Pit. In the other studies, involving the leaf-moisture HTO distribution pattern and tree ring tritium in off-site trees, the source of tritium was atmospheric HTO vapour of bomb-test and local reactor origin.

P. BOVARD: Has the relative humidity of the air been fairly constant over your sampling period? Also, does the relative humidity have any effect on the ratio between the tritium in the atmosphere and in the leaf?

R.M. BROWN: The humidity was relatively constant during our sampling operations, and so we did not see effects that we would attribute to humidity fluctuations. The finite time for equilibration of leaf HTO and atmospheric HTO would serve to smooth out such effects. We have not studied the effect of air humidity on leaf HTO specifically.

L.A. KÖNIG: In the forests surrounding the Karlsruhe Nuclear Research Centre we measured the tritium concentration simultaneously in air and soil humidity as well as in leaves and pine needles. The tritium concentration in the air humidity was found to have a marked effect on that of the leaves and needles. However, rapid changes in the tritium concentration in the air humidity were observed.

W. ROETHER: Some years ago we made a study of tritium concentrations in wine and we looked into the relationship in tritium concentration between air humidity and water in, e.g. the leaves. I think it is quite obvious that there
has to be a dependence on humidity. In a case where the air humidity is 100% no water will be sucked up the stem, but the stomata of the leaves will be open and the water in the leaves will approach the tritium concentration of the air moisture, whereas at 0% humidity the leaves will only contain water drawn up from the soil. Therefore, the higher the humidity, the more will the tritium concentration in the leaves represent that of the atmospheric moisture.
TRANSFER OF TRITIUM RELEASED BY NUCLEAR FACILITIES TO THE FOOD SUPPLY.

The use for agricultural purposes of river waters receiving releases or discharges of tritium results in contamination of irrigated crops and of animals given such water to drink or consuming the contaminated crops. It therefore seemed of importance to assess the part played by tritium in the contamination of the food chain, together with its possible effects on organisms. With this in mind, French, Belgian and Netherlands laboratories have joined forces to study, more especially, the relationship between environmental contamination rates and those of produce harvested in the Mediterranean region and in a humid temperate climate, the transfer process in the chain: water – fodder – bovines – dairy produce, and the role of technology in the contamination of the food chain. The present status of research undertaken jointly by organizations in the three countries is reviewed. In the Atlantic environment the experiments involved four annual crops consumed on a large scale: potatoes, sugar beet, carrots and peas, and in the Mediterranean environment several perennial species such as vine, olive, orange and apple were studied. The results obtained relate to the residence time for

* Les travaux présentés dans ce rapport ont été effectués dans le cadre du Programme coordonné de recherches de l’Agence internationale de l’énergie atomique sur le tritium et l’environnement, ainsi que dans le cadre d’un contrat avec la Commission des Communautés européennes.
tritium in the various organs of each species, the part played by evapotranspiration and the physiological functions of the different parts of the plants, the uptake of tritium by tissue water and organic matter, and the distribution of tritium in the soil profile.

TRANSFERT A LA RATION ALIMENTAIRE DU TRITIUM EMIS PAR LES INSTALLATIONS NUCLEAIRES.

L'utilisation à des fins agricoles des eaux de rivières réceptrices des rejets ou des dépôts de tritium entraîne la contamination des végétaux irrigués et celle des animaux abreuvis avec cette eau ou consommant les végétaux contaminés. Il paraît donc important d'évaluer le rôle du tritium dans la contamination de la chaîne alimentaire ainsi que ses effets éventuels sur les organismes. C'est pourquoi des laboratoires français, belges et néerlandais se sont associés pour étudier, en particulier, les relations entre les taux de contamination du milieu et ceux des produits récoltés en région méditerranéenne et en climat tempéré humide, le transfert dans la chaîne eau — fourrage — bovins — produits laitiers, et le rôle de la technologie dans la contamination de la chaîne alimentaire. Le mémoire fait le point de l'état actuel des recherches entreprises en commun par les organismes des trois pays. En milieu atlantique, les expériences ont porté sur quatre cultures annuelles de grande consommation: pomme de terre, betterave sucrière, carotte et pois, et en milieu méditerranéen sur plusieurs espèces pérennes telles que la vigne, l’olivier, l'oranger et le pommier. Les résultats obtenus concernent: le temps de résidence du tritium dans les divers organes de chacun des espèces, le rôle de l'évapotranspiration et des fonctions physiologiques des différentes parties des plantes, l'incorporation du tritium dans l'eau tissulaire et la matière organique, et la distribution du tritium dans le profil du sol.

1. INTRODUCTION

Le tritium, dont la caractéristique essentielle est d'être un radioisotope de l'hydrogène, constituant majeur de la matière vivante, est actuellement l'un des radionucléides dont les rejets dans l'environnement sont les plus importants. Le tritium produit dans les réacteurs nucléaires par plusieurs mécanismes demeure pour sa majeure partie dans le combustible; toutefois, une certaine partie évaluée à environ 2 curies par MW(e) s'échappe dans l'environnement, soit par la voie aérienne (10 à 25%), soit avec les effluents liquides sous forme de TMO (75 à 90%). Le tritium confiné dans les éléments combustibles est libéré au cours de leur retraitement (690 Ci par tonne de combustible). Là aussi le tritium est relâché en partie dans l'air et en partie dans l'eau. L'ampleur de ces décharges est due en particulier au fait que les procédés actuels d'épuration des effluents ne sont pas susceptibles de retenir des quantités appréciables de ce radionucléide. Le problème du tritium dépasse le cadre de la fissio puisqu'il se retrouvera également dans les installations qui mettront en œuvre l'énergie provenant de la fusion.
Le comportement dans l'environnement du tritium, qui est un corps éminemment diffusible, revêt donc, à long terme, une importance primordiale pour la radioprotection.

A plus longue échéance, le tritium moléculaire gazeux se transforme en THO par oxydation; aussi est-il intéressant de connaître les mécanismes de cette transformation et, en particulier, sa vitesse ainsi que les facteurs favorisant ou défavorisant cette oxydation. Certains auteurs ont mis en évidence le rôle du sol et de la végétation dans cette transformation.

D'autres auteurs se préoccupent du transfert dans le milieu de cette eau tritée, soit sous forme liquide, soit sous forme de vapeur. La diffusion de la vapeur d'eau tritée s'effectue non seulement dans l'atmosphère, mais également dans le sol, et les échanges atmosphère-sol font l'objet de recherches approfondies.

Le tritium ainsi dispersé entrera dans les écosystèmes où il aura trois étapes principales à franchir:

- le substratum que constitue le sol
- les échanges sol-plante et atmosphère-plante
- le maillon animal herbivore et carnivore auquel appartient l'homme [1].

Sur le plan sanitaire, l'utilisation à des fins agricoles ou domestiques d'eaux de rivière contenant du tritium sera la voie d'atteinte de l'homme, l'irradiation externe pouvant être négligée du fait de la faible énergie de cet émetteur [2].

Nous ne parlerons pas ici de la consommation de l'eau de boisson, voie directe qui ne pose pas de problème de discrimination, à l'inverse de l'utilisation agricole qui fait intervenir, comme intermédiaires, végétaux et animaux.

L'implantation de l'industrie nucléaire envisagée pour les prochaines années dans le bassin de grands fleuves tels que la Meuse, le Rhin ou le Rhône entraînera un niveau croissant de tritium (jusqu'à 10 000 pCi/l) dans l'eau des fleuves d'abord et dans celle de la nappe ensuite.

Or, dans certains pays comme la Belgique et les Pays-Bas, environ 70% de l'eau d'abreuvement des vaches laitières provient directement ou indirectement des fleuves qui les traversent [3].

Un apport de tritium est donc possible par cette voie, auquel il faut ajouter l'apport sous forme de matière organique par l'intermédiaire des plantes fourragères: herbe au moment du pâturage, mais et foin pendant la période de stabulation. Au cours de la période estivale, qui est celle de la croissance des plantes fourragères, l'appel d'eau du fleuve vers la nappe puis vers la plante sera plus grand, et il est important de savoir si ce flux se traduit par une augmentation de la teneur en tritium de la matière organique des plantes fourragères. Il en est de même pour l'irrigation des pâturages par aspersion, technique qui se pratique de plus en plus, même dans les pays au climat tempéré où l'élevage intensif s'impose pour des raisons économiques [4].

Ces flux tritiés tendront vers l'établissement d'un équilibre dans l'organisme animal et les produits synthétisés par lui comme le lait.
Le lait n'est pas la seule source de tritium dans l'alimentation humaine, d'autres cultures vivrières sont irriguées aussi bien en climat tempéré humide qu'en milieu méditerranéen.

Enfin, les transformations que la technologie alimentaire fait subir aux produits récoltés peuvent également influencer la teneur de l'aliment effectivement consommé, comme cela a déjà été mis en évidence pour d'autres radionucléides [5].

2. ETUDE DU CYCLE DU TRITIUM EN FONCTION DU CLIMAT

Le cycle de l'eau est largement influencé par les conditions climatiques, aussi nous a-t-il paru intéressant de comparer l'action du climat atlantique tempéré et humide et celle du climat méditerranéen chaud et sec sur le flux du tritium dans la plante et sur son comportement aux différents stades du métabolisme.

Photosynthèse et évapotranspiration par exemple ont-elles une influence sur le flux du tritium dans la plante?

Pour mener à bien cette étude, nous avons mis en commun, dans le cadre d'un contrat patronné par la Commission des Communautés européennes, les moyens des laboratoires de Cadarache et San Giuliano (France), de Mol (Belgique) et de Wageningen (Pays-Bas). Ces travaux entrent également dans le cadre du Programme coordonné de recherches de l'AIEA.

Cette expérimentation menée en conditions naturelles a pour but:
- de compléter les données acquises dans les domaines de la diffusion de l'eau dans le sol et des besoins en eau de diverses cultures dans les deux régions considérées,
- de vérifier jusqu'à quel point le comportement du tritium est analogue à celui de l'hydrogène,
- d'évaluer le rôle du tritium dans la contamination de la chaîne alimentaire ainsi que ses effets éventuels sur les organismes,
- de déterminer les relations entre les taux de contamination du milieu et ceux des produits récoltés,
- d'estimer le rôle de la technologie dans la contamination de la ration alimentaire (vinification, préparation des jus de fruits, sucrerie).

Les expérimentations ont porté en milieu atlantique sur quatre cultures annuelles de grande consommation: la pomme de terre, la betterave sucrière, la carotte et le pois, et en milieu méditerranéen sur trois espèces pérennes: la vigne, l'olivier et l'oranger.

Ces trois dernières cultures ont été choisies non seulement pour leur importance économique mais également pour leur adaptation au climat.
Certains caractères de l’olivier et de l’oranger, feuilles persistantes, et fréquence des aspersions pour l’oranger, pouvaient contribuer a priori à augmenter le risque de contamination des produits récoltés.

Accessoirement des injections de tritium ont été pratiquées chez le pommier pour corroborer certaines observations faites sur la vigne.

Dans tous les cas, les expérimentations ont été menées en respectant les pratiques agricoles courantes de plantation, fumure et entretien.

2.1. Expérimentation en milieu atlantique tempéré

Les expériences ont été effectuées à la ferme expérimentale du Département de radiobiologie du Centre d’étude de l’énergie nucléaire de Mol [6].

Au total, 7600 litres d’eau tritée (270 µCi/l) ont été déposés le même jour sur deux champs circulaires (Ø 24 m) divisés chacun en trois anneaux concentriques (Z₁, Z₂, Z₃). Des échantillons de sol et de végétaux ont été récoltés à des intervalles déterminés depuis le moment du dépôt de l’eau tritée jusqu’à la récolte finale, afin de déterminer l’incorporation du tritium dans les fractions de l’eau tissulaire et de la matière organique ainsi que la distribution de l’eau tritée dans le profil du sol.

Le dépôt d’eau tritée a eu lieu respectivement 94 jours avant la récolte des betteraves et des carottes, 24 à 39 jours avant celle des pois et 39 à 46 jours avant celle des pommes de terre.

Les résultats obtenus pour l’ensemble des trois zones contaminées sont les suivants:

a) La concentration moyenne en tritium mesurée dans la partie comestible des plantes cultivées était respectivement (pCi ³H par gramme de matière fraîche) pour un dépôt d’eau tritée normalisé à 1 mCi ³H/m²: 202 dans la racine de betterave, 87 dans la racine de carotte, 1676 dans le tubercule de pomme de terre.

b) Le pourcentage final (c’est-à-dire après récolte) de distribution de l’activité en tritium dans l’eau libre tissulaire (TFWT) et dans la matière organique (OBT) de ces végétaux indique que dans la fraction TFWT la distribution était: 39,3% dans le grain de pois, 87,8% dans la racine de betterave, 88,2% dans le tubercule de pomme de terre et 90,2% dans la racine de carotte. Inversement, dans la fraction OBT, la distribution était respectivement: 9,8 dans la racine de carotte, 11,8% dans le tubercule de pomme de terre, 12,2% dans la racine de betterave et 60,7% dans le grain de pois.

c) Les valeurs du rapport des activités spécifiques

  pCi ³H/g H dans OBT
  pCi ³H/g H dans TFWT
sont respectivement: 2,95 dans le grain de pois, 1,00 dans la racine de betterave, 1,96 dans la racine de carotte et 1,00 dans le tubercule de pomme de terre. Ces résultats montrent que chez les espèces végétales étudiées, le tritium incorporé dans la fraction organique a un temps de résidence au moins aussi long que le tritium contenu dans la fraction d’eau tissulaire.

d) L’échantillonnage du profil du sol a été effectué à différents moments après le dépôt. Les résultats montrent que la proportion (en μCi) du dépôt retenue, le premier jour, par le sol est de 73% pour l’ensemble des parcelles cultivées de la zone 2 dont 67,5% dans la couche superficielle du sol (0–10 cm), et de 93% dans le cas du sol nu dont 86,9% dans la couche superficielle. Pour l’ensemble des parcelles cultivées, 41% du dépôt initial était encore présent dans le sol 39 jours plus tard dont 0,3% seulement dans la couche superficielle. Le front de la contamination a atteint la nappe aquifère (profondeur 80 cm) entre le deuxième et le troisième mois après le dépôt; une activité maximale de 6 μCi/l a été observée dans l’eau de la nappe environ 4 mois après le dépôt; à noter que durant cette période environ 240 mm de pluie ont été enregistrés.

e) Une bonne corrélation a été observée entre, d’une part, les teneurs en tritium de l’eau du sol superficiel (0–10 cm) et, d’autre part, les teneurs en tritium de l’eau tissulaire de la partie aérienne des pomme de terre, pois, betterave et carotte. De même une excellente corrélation ($r^2 = 0,99$) a été constatée entre la teneur en tritium de l’eau de la racine de carotte et celle de la couche superficielle du sol.

Par contre, aucune relation significative n’a été trouvée entre les teneurs en tritium de l’eau des tubercules de pomme de terre, des racines de betterave et des grains de pois d’une part, et celles de l’eau du sol (couches 0–10 cm et 10–80 cm).

2.2. Expérimentation en région méditerranéenne

Par rapport au climat océanique, le climat méditerranéen se caractérise par les sécheresses estivales et la grande luminosité du ciel. La durée de l’insolation est de 2600 heures. La température moyenne s’élève au-dessus de 25°C au cours d’une période approximative de 90 jours. En hiver, de brusques abaissements de température correspondent souvent à un vent froid et violent. Les pluies relativement abondantes, 700 à 800 mm/an, sont réparties sur 70 à 80 jours environ.

Du fait de la faible hygrométrie, de l’existence de vents chauds et secs l’été et plus ou moins violents l’hiver, l’évapotranspiration est importante.

Ces caractéristiques ont des incidences sur le comportement de l’eau et des plantes et sont à l’origine du développement de modes particuliers de cultures que nous pouvons classer en cultures sèches (vigne-olivier) et cultures irriguées (oranger-pommier).
TABLEAU I. CONDITIONS CLIMATIQUES DES PERIODES D'ASPERSION

<table>
<thead>
<tr>
<th>Espèce</th>
<th>Température de l'air (°C)</th>
<th>Vitesse du vent (m/s)</th>
<th>Hygrométrie (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Min.</td>
<td>Max.</td>
<td>Appl. THO</td>
</tr>
<tr>
<td>Vigne</td>
<td>18</td>
<td>28</td>
<td>28</td>
</tr>
<tr>
<td>Oranger</td>
<td>2</td>
<td>14</td>
<td>10</td>
</tr>
<tr>
<td>Olivier</td>
<td>14</td>
<td>27</td>
<td>27</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>25</td>
<td>20</td>
</tr>
</tbody>
</table>

2.2.1. Schéma expérimental

Les expérimentations en conditions naturelles ont été réalisées sur des parcelles situées respectivement dans l’orangerie de la station de recherches agrumicoles de l’INRA\(^1\) de San Giuliano (Corse) [7], dans le vignoble expérimental de la station de technologie agricole de l’INRA à Narbonne [8] et dans l’oliveraie du CTGREF\(^2\) en Provence. Elles ont mis en œuvre:
— des pulvérisations d’eau triée sur le feuillage de vigne, d’oliviers et d’orangers,
— des injections dans le tronc de vigne et de pommiers.

Les aspersions d’eau triée appliquées au début de la floraison correspondent à des époques différentes de l’année, dont les caractéristiques figurent au tableau I. Pour simuler des conditions moyennes, la pulvérisation correspondait à une hauteur d’eau de 1 mm; 20 litres d’eau ont été distribués à chaque fois sur 20 m\(^2\) (tableau II).

Dans le cas des injections, 2,3 mCi d’eau triée ont été introduits dans le tronc de trois pommiers le 5 mai 1976 et le 16 juin 1976 et 8,9 mCi par souche dans six souches de vigne, les 7 juillet 1976, 25 août 1976 et 14 septembre 1976.


L’activité de l’eau libre et de la matière sèche a été mesurée sur des feuilles, des bois et des fruits.

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1 Institut national de la recherche agronomique.
2 Centre technique du génie rural, des eaux et des forêts.
TABLEAU II. TENEURS EN TRITIUM APRÈS IRRIGATION PAR ASPERSION

<table>
<thead>
<tr>
<th>Espèce</th>
<th>Tritium (pCi/ml)</th>
<th>Rapport B × 100 A</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A Eau d'irrigation</td>
<td>B Vapeur d'eau atmosphérique</td>
</tr>
<tr>
<td>Vigne</td>
<td>2,09 × 10^6</td>
<td>4 000</td>
</tr>
<tr>
<td>Olivier</td>
<td>3,7 × 10^6</td>
<td>17 670</td>
</tr>
<tr>
<td></td>
<td>1,37 × 10^6</td>
<td>14 100</td>
</tr>
<tr>
<td>Oranger</td>
<td>0,43 × 10^6</td>
<td>138</td>
</tr>
</tbody>
</table>

* Cette teneur est obtenue par condensation durant 15 heures.

TABLEAU III. CONCENTRATION (pCi/g) DU TRITIUM DANS LE SOL 19 HEURES APRÈS UN ARROSAGE PAR ASPERSION

<table>
<thead>
<tr>
<th>Type de sol</th>
<th>Sol vigne</th>
<th>Sol oranger</th>
<th>Sol olivier</th>
<th>Sol olivier</th>
</tr>
</thead>
<tbody>
<tr>
<td>Apport d'eau (litres/m²)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0,5</td>
</tr>
<tr>
<td>Apport de ³H (pCi/m²)</td>
<td>2090 × 10^6</td>
<td>433 × 10^6</td>
<td>1370 × 10^6</td>
<td>1850 × 10^6</td>
</tr>
<tr>
<td>Couche de sol</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0 – 10 cm</td>
<td>629</td>
<td>204</td>
<td>761</td>
<td>340</td>
</tr>
<tr>
<td>10 – 20 cm</td>
<td>267</td>
<td>119</td>
<td>562</td>
<td>246</td>
</tr>
<tr>
<td>20 – 30 cm</td>
<td>111</td>
<td>41</td>
<td>319</td>
<td>202</td>
</tr>
<tr>
<td>Moyenne 0 – 30 cm</td>
<td>335</td>
<td>121</td>
<td>547</td>
<td>262</td>
</tr>
<tr>
<td>T1/2 (humidité du sol 15%)</td>
<td>1 h 45</td>
<td>2 h</td>
<td>2 h 15</td>
<td>1 h 50</td>
</tr>
</tbody>
</table>

2.2.2. Comportement du tritium

Sur les parcelles d’essai, non recouvertes d’herbe et de perméabilité moyenne, l’eau tritiée se répartit dans le sol sur une couche au moins égale à 30 cm lorsque la teneur en eau est inférieure à la capacité au champ (tableau III). Ces conditions associées aux caractéristiques climatiques permettent un retour rapide de THO à l’atmosphère par évaporation directe; les demi-temps de résidence de THO dans les sols calculés sur 24 h sont compris entre 1 h 45 et 2 h 15.
A la suite des échanges avec l'atmosphère et le sol, le $^3$H se retrouve dans la plante incorporé à l'eau libre ou à la matière organique.

2.2.2.1. *L'eau libre* que l'on peut extraire du tissu végétal par évaporation sous vide est de l'eau que la plante évacue par transpiration dans l'atmosphère. Cette transpiration, régulée par l'ouverture et la fermeture des stomates à la suite de variations de pression osmotique dans les cellules, est directement liée aux conditions atmosphériques.

L'étude du dépôt de la vapeur d'eau tritiée sur les feuilles de vigne montre que la concentration du tritium dans l'eau des feuilles atteint son niveau d'équilibre d'autant plus lentement que les résistances stomatiques sont plus élevées. Le temps pour atteindre l'équilibre peut varier de quelques dizaines de minutes à plusieurs heures. Les expériences ont également montré que le rapport à l'équilibre entre la concentration du tritium dans l'eau des feuilles et celle dans l'air ne dépend que de la température.

Compte tenu de l'intérêt de l'évapotranspiration, il a paru intéressant de rechercher s'il existait une relation entre la teneur en tritium de l'eau transpirée et l'évapotranspiration potentielle calculée (ETP), selon la formule de Piche corrigée.

La figure 1 montre l'évolution du tritium de l'eau transpirée de la vigne et du pommier pendant les 7 jours qui suivent l'injection d'eau tritiée dans leurs journalières moyennes calculées sur 7 jours et le demi-temps de résidence du tritium.

Cette diminution de la teneur en tritium de l'eau libre de la plante peut provenir du fait qu'il n'y a pas renouvellement de l'apport de tritium dans la plante mais que, par contre, il y a apport continu d'eau non tritiée. La relation trouvée peut donc ne s'appliquer qu'au cas d'un rejet ponctuel dans l'environnement.

2.2.2.2. En ce qui concerne la *matière organique*, les demi-temps de résidence du tritium mesurés sont plus courts pour les feuilles que pour les fruits (tableau IV). Les pertes sont dues aux exportations vers les organes de réserve et de reproduction. Il est vraisemblable que l'enrichissement des fruits en matières hydrocarbonées aux dépens des feuilles contribue à maintenir leur concentration en tritium.

Chez l'olivier, le taux d'incorporation du tritium dans la matière sèche des feuilles semble être indépendant du volume d'eau apporté, c'est-à-dire de la dilution du radionucléide.

Le tableau V montre également que si, 36 jours après la pulvérisation, les taux d'incorporation en fonction du tritium déposé par unité de surface sont voisins, il n'en est pas de même pour les taux exprimés en fonction des concentrations de tritium par unité de volume. Il semblerait donc que la plante
incorpore dans sa matière sèche une même fraction du tritium déposé par unité de surface, quel que soit le mode d’apport, pour des volumes d’eau variant du simple au double et des conditions climatiques voisines au moment de la contamination.

2.2.3. Rôle de la vinification

A partir des vendanges tritiées obtenues sur les parcelles expérimentales ont été fabriqués selon les méthodes classiques du vin rosé (pressurage sans macération avant fermentation) et du vin rouge (pressurage après macération et fermentation).
FIG. 2. Demi-temps de résidence du tritium dans l'eau de transpiration (vigne, pommier) et ETP.
ETP = α E_p \{1 + \beta (T)\} avec α coefficient abri, E_p évaporation selon Piche (mm),
\( \theta = (t_x + 3 t_m)/4 \), \( t_x \) température maximale, \( t_m \) température minimale.

### TABLEAU IV. DEMI-TEMPS DE RESIDENCE (jours) DU TRITIUM DANS LA VIGNE, L'ORANGER ET L'OLIVIER

<table>
<thead>
<tr>
<th></th>
<th>Feuilles</th>
<th>Fruits</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MF(^a)</td>
<td>MS(^b)</td>
</tr>
<tr>
<td>Vigne</td>
<td>1,5 à 2</td>
<td>2 à 4</td>
</tr>
<tr>
<td>Oranger</td>
<td>50</td>
<td>12 à 100</td>
</tr>
<tr>
<td>Olivier</td>
<td>8</td>
<td>50</td>
</tr>
</tbody>
</table>

\(^a\) MF — matière fraîche; \(^b\) MS — matière sèche.
TABLEAU V. INFLUENCE DE LA DENSITE D’ARROSAGE SUR LA RETENTION DU TRITIUM PAR L’OLIVIER (FEUILLES)

<table>
<thead>
<tr>
<th>Densité d’arrosage (litres/m²)</th>
<th>Tritium</th>
<th>Facteurs de transfert</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>dans l’eau (pCi/l)</td>
<td>déposé sur le sol (pCi/m²)</td>
</tr>
<tr>
<td>0,5</td>
<td>$3700 \times 10^6$</td>
<td>$1850 \times 10^6$</td>
</tr>
<tr>
<td>1</td>
<td>$1372 \times 10^6$</td>
<td>$1372 \times 10^6$</td>
</tr>
</tbody>
</table>

La distillation de ces vins fait apparaître une différence de teneur en tritiurn entre le distillat constitué par de l’eau et des matières organiques volatiles (alcool, esters) et le résidu sec. Les matières sèches sont plus riches en tritiurn que le distillat particulièrement dans le cas du vin rouge. Ceci permet de supposer qu’une partie du tritiurn est transférée au vin au cours de la macération par l’intermédiaire de produits organiques tels que les tannins.

3. CONCLUSION

De nombreux points restent encore à éclucider et les travaux se poursuivent dans ce but dans nos différents laboratoires; néanmoins, certains points peuvent déjà être dégagés.

Pendant la période de croissance des plantes, en région méditerranéenne, les demi-temps de résidence du tritiurn dans les sols et l’eau de transpiration sont courts. Par contre, même en région méditerranéenne, ces mêmes temps de résidence pour la matière sèche sont relativement longs. Il y a donc deux cycles du tritiurn dans l’écosystème : un cycle court passant par l’eau libre, un cycle long passant par la matière organique. Sur le plan de la radioprotection, le cycle court réalimentera le milieu, le cycle long pourra influencer la ration alimentaire.

REFERENCES


DISCUSSION

Y. UENO: The uptake of tritium by leaves sprayed with tritiated water is strongly dependent on the age of the leaves. Did you consider this point in your experiment?

P. BOVARD: All sprayings were carried out during flowering. The results showing seasonal variations were obtained from injections into the stem, and absorption by the leaf itself played no part. It was therefore not possible to identify the role of leaf age. What we were trying to do was establish the fact that the behaviour of tritium varies with time. Leaf age may be one of the causes of this variation, but we cannot say what its relative importance is.

D.J. MEWISSEN: Could it be that seasonal variations in the residence time of organic components, as well as the age-dependent variations in plants, are somehow related to the metabolic rate as a general parameter? In mice we find different incorporation rates in newborn as opposed to juvenile or adult animals, as far as biologically bound tritium is concerned.

P. BOVARD: In our experiments we were interested exclusively in the action of external factors, particularly climatic conditions, and we demonstrated the role of evapotranspiration. One of the components of the latter, transpiration, is connected with the metabolic activity of the plant. It is therefore fully to be expected that metabolic activity would be related to the phenomena observed.
TRANSFER AND INCORPORATION OF TRITIUM IN MAMMALS*

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Abstract

TRANSFER AND INCORPORATION OF TRITIUM IN MAMMALS.

The metabolism of tritium in mammals has been studied in a number of laboratories which have participated in the IAEA Co-ordinated Research Programme on the Behaviour of Tritium in the Environment. The results of these studies are discussed and related to data obtained elsewhere. The animals studied are small laboratory and domestic animals. Tritium has been administered as THO, both in single and long-term dosing experiments, and also as organically bound tritium. The biological half-life of tritium in the body water pool has been determined in different species. The following values have been found: 1.1 days in mice; 13.2 days in kangaroo rats; 3.8 days in pigs; 4.1 days in lactating versus 8.3 in non-lactating goats and 3.1–4.0 days in lactating cows and steers. Much attention has been paid to the incorporation of tritium into organic constituents, both in the animal organism (organs, tissues) and in the secretions of the animal after continuous administration of tritium, mostly as THO. When compared with tritium levels in body water, and expressed as the ratio of specific activities, values of 0.25 and 0.40 have been found in mice liver and testis respectively. In cow’s milk, these ratios vary from 0.30 for casein to 0.60 for lactose. The transfer of tritium into milk after continuous ingestion of THO by a lactating cow is about 1.50% of the daily ingested tritium per litre of milk. Some results of experiments, utilizing organically bound tritium, are also presented.

* Part of the IAEA Co-ordinated Research Programme on the Behaviour of Tritium in the Environment.
1. INTRODUCTION

The main aims of the IAEA Co-ordinated Research Programme are to obtain information on the residence time, pattern of movement and distribution of tritium in typical ecosystems, its incorporation into biological compounds and its transfer along food chains to determine the biological significance of this radionuclide.

Knowledge on the pattern of movement, metabolism and incorporation of tritium in mammalian species is of importance for answering questions concerning radiobiological and health hazard problems, and also because of the importance of certain mammalian species as a source of human food. For these reasons, research projects designed to obtain information on the parameters mentioned above have been included in the programme. These projects have been carried out in Belgium, the Netherlands, the Philippines and the United States of America.

Tritium metabolism has been investigated in small laboratory and domestic animals. The former group has the advantage of experimental simplicity, and is particularly suitable for investigations on the incorporation of tritium in important biological molecules. The experiments involving domestic animals have produced information on the tritium content of such important food items as meat and milk, and on the incorporation of tritium into the organic constituents of these products. Furthermore, in one study, a small rodent (kangaroo rat, Dipodomys deserti) which had been living for several generations in a natural environment in which tritium levels were relatively high, was taken to the laboratory for detailed study of metabolism and distribution of tritium in the animal organism.

The studies mentioned above can be divided roughly into experiments in which tritium was administered as tritiated water (THO), and experiments in which organically bound tritium was ingested by the animals. The experimental results of some of these studies have been published during the operation of the co-ordinated research programme. It is the purpose of this paper to review and discuss the main results, supplemented with non-published results of more recent experiments, and to relate these to data obtained elsewhere.

2. METABOLISM OF TRITIUM, ADMINISTERED AS THO

2.1. Biological half-life of tritium

2.1.1. Experimental conditions

Single doses of tritium may be administered either orally or parenterally. This means ingestion via drinking water in the former case, and intravenous, intramuscular or intraperitoneal application in the latter. Single dose experiments have been carried out on domestic animals in all participating laboratories.
TABLE I. BIOLOGICAL HALF-LIFE OF TRITIUM IN BODY WATER AFTER ADMINISTRATION OF THO

<table>
<thead>
<tr>
<th>Species</th>
<th>Biological half-life (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st component</td>
</tr>
<tr>
<td>Mouse</td>
<td>1.1</td>
</tr>
<tr>
<td>Kangaroo rat</td>
<td>13.4 ± 0.7; 13.2 ± 1.3</td>
</tr>
<tr>
<td>Goat (lactating)</td>
<td>4.1 ± 0.1 (2.9 - 5.3)</td>
</tr>
<tr>
<td>Goat (non-lactating)</td>
<td>8.3 ± 0.5 (6.7 - 10.4)</td>
</tr>
<tr>
<td>Miniature goat (non-lactating)</td>
<td>4.3 ± 0.2</td>
</tr>
<tr>
<td>Pig</td>
<td>3.8 - 4.3</td>
</tr>
<tr>
<td>Cow (lactating)</td>
<td>3.1; 3.3; 3.5; 4.0</td>
</tr>
<tr>
<td>Cow (non-lactating)</td>
<td>4.0 ± 0.2</td>
</tr>
<tr>
<td>Chicken</td>
<td>4.6</td>
</tr>
</tbody>
</table>

Continuous administration of THO is limited under most experimental conditions to the oral route. It is essential that the specific activity of the drinking water remains constant in the course of the experimental period to ensure that equilibrium conditions between intake and excretion are reached. This condition requires special experimental precautions in the case of lactating dairy cows which have a water consumption of about 40 litres per day while the duration of the experiment is thirty days or more. Such investigations have been performed in Belgium, the Netherlands and the United States of America on both laboratory and domestic animals.

2.1.2. Results and discussion

Table I summarizes the values of the biological half-life of tritium in the body water of a variety of species, obtained in different studies. The values for a second, long half-life component, have been derived from long-term intake experiments.

The data in Table I are in good agreement with results published in the literature. Richmond and co-workers [1] give values of 1.13 ± 0.14 days for mice and of 11.82 ± 2.96 days for kangaroo rats. Cunningham [2] found a half-life of 3.3 days in pigs with free access to water and of 6.3 days in pigs with restricted water intake. This shows that the availability of water may influence water turn-over. Another factor which may have an effect is the ambient temperature. Variations in ambient temperature are often seasonal.
and seasonal changes in the biological half-life of tritium have been reported for animals [3] and man [4]. An average value of 3.54 ± 0.10 days (range 3.0–3.9 days) for lactating dairy cows and of 3.4 ± 0.18 days (range 2.8–4.1 days) for non-lactating, male cows between 1–1.5 years old has been reported [5]. It appears from these data that lactation does not influence water turn-over in the cow. The results presented in Table I suggest that lactation in goats brings about a shortening of the residence time of tritium in the body water. The physiological effect of lactation on water metabolism would be an increase in turn-over rate of water for a body water pool of the same size, and consequently a shorter biological half-life. Apparently, lactation has different consequences on water metabolism in the cow and in the goat, both ruminant animals. More detailed investigations are necessary to provide quantitative information on this interesting subject.

Another point of interest is the correlation between water turn-over and body weight. Richmond and co-workers [1] have shown that a log-log correlation between these two parameters exists in the mouse, rat, rabbit, man and horse. This was extended by Yousef [6] to some other species including small and large mammals living in the desert. Only the kangaroo rat is exceptional. Its unique water metabolism has been the subject of many investigations. The dairy cow occupies a special place, having a water turn-over comparable with that of a rabbit. This cannot be ascribed to the physiological effects of being a ruminant animal since the correlation between body weight and water turn-over does in fact hold for goat, sheep, reindeer and other ruminants.

A slower component has been established for the kangaroo rat [7] and for the lactating [8] and non-lactating dairy cow, as shown in Table I. Both values have been derived from continuous administration experiments. Figure 1 shows the evolution of tritium activity in milk of a lactating cow which had been ingesting tritiated water for nearly six weeks, quite long enough for equilibrium conditions between intake and secretion into milk water to occur. After discontinuing the administration of tritiated water, a biological half-time of four days of tritium in milk water was found. The slower component represents less than 1% of the tritium activity at equilibrium and is of limited importance for the total body dose from tritium. A similar situation exists in the kangaroo rat [7] and in man [9].

The possibility of an isotope effect occurring in the passage of tritium across body membranes has been examined by various authors. In the case of tritiated water, the lung has been found to be the only site where discrimination is significant, the specific activity in the expired air being less than in the body water. In mice and kangaroo rats, the specific activity in the expired air was found to be 0.65 and 0.44 respectively of the mean tissue water specific activity [10]. In our own experiments on lactating cows, calves and pigs, differences in specific activity of tritium in its passage in liquid form through the body have not been observed.
2.2. Incorporation of tritium in organic constituents

2.2.1. Experimental conditions

These have been identical with those described for the determination of the biological half-life. Both single and continuous dose studies on small laboratory and domestic animals have been carried out.

The radiochemical analysis of organically bound tritium (OBT) after administration of tritiated water requires special attention because of the high tritium activity of the water fraction relative to that in the OBT fraction. Therefore, complete evacuation of THO, for example by drying under vacuum conditions, is essential. Combustion of the sample is another critical step. It has been carried out originally by the oxygen flask combustion technique, first introduced by Schöniger. The availability of automatic sample oxidizers has facilitated greatly this part of the analysis. Nevertheless, we feel that adoption of a standard procedure of proven reliability is essential in order to obtain reproducible analytical results.

2.2.2. Results and discussion

2.2.2.1. Incorporation in organic matter

A cow that had been given orally a large single dose of tritiated water had to be killed three weeks afterwards. Tritium activities were determined in the tissue-free water (TFWT) and in the dry tissue solids, the OBT fraction. The ratio of the specific activities of these two fractions varied from 0.19 (intestinal wall) to 0.63 (liver), with intermediate values for muscle (0.33) and kidneys (0.44). The organic milk constituents (casein, lactose, milk fat) also contained tritium, and the highest values were found from 12 to 24 hours after ingestion of THO. Expressed as percentage of the ingested tritium, secreted per litre of milk, the transfer coefficient for milk water is about 0.2% when milk activity is at a maximum and 2–3 orders of magnitude smaller for the organic constituents. In mice, killed 50 days after an acute exposure, highest tritium levels were observed in brain tissue, followed by muscle, kidney and liver [10]. Radwan and co-workers [11] determined $^3$H activity in various rat tissues at different intervals after a single subcutaneous dose of THO. Maximum incorporation was found between day 4 and 7 after dosing, with lowest levels in muscle and highest activity in liver. Species differences and time-dependent factors may influence tritium incorporation into tissues after an acute exposure.

Conditions of chronic intake of tritiated water are more likely to occur under practical circumstances, and therefore studies involving continuous administration are of greater interest. In mice, kept on THO for 40–147 days, the specific activity
TABLE II. AVERAGE TRANSFER COEFFICIENTS FOR MILK AND MILK CONSTITUENTS, EXPRESSED AS PERCENTAGE OF THE DAILY TRITIUM INTAKE, SECRETED IN ONE LITRE OF MILK, AFTER CONTINUOUS INGESTION OF THO

<table>
<thead>
<tr>
<th>Product</th>
<th>Transfer coefficient (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Milk THO</td>
<td>1.50</td>
</tr>
<tr>
<td>Dry matter</td>
<td>0.067</td>
</tr>
<tr>
<td>Milk fat</td>
<td>0.026</td>
</tr>
<tr>
<td>Lactose</td>
<td>0.029</td>
</tr>
<tr>
<td>Casein</td>
<td>0.011</td>
</tr>
<tr>
<td>Albumin</td>
<td>0.002</td>
</tr>
<tr>
<td>Total</td>
<td>1.57</td>
</tr>
</tbody>
</table>

TABLE III. RATIO OF THE SPECIFIC ACTIVITIES (pCi\(^{15}H/gH) OF SOME ORGANIC MILK COMPONENTS AND OF INGESTED WATER AND FOOD IN A LACTATING COW AFTER CONTINUOUS DAILY INGESTION OF THO

<table>
<thead>
<tr>
<th>Component</th>
<th>S.A. Component</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>S.A. (H_2O) ingested</td>
</tr>
<tr>
<td>Total dry matter</td>
<td>0.49</td>
</tr>
<tr>
<td>Lactose</td>
<td>0.60</td>
</tr>
<tr>
<td>Milk fat</td>
<td>0.40</td>
</tr>
<tr>
<td>Casein</td>
<td>0.30</td>
</tr>
<tr>
<td>Albumin</td>
<td>0.32</td>
</tr>
</tbody>
</table>
FIG. 1. The evolution of $^3$H activity in milk and in milk constituents after continuous administration of THO for 41 days to a lactating cow.

(S.A.) of the OBT fraction from liver and testis was 0.25 and 0.40 respectively, of the S.A. of tissue water [12]. The higher ratio for testis could not be explained. Liver DNA was shown in this study to contain tritium in all four bases. A mean value of 0.25 for the OBT/TFWT specific activity was also found in 11 organs of a baby goat whose mother had been ingesting daily tritiated water (46.8 $\mu$Ci/l) during the 124-day gestation period [13]. Similar values have been obtained for organs of calves after chronic ingestion of THO. The mean value is 0.20, the range 0.19–0.25 in five organs. The same organs of pigs show lower values (mean: 0.13, range: 0.10–0.15) [14]. There is no apparent reason for these differences. It may be that equilibrium conditions between intake and excretion...
were not obtained in the experiments with pigs. These studies show reasonably good agreement with earlier experiments [15].

The incorporation of tritium into the organic constituents of cow’s milk is of special interest because of the importance of milk for the contamination of the food chain. Table II shows the transfer of tritium into various milk components at equilibrium after continuous ingestion of THO by a lactating cow, expressed as the percentage of ingested tritium secreted per litre of milk. It follows from Table II that the overall transfer for milk is a little over 1.5% of the daily ingested tritium per litre. Milk dry matter, which constitutes about 13% of milk, contains only 4% of the tritium activity. Calculation of the ratio of the specific activities of milk constituents, and of ingested water and feed (Table III) indicates the measure of incorporation of tritium into organic material. Considerable differences between the various organic milk components can be noted in this respect, the difference between the values for lactose (0.60) and for the two milk proteins, casein (0.30) and albumin (0.32), being particularly noteworthy. Glucose is the only precursor of lactose, and in ruminant animals, it is newly formed from propionic acid. This may explain why more than half the hydrogen of lactose originates from water.

2.2.2.2. Biological half-life

After discontinuing the daily intake of THO by the lactating cow, tritium activity in the milk water decreases with a half-life of four days (Fig. 1), which is in close agreement with the values observed after a single dose. A similar value can be determined for the organic milk constituents over the period of three weeks following the end of THO administration, as represented in Fig. 1. This is what one would expect since the organically bound tritium will be derived entirely from tritium in the body water pool. The milk proteins are behaving somewhat differently in that a slower component becomes visible after about ten days. Analyses are in progress to clarify this point. Kirstner [16] has reported different half-lives for whole milk (five days) and dry matter (seven days) after a daily intake of THO for 40 days. This is an interesting observation which needs to be verified since there is no obvious physiological reason for the differences in half-life observed. Most of the organic material in milk is newly synthesized by the mammary epithelial cells from blood precursors. These precursors originate for the greater part from the animal’s feed, and presumably would not contain any tritium before being taken up by the animal.

In mice longer biological half-times of 11 days [10] and of 90 days and longer [15] have been reported. These were interpreted as representing excretion of tritium incorporated into organic tissue components.
TABLE IV. DISTRIBUTION OF TRITIUM IN ORGANIC FRACTION OF CALF ORGANS (% of total activity ingested)

<table>
<thead>
<tr>
<th>Organs</th>
<th>$^3$H administered in $\theta$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Water</td>
</tr>
<tr>
<td>Kidney</td>
<td>0.033</td>
</tr>
<tr>
<td>Spleen</td>
<td>0.020</td>
</tr>
<tr>
<td>Brain</td>
<td>0.023</td>
</tr>
<tr>
<td>Lungs</td>
<td>0.051</td>
</tr>
<tr>
<td>Muscles</td>
<td>2.46</td>
</tr>
<tr>
<td>Tongue</td>
<td>0.030</td>
</tr>
<tr>
<td>Heart</td>
<td>0.046</td>
</tr>
<tr>
<td>Liver</td>
<td>0.14</td>
</tr>
<tr>
<td>Sum</td>
<td>2.803</td>
</tr>
</tbody>
</table>

3. METABOLISM OF TRITIUM, ADMINISTERED AS ORGANICALLY BOUND TRITIUM (OBT)

3.1. Incorporation of tritium in organic matter

3.1.1. Experimental conditions

As has been discussed in the previous section, administration of THO leads to tritiation of organic components in animal tissues and organs, and in animal products such as milk. Thus, when considering the question of how much tritium becomes incorporated after ingestion of organically bound tritium by an animal, e.g. through its feed, it is necessary that no THO reaches the animal or, if it does, that the amount of it be known. In the latter case, it is preferable that the amount of THO is small relative to the organically bound tritium. Another point to remember is that a relatively substantial amount of homogeneously tritiated feed should be available for equilibrium conditions to be reached. Obviously, this kind of investigations is not easily realized experimentally. A few studies, partly of a preliminary character, have been carried out within the framework of the IAEA Co-ordinated Research Programme. These are briefly discussed below.
3.1.2. Results and discussion

Two calves were fed tritiated milk powder for 28 days before they were killed. Various organs and tissues were analysed for tritium content, and the results are shown in Table IV. Similar data from calves that had been ingesting THO for periods of between 4–6 weeks are given also for comparison. The much higher incorporation of $^3$H into eight organs and tissues after ingestion of OBT in the feed is obvious: on the average, fifteen times more tritium has been incorporated, representing 4.1% of the ingested tritium. Pigs fed tritiated milk powder incorporated 4.2% of the ingested tritium in a slightly different set of
organs, and nearly 11% after feeding on tritiated potatoes [14]. This would appear to illustrate clearly the importance of the tritiated ‘precursor’.

Figure 2 shows incorporation of $^3$H in various milk components after the feeding of tritiated hay to two lactating cows for eight days. Although equilibrium conditions were almost certainly not reached in so short a period of time, the following conclusions may be drawn. Firstly, that milk fat and casein contain fifteen and ten times more $^3$H on a weight basis than milk water, whereas lactose has a surprisingly low tritium content. Secondly, the half-life of tritium in milk fat, casein and milk dry matter is of the order of two days for the first week after stopping the feeding of tritiated hay. For milk water, a value of four days is found. After about two weeks, a slower component of 8–10 days for the decrease of tritium activity in milk fat appears. No data are available for the other milk components.

The results reported here for the incorporation of tritium taken up as OBT, are clearly of a preliminary character. Plans are being made to continue this line of research where extension of existing knowledge is desirable.

REFERENCES

DISCUSSION

S. STRACK: In your Fig. 1, showing tritium activities in milk and in milk constituents after continuous administration of HTO, I discern differences in the biological half-lives of the milk constituents, because the eliminating curves are intersecting. Did you calculate these biological half-lives? I think it is rather important to do this, since the ratios given in Table III will then be shifted with time.

J. VAN DEN HOEK: The elimination curve of tritium in milk fat and lactose follows closely that of tritium in milk water. This is what one would expect, since tritium in organic milk constituents is derived entirely from tritiated water from the body water pool. There is no apparent reason why casein and albumin should behave differently, although it appears from the data that they deviate slightly in the direction of a longer half-life. Analyses are in progress to clarify this point. It is quite likely that the first component of the elimination of tritium from casein and albumin will be the same as for lactose and milk fat.
TRANSFER OF TRITIUM TO FOETUSES AND NEWBORNS FROM MOTHER MICE ADMINISTERED WITH TRITIATED WATER

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Abstract

TRANSFER OF TRITIUM TO FOETUSES AND NEWBORNS FROM MOTHER MICE ADMINISTERED WITH TRITIATED WATER.

The kinetics of tritium release from the skin, liver and brain of pregnant and nursing mice or foetuses and sucking newborns were studied after single subcutaneous or intraperitoneal injections and the per oral uptake. When female mice were injected subcutaneously at various times during pregnancy, tritium in wet tissues at delivery was reduced linearly on a semi-log scale with prolongation of the exposure time in both pregnant females and foetuses. Tritium in dry tissues was not reduced for a short time before delivery. The estimated half-lives of tissues were longer in pregnant females than in non-pregnant ones. The half-lives of wet tissues were shorter in pregnant females than in foetuses but those of dry tissues were shorter in foetuses. The estimated doses in wet and dry tissues for 10 days after delivery were larger in foetuses than in pregnant females. In the oral uptake, the tritium in wet tissues of both pregnant females and foetuses increased with prolongation of the drinking period, reached a maximum after about 200 h and fell with the further prolongation of the period. The patterns of tritium release in nursing females injected intraperitoneally immediately after delivery were similar to those of normal females, and those of sucking newborns depended upon those of nursing females. The estimated doses in tissues at 300 h after delivery were larger in nursing females than in sucking newborns except for the amount in the skin.

1. INTRODUCTION

Studies on the metabolism of tritium in tritiated water are based on factors in the present condition, that is, the industry in which extremely large amounts of tritium are handled [1, 2] and inadequate assessment of physiology or physiochemistry regarding water metabolism. Furthermore, the data in these studies are
based on radiobiological effects of tritium as a contribution to such subjects as physiology. These problems have been surveyed by the present authors [3]. Data on the metabolism of tritium in water molecules were collected by the present authors [4]. The results of analysis suggested that the effective half-lives of tritium released from the bodies of many kinds of mammals depended upon body weights, as pointed out elsewhere [5]. The dependency is as follows:

\[
\begin{align*}
y &= 4.04 \times 10^{0.04} & \text{for the first fraction} \\
y &= 18.19 \times 10^{0.14} & \text{for the second fraction} \\
y &= 173.78 \times 10^{0.25} & \text{for the third fraction}
\end{align*}
\]

where \( y \) is effective half-life in days and \( x \) is body weight in kilograms.

In spite of the accumulation of papers on tritium metabolism, there is a lack of data on the transfer of tritium in water molecules from pregnant or nursing females to their foetuses or sucking newborns. The present report deals with these problems.

2. MATERIALS AND METHODS

2.1. Animals

The dd/YF, 13-weeks-old female mice used in the present experiment were housed in individual cages maintained in a special apparatus to protect the researchers from tritiated vapour released from the contaminated mice and drinking water. Virgin females were mated in a colony in our laboratory. In the first experiment, pregnant females were injected subcutaneously with tritiated water at various times during pregnancy, and in the second experiment were continuously provided with tritiated drinking water at various times during pregnancy to delivery. In the third experiment, nursing females were injected intraperitoneally immediately after delivery.

2.2. Tissues

Three small pieces of the liver, the brain and the skin were removed immediately after delivery from the mother and three offsprings in the first and second experiments. In the third experiment, the pieces were removed from a sucking newborn every 24 h after delivery and from the nursing female. Three pieces were used for each measurement. The pieces were weighed and kept in 1 ml of Soluene (Packard) at room temperature in a dark room. Another three pieces were dried by a frozen-dehydration method for 48 h, weighed and dissolved in Soluene. The tritium counting was carried out as reported previously [6].
2.3. Tritiated water

Tritiated water (specific activity: 92.3 MBq/ml) was used. Mice were injected with tritiated water at the rate of 370 kBq/g of body weight in the first and third experiments, and drank tritiated water with an activity of 370 kBq/ml in the second experiment. Estimation of absorbed doses was carried out as reported previously [7, 8].

3. RESULTS

3.1. Experiment 1

Radioactivities of three wet tissues from the mother and offsprings were plotted according to the time intervals between injection and delivery on a semi-logarithmic scale. The activities in the liver were reduced linearly for 300 h before delivery, as the time interval was prolonged. The results are shown in Figs 1 and 2. The ratios of the activities (foetuses/pregnant females) were calculated, with the results shown in Fig.3. The ratios were larger than 1.0 and increased as the time interval was prolonged and reached 1.7 in the brain. Activities in dry tissues were constant for about 100 h before delivery and decreased linearly as the time interval was prolonged more than about 100 h (Figs 1 and 2). The ratios were constant for some intervals before delivery and decreased further before these intervals. The ratios were more than 1.0 for about 250 h before delivery. The half-lives calculated using slopes in Figs 1 and 2 correspond to the effective half-lives of pregnant females and foetuses. The effective half-lives were longer in pregnant females than in non-pregnant females [7, 8]. Those in the wet tissues...
FIG. 2. Tritium activity in the liver of foetuses in females injected subcutaneously with tritiated water. The time interval shows the time from injection to delivery.

FIG. 3. The ratios of tritium activities in tissues of pregnant females and foetuses (foetuses/ pregnant females).

were longer in foetuses than in pregnant females, whereas those in the dry tissues were longer in pregnant females than in foetuses. The results are shown in Table I. The estimated doses of mother and offsprings for 10 days after delivery using the half-lives in pregnant females and foetuses are shown in Table II. The ratios of absorbed doses were more than 1.0 in all cases and reached 2.75 in the dry skin.

3.2. Experiment 2

The activities in the liver in pregnant females were constant for about 100 h of the prolonged drinking period, and fell when the prolonged drinking period exceeded 200 h. The results are shown in Figs 4 and 5. The ratios of activities varied from 0.2 to 3.0 and did not depend linearly upon the drinking period.
TABLE I. EFFECTIVE HALF-LIVES OF FOETAL, PREGNANT AND NON-PREGNANT MICE (hours)

<table>
<thead>
<tr>
<th></th>
<th>Foetal mice</th>
<th>Pregnant mice</th>
<th>Non-pregnant mice</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wet skin</td>
<td>71.44</td>
<td>67.28</td>
<td>62.41</td>
</tr>
<tr>
<td>liver</td>
<td>67.94</td>
<td>55.89</td>
<td>50.48</td>
</tr>
<tr>
<td>brain</td>
<td>73.41</td>
<td>54.14</td>
<td>57.65</td>
</tr>
<tr>
<td>Dry skin</td>
<td>57.27</td>
<td>130.75</td>
<td>62.45</td>
</tr>
<tr>
<td>liver</td>
<td>61.88</td>
<td>63.58</td>
<td>62.46</td>
</tr>
<tr>
<td>brain</td>
<td>68.61</td>
<td>86.19</td>
<td>72.04</td>
</tr>
</tbody>
</table>

TABLE II. ESTIMATED ABSORBED DOSES FOR 10 DAYS AFTER DELIVERY (SINGLE TRANSPLACENTAL TRANSFER) (mGy)

<table>
<thead>
<tr>
<th></th>
<th>Newborn</th>
<th>Mother</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wet skin</td>
<td>84.5</td>
<td>60.3</td>
<td>1.40</td>
</tr>
<tr>
<td>liver</td>
<td>75.2</td>
<td>73.2</td>
<td>1.03</td>
</tr>
<tr>
<td>brain</td>
<td>84.8</td>
<td>74.3</td>
<td>1.14</td>
</tr>
<tr>
<td>Dry skin</td>
<td>46.4</td>
<td>16.9</td>
<td>2.75</td>
</tr>
<tr>
<td>liver</td>
<td>33.3</td>
<td>27.7</td>
<td>1.20</td>
</tr>
<tr>
<td>brain</td>
<td>39.3</td>
<td>14.4</td>
<td>2.73</td>
</tr>
</tbody>
</table>

FIG. 4. Tritium activity in the liver of pregnant females drinking tritiated water. The period of drinking shows the time from the start of drinking tritiated water to delivery.
3.3. Experiment 3

The activities in the wet liver in pregnant females increased rapidly to reach a maximum about 20 hours after injection; those in the dry liver reached a maximum after about 60 h. Later, the activities in the wet and dry livers fell until about 300 h and thereafter were constant. The kinetics in the wet liver of sucklings were similar to those in nursing females, but the maximum in activity was observed about 50 h later. The activity in the dry liver reached its maximum after 40 h and thereafter fell slightly followed by a slight increase, and thereafter fell from 120 to 300 h. The slight wave form in the activity in the dry liver was observed in other tissues. The absorbed doses of nursing females and sucklings were estimated for 300 h after delivery. The results are shown in Table III. The ratios (sucklings/nursing females) of absorbed doses were less than 1.0 except in the skin.

4. DISCUSSION

Although water metabolism in pregnant females and foetuses is a very complex process [9, 10] and could be a particularly interesting problem in the biology of reproduction, the present discussion is concerned with the problems of kinetics of tritium and health physics.

The patterns of tritium metabolism in wet tissues as shown in Figs 1 and 2 were similar to those in singly injected normal females [7, 8]. The reason why the tritium content is higher in foetuses than in pregnant females could be reasonably
explained by mechanisms specific to pregnancy and foetal life. The pattern in dry tissues including a plateau phase cannot be observed in normal females [7, 8]. This could be new information specific to pregnancy. The long effective half-lives in the pregnant females seem to be based on a specific water balance and biochemical processes during pregnancy [9, 10]. The short effective half-lives in dry foetal tissues compared with those of pregnant females could be explained by the reason that a relatively large amount of non-labelled substances transfer from pregnant females to foetuses to induce a relative small tritium content in foetuses. The phenomenon does not imply the fast excretion of tissue-bound tritium from foetuses, but a kind of self-burial. The estimated absorbed doses were larger in foetuses than in pregnant females. This is an important observation, because the absorbed doses in the case of lacteal transfer as shown in Table III were smaller in sucklings than in nursing females.

The patterns in wet tissues in the case of chronic administration were interesting. The unexpected occurrence of peaks as shown in Figs 4 and 5 and of a plateau in Fig.4 [11, 12] might have been induced by disturbances of very complex paths of tritium in metabolism due to pregnancy, though the disturbances have not yet been analysed in the study on reproduction. The plateau observed in Fig.4 could be based on a mechanism similar to that considered in the patterns of dry tissues in the case of a single injection. The ratios of activity were never larger than 1.0. The ratios in the dry skin, brain and liver varied during pregnancy, and the maximum was more than 3.0 in the skin. The ratios in the liver were less than 1.0 except for 1.5 at the maximum. The relative absorbed doses of foetuses were not as high as surmised from the results in the case of a single injection. However, it is very interesting and important that the tritium uptake is greater when pregnant females start to drink tritiated water in the late organogenesis

<table>
<thead>
<tr>
<th></th>
<th>Newborn</th>
<th>Mother</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wet</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>skin</td>
<td>34.8</td>
<td>33.9</td>
<td>1.03</td>
</tr>
<tr>
<td>liver</td>
<td>32.0</td>
<td>54.4</td>
<td>0.59</td>
</tr>
<tr>
<td>brain</td>
<td>34.2</td>
<td>64.5</td>
<td>0.53</td>
</tr>
<tr>
<td>Dry</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>skin</td>
<td>16.8</td>
<td>11.0</td>
<td>1.53</td>
</tr>
<tr>
<td>liver</td>
<td>15.1</td>
<td>17.6</td>
<td>0.86</td>
</tr>
<tr>
<td>brain</td>
<td>16.0</td>
<td>20.4</td>
<td>0.78</td>
</tr>
</tbody>
</table>
of foetal life (Figs 4 and 5), so that there is a critical time in chronic oral uptake. The kinetics of radioactivities in the tissues of nursing females were similar to those in normal females injected with tritiated water. The slight wave form of patterns in dry tissues of sucklings might be related to the foetal growth specific to this period.

The present results show that the kinetics of tritium in pregnant females differ from those in non-pregnant females and the transplacental transfer to foetuses was more harmful compared with the lacteal transfer.

REFERENCES


METABOLISME DE MOLECULES TRITIEES CHEZ LE RAT
Etude sous l'angle de la radioprotection

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Abstract—Résumé

METABOLISM OF TRITIATED MOLECULES IN RATS: A STUDY FROM THE STANDPOINT OF RADIOLOGICAL PROTECTION.

To promote the radiological protection of workers and the population at large, a study was made of the metabolism of tritiated molecules in rats. The molecules selected so far are sodium acetate, thymidine, oleic acid, benzoic acid and, for purposes of comparison, tritiated water. Elimination and retention were traced for at least 500 days so as to make an optimum evaluation of the long-half-life component. The retention and cumulative activity vary according to the organ, the type of molecule and the method of administration.

METABOLISME DE MOLECULES TRITIEES CHEZ LE RAT: ETUDE SOUS L'ANGLE DE LA RADIOPROTECTION.

Pour assurer la radioprotection des travailleurs et de la population, l'étude du métabolisme de molécules tritées a été entreprise chez le rat. Les molécules retenues jusqu'à présent sont l'acétate de sodium, la thymidine, l'acide oléique, l'acide benzoïque et à titre de comparaison l'eau tritée. L'élimination et la rétention ont été suivies pendant au moins 500 jours de façon à apprécier le mieux possible la composante à longue période. La rétention et l'activité cumulée varient selon l'organe, la nature de la molécule, le mode d'administration.

1. INTRODUCTION

La surveillance radiotoxicologique du personnel travaillant avec du tritium est effectuée par des mesures de l'activité dans l'urine. L'interprétation des résultats est simple avec l'eau tritée, mais il n'en est pas de même lorsque l'agent contaminant est une molécule organique marquée, car les informations sur le devenir du tritium manquent la plupart du temps.

D'autre part, le développement industriel de l'énergie nucléaire s'accompagne de rejet dans l'environnement d'une certaine quantité de tritium
dont une partie se retrouve sous forme organique dans la chaîne alimentaire et pénètre ainsi dans l'organisme humain.

Il est donc important non seulement pour la radioprotection des travailleurs mais aussi pour celle du public de connaître le devenir du tritium lié aux molécules dans les organismes vivants. Ce sont les raisons pour lesquelles des études ont été entreprises sur le métabolisme d'un certain nombre de molécules tritiées chez un animal de laboratoire, le rat.

2.- Méthodes

Les molécules étudiées jusqu'à présent sont l'acétate de sodium, la thymidine, l'acide oléique, l'acide benzoïque et à titre de comparaison une tritiée. Elles ont été administrées en une fois à des rats par voie digestive ou intrapéritonéale (IP). L'élimination du tritium est suivie régulièrement dans les urines fraîches et les selles sèches. Des rats sont sacrifiés à partir du quatrième jour, à intervalles plus ou moins rapprochés en fonction de la vitesse d'élimination du tritium. On s'efforce de suivre l'expérimentation pendant la durée de vie normale des rats c'est-à-dire jusqu'à 600 jours après l'administration. Sur chaque animal le tritium est mesuré dans vingt à vingt-cinq tissus frais et secs pour connaître la rétention globale et la rétention de la fraction fixée. L'ensemble des valeurs permet de calculer par interpolation les activités cumulées. Les rapports (1 - 4) contiennent tous les résultats expérimentaux obtenus sur les tissus, ce qui permet au lecteur d'utiliser les résultats à sa convenance et d'en tirer sa propre interprétation.

3.- Résultats

3.1.- Élimination

Parmi les molécules étudiées, l'acide benzoïque s'est comporté de façon particulière. L'acide benzoïque marqué sur le cycle benzénique est caractérisé par une élimination très rapide. Après vingt-quatre heures on retrouve 50 à 80 % de l'activité dans l'urine et 20 à 40 % dans les selles. L'acide benzoïque est éliminé tel quel ou bien sous forme d'un composé d'addition avec le glycollate : l'acide hippurique. Le quatrième jour, on retrouve dans le rat moins de 1 % de l'activité injectée par voie IP. Seuls les poils montrent une certaine activité, mais il s'agit probablement d'une contamination externe, car les valeurs diminuent après lavage de l'échantillon.
Dans les autres cas le tritium est éliminé préférentiellement sous forme d'eau tritée après que la molécule marquée ait été soumise aux réactions métaboliques dans l'organisme. De ce fait, même si la transformation est très rapide, la période d'élimination de l'activité sera au moins celle de l'eau. Les molécules étudiées dans la suite de ce rapport ont un comportement de ce type.

3.2. - Rétention

Le tritium est présent dans tous les tissus ou organes à la fois sous forme d'eau tritée et sous une forme fixée qui peut être différente de la molécule initiale.

Les résultats expérimentaux semblent indiquer que l'on pourrait décrire l'évolution de la rétention dans un organe par la somme de trois exponentielles. La réalité ne semble cependant pas obéir à une loi aussi simple.

- La première exponentielle, bien définie avec une période de quatre jours, correspond à l'élimination de l'eau libre et elle représente la majeure partie de l'activité.

- Les deux autres périodes ne peuvent être calculées avec précision, car leurs valeurs apparentes augmentent avec la durée de l'expérimentation ; en outre, la dispersion des résultats a tendance à croître au cours du temps d'expérience, alors qu'une meilleure précision serait d'autant plus nécessaire que la période que l'on essaie d'évaluer est plus longue.

C'est pourquoi en général nous évitons de calculer des courbes de rétention qui nous semblent artificielles. Les résultats sont portés sur une figure où est inscrite une courbe de rétention théorique calculée par intégration de l'élimination urinaire.

La comparaison des courbes de rétention montre leur variété. Même pour des tissus à première vue assez semblables, des différences sensibles peuvent apparaître au cours du temps. Un bon exemple est donné par le système nerveux central. Après administration d'eau tritée, la rétention des diverses parties montre des écarts systématiques, les valeurs décroissant dans l'ordre suivant : moelle épinière, bulbe, cervelet, hémisphères cérébraux (figure 1). Un phénomène analogue se produit avec la thymidine et l'acide oléique ; dans ce dernier cas les résultats sont dans l'ensemble trois fois plus élevés qu'avec l'eau tritée.
FIG.1. Fraction de l'activité retenue par gramme de tissu frais dans différentes parties du système nerveux central. La courbe en tirets est la courbe de rétention calculée.

Ayant à l'esprit les difficultés citées au début de ce chapitre on peut tout de même dire que les courbes de référence ont une période intermédiaire comprise entre 10 et 20 jours et une période longue qui va de 70 à 200 jours. La fraction qui correspond à la période la plus longue représente environ 1% de l'activité présente au quatrième jour pour l'acétate et la thymidine, et 10% pour l'acide oléique.

Nous avons comparé nos résultats correspondant à la fraction à longue période de rétention avec ceux que THOMPSON (5) a obtenus après ingestion continue d'eau tritée. Les valeurs absolues sont différentes, mais l'ordre de classement des organes est le même (figure 2). Nous avons également noté au cours d'un essai préliminaire que la fraction retenue à longue période varie avec l'âge auquel on contamine le rat.

3.3.- Activité cumulée

Les courbes d'élimination et de rétention sont intéressantes qualitativement, mais c'est sous la forme d'activité cumulée que les données sont
FIG. 2. Comparaison des fractions de tritium à longue période de rétention trouvées après administration unique (notre étude) et continue (Thompson et Ballou [5]) d'eau tritée.

utilisées de façon pratique. La figure 3 indique les activités cumulées par gramme de tissu frais de divers organes après contamination par l'eau tritée, la thymidine par ingestion, la thymidine par injection IP, l'acétate de sodium et l'acide oléique par ingestion.

Le maximum de chaque valeur en ordonnée représente l'activité cumulée totale du 4ème jour au 60ème jour environ, en microcurie/jour, après administration de 1 mCi par rat de 300 grammes. Pour chaque organe, la partie inférieure représente ce qui revient à l'eau tritée présente et la partie supérieure ce qui revient à la fraction fixée. Du fait de l'importance des écarts il a fallu employer une échelle logarithmique des valeurs ; aussi l'importance relative des deux fractions n'est-elle malheureusement pas directement
FIG. 3. Activités cumulées par gramme de tissu frais depuis le quatrième jour. Pour chaque organe la partie inférieure correspond à l'eau, la partie supérieure à l'activité fixée.
proportionnelle à leur longueur. Cet inconvénient est en partie atténüé par la valeur assez constante de la fraction eau libre qui est voisine de 10 µCl/j ; seuls l’os et la graisse donnent des valeurs inférieures du fait de leur faible teneur en eau.

La figure 3 montre que la comparaison avec l’eau tritiée présente plus d’un avantage. Pour les autres molécules marquées, la fraction fixée dans certains organes n’est pas toujours très différente de la fraction fixée après administration d’eau tritiée (voir dans la thyroïde les valeurs obtenues avec l’acétate, la thymidine ingérée et l’eau tritiée) ; de plus, pour l’eau tritiée l’activité cumulée de la partie fixée varie d’un facteur 5 entre deux organes (testicules et moelle épinière).

Si l’activité cumulée était calculée depuis le moment de l’administration et non depuis le quatrième jour, la fraction eau atteindrait à peu près le double des valeurs qu’atteint l’eau libre après administration d’eau tritiée soit 20 à 25 µCl/j. L’activité cumulée de la fraction fixée augmenterait sans doute assez peu dans le cas de l’eau tritiée, mais plus fortement avec les autres molécules étudiées, en particulier pour les organes comme le foie qui ont une activité métabolique importante.
FIG. 5. Comparaison des activités cumulées par gramme de tissu frais après ingestion et injection IP de thymidine.
La figure 3 montre encore que l'activité cumulée de la fraction fixée des molécules marquées étudiées peut être bien plus grande que celle de la fraction eau dans l'espace de temps considéré.

Pour certains tissus comme le sang ou le foie, l'activité cumulée a pratiquement atteint sa valeur maximale bien avant la fin de la période d'étude (figure 4). Pour d'autres, comme la moelle épinière ou l'os, la limite peut être seulement calculée et sa valeur dépend des hypothèses retenues. Entre ces deux cas, l'examen des courbes de rétention et d'activité cumulée permet d'apprécier la valeur limite.

La figure 5 montre que le comportement de la thymidine n'est pas le même selon qu'elle est administrée par ingestion ou par injection intra-péritonéale. En portant sur le même graphique les activités cumulées respectives des tissus frais (figure 5) on constate que la plupart des organes des cavités abdominale et thoracique sont beaucoup plus contaminés après une injection IP. Pour les autres organes, le mode d'administration a beaucoup moins d'influence.

4. - CONCLUSION

Nous avons entrepris l'étude du métabolisme de molécules marquées dans un but de radioprotection. Afin de réserver les différentes possibilités d'interprétation et d'extrapolation à l'homme les rapports contiennent les informations de base sur la rétention et l'activité cumulée. L'expérimentation doit se poursuivre aussi longtemps que possible pour mieux apprécier l'importance de la composante à longue période. Il faut également distinguer la fraction eau de la fraction fixée, car chez l'homme les périodes retenues ne seront peut-être pas dans le même rapport que chez le rat.

Les exemples portant sur cinq molécules tritiées montrent que la rétention et l'activité cumulée pendant 500 jours ou plus varient selon l'organe, la nature de la molécule, le mode d'administration.

REFERENCES

(2) JEANNAIRE L., VERNOIS Y., BULLIER D., GROS R., Métabolisme de la thymidine tritiée chez le rat, Rapport CEA-R-4722 (1976).
DISCUSSION

S. PORTER: Can you expand a little on your statement that the long-term bound $^3$H varies with the age of the rat? Have you any similar human data?

L. JEANMAIRE: Long-term retention seems to decrease with the age of the rat, at least in the tissues of the central nervous system. This may be due to a diminution of metabolic activity. We have no data on humans.

V. KASCHE: Would not the non-homogeneous distribution of tritium a long time after exposure have an influence on the accumulated dose?

L. JEANMAIRE: It is indeed probable that the non-uniform distribution of the tritium would have an effect on dose, but we have not investigated this point. We calculated only the activity accumulated per gram of tissue.

A.A. MOGHISSI: I would like to congratulate the authors on providing us with data that are fundamental for an understanding of tritium dosimetry. I hope they will be able to continue their work.

L. JEANMAIRE: Thank you.
RADIATION DOSIMETRY IN EXPERIMENTAL ANIMALS EXPOSED TO TRITIATED WATER UNDER DIFFERENT CONDITIONS*

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United States of America

Abstract

RADIATION DOSIMETRY IN EXPERIMENTAL ANIMALS EXPOSED TO TRITIATED WATER UNDER DIFFERENT CONDITIONS.

The radiation dose to the germ cells of male mice, which sired the offspring scored in a specific-locus mutation test of injected tritiated water, has been calculated. The weighted mean dose for germ cells which received all of the radiation at post spermatogonial stages was 430 rad, while that for germ cells irradiated almost entirely as spermatogonia was 615 rad. Most of the animals received a single intraperitoneal injection of HTO of 0.50 mCi/g body weight, but a few of them received 0.75 mCi/g. These weighted mean doses reflect the contribution of both groups. The percentage of the total dose delivered from $^3$H incorporated into macromolecules is small — less than 0.5%. The percentage of the total radioactivity in dry material from the testis on day 1 post injection is 0.6%, on day 7 it is 3.2%, and on day 605 it is 95%. Tritium is incorporated into testicular DNA from tritiated water, and peak levels of radioactivity in this macromolecule are reached from three to nine days following injection. The stable incorporation of tritium into trichloroacetic acid insoluble materials is about 75% in protein and 25% in nucleic acids at all time periods following injection. Doses from single injections of tritiated water are inherently more variable than for protracted low-level exposures. This is because small differences in water balance near the time of injection can make a very large difference in total radiation dose.

INTRODUCTION

Human exposure to environmental tritium would be expected to be largely from tritiated water at low levels for protracted periods. In obtaining quantitative biological data for use in the calculation of potential risk to human populations, it is frequently necessary to choose other exposure conditions for the treatment of the experimental animal models. It is the purpose of this report to explore one such set of conditions, the single intraperitoneal injection of

* Research sponsored by the Division of Biomedical and Environmental Research,
relatively high levels of HT0, and to examine the nature and magnitude of errors which could result from departing from chronic low-level exposure in experiments using this technique.

The specific-locus mutation test in mice [1] provides the best currently available method of obtaining quantitative transmitted gene mutation data in an intact mammal. However, this method, or any method designed to obtain such data, requires the examination of relatively large numbers of offspring of individuals exposed to doses which are high compared to the expected environmental exposure. In addition, it is useful to perform the experiment in such a way that effects due to exposure of different germ-cell stages can be distinguished. The only type of exposure with which one can practically achieve these objectives for a specific-locus test in mice is that of the single injection of HT0.

For any type of exposure conditions the total dose is a function of the product of the average radiation intensity (dose rate) and time. For tritiated water the time-intensity curves are very different in shape for single exposure and chronic exposure conditions. In principle the areas under both types of curves could be integrated and total dose to any germ-cell stage calculated. These total dose figures would be strictly comparable with each other only if there is complete reciprocity for time and intensity, i.e., that biological effect per unit of dose is independent of dose rate over the range of dose rates tested. The highest dose rates obtained by injecting 0.50 mCi/g body weight of tritiated water are below the levels where dose rate effects are seen in males from other types of radiation exposure in the specific-locus test. Thus dose rate, per se, is not likely to produce major differences in results between the two types of experimental protocol.

There are, however, other complicating factors in comparing chronic vs. acute exposures to tritiated water. One such factor is the possibility that for the two sets of exposure conditions, different proportions of the total dose may be from tritium in different molecular sites. Another is that the precision of dose calculations for acute exposures is inherently less precise than that for chronic exposure conditions.

Differences of this type between chronic and acute exposure conditions must be effectively dealt with to achieve a realistic assessment of the environmental impact of man-made tritium release. The dosimetry of biologically significant radiation from exposure to tritiated water is extremely complex. However, it is hoped that useful values can be obtained for the purposes of risk calculations.

Preliminary accounts of this work have appeared in the Annual Reports of the Biology Division of the Oak Ridge National Laboratory [2, 3, 4].

METHODS

Exposure conditions

Male mice, strain (101 x C3H)F1, were individually weighed and injected intraperitoneally with tritiated water which had been adjusted in specific activity to 25 mCi/ml. Each mouse received a single injection of 20 μCi/g of body weight to give a total injected radioactivity of 0.50 mCi/g or 500 mCi/kg. Eighteen male mice were injected in each group at about 10:00 a.m., and each male was placed in a disposable plastic mouse cage with 5 uninjected T-stock female mice. The cage was immediately placed in an isolator (described below) designed to minimize atmospheric contamination in the animal facility. Exposure conditions for dosimetry were identical to those for the specific-locus mutation experiments reported elsewhere [5], and some of the animals in the dosimetry experiments produced offspring scored for specific-locus mutations and included in that report.
Isolators

For two weeks after injection the animals were housed in isolators designed to recover and measure HTO released into the atmosphere surrounding their cages. The isolators were made of a heavy flexible plastic with a rigid internal aluminum frame to maintain constant volume. These isolators were 61 x 61 x 152 cm in size and had an internal volume of 565 l. Each contained 18 disposable mouse cages and enough food and water to maintain the mice for one week. Each isolator was fitted with 2 pairs of heavy rubber gloves, which allowed the operator to reach any part of the internal space without opening the isolator. Isolators were operated at negative pressure relative to the room. Air was pulled from the isolator through a series of four cold traps, which removed >99% of the moisture it originally contained. It was then pulled through a large drying filter and released through a hood. Traps were removed at intervals and the volume and specific activity of the water they contained was measured. Air flow was maintained at about 1 isolator volume every 4 min. Animals were maintained in the isolators for two weeks after injection and then were removed into a special mouse room reserved for that purpose.

Experimental protocol

Animals were injected as described above and then serially sacrificed so that the quantity and distribution of radioactivity remaining in the testes could be measured. The postinjection serial sacrifice time points for this experiment were: 1 hr, 12 hrs, ± 10 days (±1-day increments), 10 to 100 days (±5-day increments), and 605 days. Nine animals were sacrificed for each time point to give an estimate of individual variability.

Tissue preparation

The testes of each treated animal were homogenized in distilled water and brought to a constant volume (10 ml). Aliquots of these homogenates were solubilized wet or dried under a vacuum on filter paper and counted for radioactivity in a liquid scintillation spectrophotometer. Details of tissue handling procedures are as described previously [6]. Dried aliquots were oxidized by combustion prior to counting to obtain optimum samples for the determination of absolute activity. DNA and protein were extracted from pooled extracts at each time point and the specific activity determined.

Calculation of dose

Mean total tissue specific radioactivity was measured at the various time points from one hour to 605 days postinjection. These points were joined using an exponential extrapolation between points to generate a radioactivity time curve for the single exposure. Areas under this curve were determined by integration to give radiation dose estimates to germ cells at various injection-to-conception intervals. These doses were weighted for the number of offspring sired in each interval to give weighted mean doses in the specific-locus experiments for offspring resulting from germ cells irradiated only in postpubescent stages and offspring resulting from conceptions involving germ cells which were irradiated primarily as spermatogonia.

RESULTS AND DISCUSSION

An estimate of the radiation dose to the testes of mice involved in a specific-locus mutation experiment [5] has been calculated. This estimate was based on data obtained from mice injected with 0.50 mCi/g of HTO and handled and
TABLE I. SUMMARY OF DATA ON RADIATION DOSE TO THE TESTES OF MALE MICE FOLLOWING A SINGLE INJECTION OF 0.50 mCi/g OF HTO

<table>
<thead>
<tr>
<th>Time postinjection (days)</th>
<th>Tissue radioactivity (DPM/mg)</th>
<th>Dose rate (rads/hr)</th>
<th>Cumulative dose (rads)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 h</td>
<td>$13.8 \times 10^5$</td>
<td>7.23</td>
<td>—</td>
</tr>
<tr>
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<td>5.94</td>
<td>157.9</td>
</tr>
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<td>281.8</td>
</tr>
<tr>
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<tr>
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</tr>
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</tr>
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<td>576.7</td>
</tr>
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<td>$2.7 \times 10^{-2}$</td>
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<tr>
<td>605</td>
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<td>$2.3 \times 10^{-4}$</td>
<td>594.6</td>
</tr>
</tbody>
</table>

...mated identically to those in the specific-locus experiment. The tissue specific activity of the testes was monitored through time by serial sacrifice and liquid scintillation counting of tissue homogenates. A summary of some of the data obtained is presented in Table I. By exponential extrapolation between points a curve is generated and the area under this curve can be integrated to give the dose to any point in time after injection. The upper limit of the dose for these conditions and with this quantity of radioactivity is just under 600 rads. The animals injected with 0.75 mCi/g in the specific-locus experiment are assumed to receive 1.3 times the dose received by animals given 0.5 mCi/g at any equivalent time point. Using these values to calculate weighted mean doses to germ cells which produced the over 40,000 offspring scored for the specific-locus test [5], a value of 430 rads is obtained for germ cells irradiated in post spermatogonial stages and of 615 rads for spermatogonia.
FIG. 1. Specific activity of DNA extracted from the testes of mice as a function of time after a single injection of 0.50 mCi/g of tritiated water.

When aliquots of testis homogenates are counted both wet and after drying under a vacuum, one can obtain an estimate of the percentage of the radioactivity present in the form of water. The radioactivity which remains after such drying procedure is assumed to be incorporated into some nonvolatile molecule. Only 0.6% of the radioactivity remains after drying on samples taken 24 hours after injection, while 3.2% is stable to drying in samples taken 7 days after injection. In samples taken 605 days after injection of HTO, 95% of the remaining activity is in nonvolatile molecules. This nonvolatile radioactivity forms the upper limit for tritium incorporated into macromolecules (proteins and nucleic acids). When nonvolatile radioactivity is integrated with time after injection, less than 0.5% of the total dose comes from tritium in this nonvolatile form. Thus a very small fraction of the total radiation dose comes from tritium incorporated into macromolecules.

In a procedure described earlier [6] it is possible to get an estimate of the ratio of $^3$H incorporated into protein to that incorporated into nucleic acids. For all time periods following HTO injection this ratio is about 3 in testis homogenates. This indicates that about 75% of the total macromolecular incorporation is in protein and the other 25% in nucleic acids (both DNA and RNA).

If DNA is extracted from testis homogenates of animals sacrificed at various times following the injection of HTO, tritium incorporation into this macromolecule is readily measured. There is a broad peak of activity in DNA from 3 to 9 days postinjection. (See Fig. 1.) After this time the specific activity of DNA decreases, presumably due to dilution from new DNA synthesis incorporating less tritium. The fraction of the total radiation dose received from tritium incorporated into DNA is extremely small (<0.1% of the total).

We have a number of indications that total radiation dose from a single injection of HTO can be greatly influenced by rather minor changes in the experimental conditions. Water intake during the first few weeks following injection is clearly very important. It is also clear that the quantity and quality of the food consumed plays a role as does the number of animals housed in a single cage. Low-level chronic exposure conditions, such as those used by Carsten and Commerford [7] would be expected to be less subject to experimental variability.
This is because the specific activity of the administered water remains constant and fluctuations in daily intake are relatively unimportant. The chronic low level exposures also provide a better model for the exposure conditions which humans are likely to encounter. However, the single injection protocol does make practical large genetic experiments such as the specific-locus test and thus yields vital information.

REFERENCES


DISCUSSION

R.V. OSBORNE: In view of the great influence which even minor changes in the experimental conditions have on the radiation dose, what confidence limits would you place on your dose estimates?

R.B. CUMMING: Somewhere in the 30% range. The coefficient of variation for individual time points increases with the lapse of time after injection, i.e. the confidence limits on the early time points are relatively much better, and of course these early time points have very much more of the total radiation dose. Thus, as we go out to the 605-day point the coefficient of variation is relatively much greater, about 100%, but we are then dealing with very small amounts of activity.
TRITIUM DISTRIBUTION AND INCORPORATION FROM TRITIATED WATER OR TRITIATED PRECURSORS OF DNA, RNA OR PROTEINS

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Abstract

TRITIUM DISTRIBUTION AND INCORPORATION FROM TRITIATED WATER OR TRITIATED PRECURSORS OF DNA, RNA OR PROTEINS.

Following administration of tritiated thymidine to newborn C57 Black/6M mice in the dose range of 0.3 to 1.5 μCi/g of body weight, an increased incidence of lymphosarcomas was observed in both sexes. It appeared that administration of tritiated thymidine to newborn mice resulted in a definite modulation of the tumour spectrum. This observation brought up the question as to whether tritiated precursors of RNA or proteins, namely tritiated uridine or leucine, or even tritiated water, would possibly trigger a similar shift of the tumour spectrum in the C57 Black/6M mice. As a prerequisite to testing this hypothesis, an endeavour was made to investigate the comparative distribution of tritiated thymidine, uridine or leucine versus tritiated water, in mice under various experimental conditions. Newborn and juvenile C57 BL/6M mice were injected with 1 or 10 μCi/g of tritiated water, tritiated thymidine, tritiated uridine or tritiated leucine. Young adult mice were similarly injected. Animals were sacrificed after varying lengths of time. The data show that administration of tritiated precursors such as thymidine, uridine and leucine or of tritiated water results in a very complex time-dependent and, in some respects, age-dependent residual activity dynamics both in the organic component and in the aqueous fraction of tissue. Groups of juvenile male mice, 15 days old, were injected on three consecutive days with 100 μCi of HTO/g of body weight. A significant although small incorporation of tritium into DNA, RNA and proteins was observed in all organs tested under the experimental conditions. In addition, comparative dose estimates were made from tritiated thymidine or tritiated water in the testis.

1. INTRODUCTION

Tritium is classically considered a relatively innocuous radionuclide on account of its low beta energy and because it is generally assumed to hold a homogeneous distribution throughout tissues, mostly in the form of tritiated water. The disappearance of tritium from body water in mammals is relatively rapid; the half-life of tritiated water is about 1.5 days in the mouse and 9.7 days in man. Long-term studies, however, have shown that the overall radioactivity
FIG. 1. Tritium activity in free water pool following a single administration of HTO (1 or 10 μCi/g) to newborn (NB), juvenile (JUV), or adult (AD) mice.

FIG. 2. Tritium activity in organic component following a single administration of HTO (1 or 10 μCi/g) to newborn (NB) or juvenile (JUV) mice.
FIG. 3. Tritium activity in organic component following a single administration of HTO (1 or 10 μCi/g) to adult (AD) male or female mice.

disappears according to a complex kinetic pattern. In man the excretion curve can be approximated using three exponentials, with half-times of disappearance of approximately 6, 23 and 35 days (1).

The NCRP-ICRP have set the maximum permissible body tritium for occupational exposure at 1,000 μCi with body tissue as the critical organ (2, 3). Assuming a uniform distribution of tritium in the human body, the equivalent dose rate absorbed, as calculated for a standard man with a 70 kg mass, is short of 100 mrem/week, in accordance with the recommended maximum permissible occupational exposure of 5 rem/year. In 1970 the annual dose from tritium in the biosphere was estimated to be 0.04 mrem/person whereas the concentrations of tritium in the oceans and ground water in the United States typically ranged from 0.2 to 1.5 nCi/liter (4). At equilibrium in a steady state, the tritium to hydrogen ratio in man's body water is generally assumed to approach the same value as in the environmental water. There is some indication, however, that the actual ratio may be lower, i.e., of the order of 0.40 (5).

Most tritium, as well as hydrogen, that is incorporated into the living tissues originates from the hydrogen that is organically bound in food. It was found (6) that tritium bound to the organic fractions of cow's milk, largely in the milk fat, was 10 times higher in cows fed grass with organically bound tritium than in those given equal amounts in their drinking water. The transfer of tritium from tritiated
FIG. 4. Tritium activity in free water pool following a single administration of HTO, tritiated uridine, thymidine or leucine (1 μCi/g or 10 μCi/g) to newborn mice.

water is largely by exchange with the labile hydrogens of organic fractions after the new tritiated water has mixed with the existent body water. Carbon bound hydrogens are very stable and it requires enzymatic processes to produce a limited exchange between them and the hydrogens of free body water (7). In a study with mice given daily injections of tritiated water for 14 days, 12 was found in organically bound form and there was but 12% in all of the body presumably as free body water. Except for blood, where tritium was largely as free water, all tissues had only a small amount of tritium that was largely labile in the organic form (8). In rats and mice exposed to tritiated water for extended periods, the highest concentrations were in the brain lipids (9) followed by the skin and muscle (10).

Much of the present guidelines are based on the assumption of a homogeneous distribution for the absorbed dose from tritium in living tissues. It has been argued that "a given dose of radiation to an organ from the beta rays of tritium has the same radiobiological meaning as the given dose from x-rays or gamma rays (same dose pattern), and no significant added hazard is to be attached by virtue of the fact that dose may have been derived from tritium" (11). This view is well substantiated by numerous experimental data related to biological effects of tritium within the medium dose range, at the cellular level. The question remains, however, as to whether the same conclusions are valid in the low dose range, especially at the level at which radiation protection recommendations are applicable.
FIG. 5. Tritium activity in organic component versus free water pool following a single administration of HTO (1 μCi/g) to newborn (NB), juvenile (JUV) or adult (AD) mice.

The validity of currently accepted dose estimates are predicated on the assumption that the mean range of the particle is small compared with the dimensions of the irradiated target, an essential prerequisite to microscopic physical homogeneity of the dose distribution. This concept becomes less and less acceptable with decreasing dose rates, as microsites of energy absorption tend to cluster up. Furthermore, as pointed out in a recent Brookhaven report (12) there are also extremely important factors of microscopic biological heterogeneity due to possible differences in tritium concentration in particular sub-cellular components (nucleus) or even molecular components (nucleic acids).

Our interest in tritium distribution and incorporation was triggered by the investigation of a possible carcinogenic effect of tritium at low dosage in mammals. Following the pioneer work of Lisboa, Baserga and Kitieński (13), tritiated thymidine was further tested (Mewissen, 14) for potential carcinogenic action. Following administration of tritiated thymidine to newborn C57 Black/6H mice in the dose range of 0.3 to 1.5 μCi/g of body weight, an increased incidence of lymphosarcoma was observed in both sexes. It appeared that administration of tritiated thymidine to newborn mice resulted in a definite modulation of the tumor spectrum. This observation brought up the question as to whether tritiated precursors of RNA or proteins, namely
FIG. 6. Tritium activity in the organic component versus free water pool following a single administration of either HTO or thymidine (1 μCi/g) to newborn (NB) or juvenile (JUV) mice.

tritiated uridine or leucine, or even tritiated water would possibly trigger a similar shift in the tumor spectrum in our C57 Black/6M mice. As a prerequisite to testing this hypothesis, we endeavored to investigate the comparative distribution of tritiated thymidine, uridine or leucine versus tritiated water, in those mice under various experimental conditions. Data available to date are presented in this report.

2. COMPARATIVE DISTRIBUTION OF TRITIATED WATER VERSUS TRITIATED THYMIDINE, URIDINE OR LEUCINE

a. Materials and methods

Newborn (less than 24 hours old) and juvenile (2 weeks old) C57 BL/6M mice were injected subcutaneously or intraperitoneally with 1 or 10 μCi/g of tritiated water, tritiated thymidine or tritiated leucine. Animals were sacrificed after 24 hours, 8, 15, 22 or 29 days.

Young adult mice (aged 3 months) were similarly injected and sacrificed. Tritium incorporation was assessed both in the aqueous fraction and in the organic component of tissues from various organs, including the thymus, spleen, lungs, liver, brain and kidneys. The remaining organs and the carcass were pooled for
similar determination. The tritium assay was carried out by dehydration and subsequent oxidation of the tissue residue in a Packard Tri-Carb Oxidizer according to a procedure involving several steps, including progressive dehydration, multiple flushing of the activity memory, dehydration of the blank filter paper, reflushing of the activity memory, and final oxidative combustion of the sample. This procedure is roughly equivalent to lyophilization of living tissue. Our aim was to calculate the relative specific activity (RSA) of tritium in the organic component, i.e. the fraction of organically bound tritium per hydrogen content of dry tissue compared to the aqueous tritium per hydrogen content of water present in tissue.

All experiments were duplicated at the 1 μCi and 10 μCi/g levels in an effort to cross-check the data and assess the reproducibility of distribution patterns, as very small amounts of residual activity were present in tissue extracts following administration of 1 μCi of tritiated water or tritiated precursors. A significant difference in the pattern of leucine incorporation between newborn and juvenile animals was observed, in contradistinction to incorporation patterns for uridine and thymidine. Therefore, this particular experiment was repeated. Duplicate data yielded comparable results.

b. Results

Pooled data for whole animals are presented in Figures 1 to 6.

It can be seen in Figure 1 that, following administration of tritiated water (HTO), free-water tritium (F.W.) activity decreased almost exponentially with time in all animals. Residual activity patterns were similar at both activity levels tested (10 and 1 μCi/g). Data on adult animals were separated by sexes. In juvenile and newborn animals, however, animals of both sexes were pooled, as separation at an early age was impractical at the start of the experiment. No significant age or sex related difference between animals was apparent.

In Figure 2, the residual activity of organically bound tritium (O.B.) was plotted as a function of time. Residual activity patterns were similar at the two activity levels. The striking feature was that activity levels were consistently higher in juvenile than in newborn animals.

The residual activity levels of organically bound tritium were consistently higher in male than in female adult animals (Figure 3) at both activity levels tested.

In Figure 4, the respective residual activity levels were compared following administration of tritiated water, tritiated thymidine, tritiated uridine or tritiated leucine. Tritiated precursors generated equivalent activity levels, but diverged markedly from the lower levels generated by tritiated water.

In Figure 5, F.W. activity was compared with O.B. activity following administration of HTO to adult, juvenile and newborn mice. As observed previously, F.W. activities were similar, irrespective of age or sex. The O.B. activity progressively overtook the F.W. activity with increasing time. Whereas O.B. levels, as already noticed, were higher in juvenile (JUV) than in newborn (NB) animals, intermediate values were observed in adults (AD), with those for males higher than in females.

In Figure 6, F.W. activity was compared with O.B. activity in NB or JUV mice following administration of HTO or tritiated thymidine. As expected, tritiated precursors yielded much higher O.B. values than did HTO. Also, F.W. activity from tritiated thymidine progressively overtook F.W. activity from HTO, in adult, juvenile and newborn mice. As observed previously, F.W. activities were similar, irrespective of age or sex. The O.B. activity progressively overtook the F.W. activity with increasing time. Whereas O.B. levels, as already noticed, were higher in juvenile (JUV) than in newborn (NB) animals, intermediate values were observed in adults (AD), with those for males higher than in females.

A summary of the data pertaining to selected organs is presented in Tables 1 to IV.

F.W. refers to free water activity, i.e. in the aqueous fraction, O.B. refers to activity in the organic component, i.e. organically bound activity.
<table>
<thead>
<tr>
<th>Day</th>
<th>Thymus</th>
<th>Spleen</th>
<th>Lung</th>
<th>Liver</th>
<th>Brain</th>
<th>Kidney</th>
<th>All Organs</th>
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<tbody>
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<td>1</td>
<td>4 131</td>
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<td>3 463</td>
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<td>Juvenile</td>
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<td>0.6</td>
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<td>0.7</td>
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<tr>
<td>Adult</td>
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<td>5 566</td>
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TABLE II. RESIDUAL ACTIVITY (nCi/g) AND RSA VALUES FOLLOWING ADMINISTRATION OF TRITIATED THYMIDINE (10 μCi/g)

<table>
<thead>
<tr>
<th>Day</th>
<th>Thymus</th>
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<th>Lung</th>
<th>Liver</th>
<th>Brain</th>
<th>Kidney</th>
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<tbody>
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<td>36 447</td>
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### TABLE III. RESIDUAL ACTIVITY (nCi/g) AND RSA VALUES FOLLOWING ADMINISTRATION OF TRITIATED URIDINE (10 μCi/g)

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<tr>
<th>Days</th>
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<th>All Organs</th>
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<td>O.B.</td>
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<td>190</td>
<td>15.9</td>
<td>19</td>
<td>556</td>
<td>47.2</td>
</tr>
<tr>
<td>Adult</td>
<td>1</td>
<td>4039</td>
<td>8.610</td>
<td>3.1</td>
<td>5804</td>
<td>37.681</td>
<td>10.4</td>
</tr>
<tr>
<td></td>
<td>29</td>
<td>167</td>
<td>807</td>
<td>7.7</td>
<td>140</td>
<td>1741</td>
<td>19.7</td>
</tr>
<tr>
<td>Adult</td>
<td>1</td>
<td>3271</td>
<td>5.993</td>
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<td>4306</td>
<td>36.217</td>
<td>13.3</td>
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<tr>
<td></td>
<td>29</td>
<td>90</td>
<td>429</td>
<td>7.5</td>
<td>131</td>
<td>1403</td>
<td>16.9</td>
</tr>
</tbody>
</table>
RSA refers to relative specific activity as the ratio of specific activity of organically bound tritium to aqueous tritium, corrected for relative abundance of hydrogen in either pool.

As far as residual activity is concerned, in both the aqueous (F.W.) and non-aqueous (O.B.) fractions, thymidine and leucine followed a similar pattern of disappearance in all organs tested at the 1 μCi as well as the 10 μCi level. The reproducibility was satisfactory even at the lower dose level. In thymus, spleen and liver, incorporation and utilization appeared quite comparable with respect to the time-dependent rate of disappearance of residual activity. The specific rates for the organic component differed from those pertaining to the aqueous fraction by a factor of 10. Moreover, these rates were at any particular time significantly lower in the organic component than in the aqueous fraction, with the result that the rate difference increased from a ten-fold to a fifteenfold margin.

For lungs, kidneys and brain, the rates of residual activity in the organic component decreased much more slowly with time, and the rate difference from the aqueous fraction increased far more to attain a hundredfold divergence. These findings were common to both newborn and juvenile animals.

The time-dependent rates of disappearance of residual activity for thymidine and for uridine followed a closely similar pattern in both newborn and juvenile animals, with residual rates being about three times higher for thymidine than uridine. The specific rates for leucine were unique in one respect: in newborn animals, they closely paralleled the uridine rates; in juvenile males, however, they approached the thymidine rates.

Values of RSA were found to be time-dependent for all precursors, but the patterns varied widely. Typically, the observed values increased more rapidly for juveniles than for newborn animals. Time rates of increase were moderate for leucine and thymidine and very low for uridine.

Following a single administration of tritiated water, at the 1 or 10 μCi level, the rates of residual activity were significantly lower for the organic component than for the aqueous fraction, a phenomenon that, in our opinion, was the expression of a significantly, although transient, incorporation. It is tempting to postulate a steady rate of replacement of exchangeable hydrogens with available tritium atoms, resulting in a proportionally increased fraction of tritium atoms being trapped in the organic component.

c. Discussion of the results

From the data available at this time, the following conclusions may be drawn.

The F.W. component was essentially equivalent in all animals, irrespective of age and sex, following administration of tritiated water. The O.B. component, however, was proportionately higher respectively in juveniles, male adults, female adults and newborns, suggesting that the rate of incorporation and/or utilization varied significantly with age and sex.

Following administration of tritiated precursors, tritium was transferred into the F.W. pool at a higher rate than following administration of tritiated water, again suggesting a steady rate of exchange between the O.B. compartment and the F.W. pool.

Administration of tritiated water resulted in definite organification of tritium. Moreover, the time-dependent rates for the residual activity of the O.B. fraction from HTO closely paralleled those for the O.B. fraction from tritiated thymidine by a factor which increased from about 10 after 24 hours to about 15 after 28 days.

The data show that administration of tritiated precursors such as thymidine, uridine and leucine or of tritiated water result in a very complex time-dependent and, in some respects, age-dependent residual activity dynamics both in the organic component and in the aqueous fraction of tissue. In view of our findings, it would seem that a realistic estimate of the internal dose to the radiosensitive nucleus ought to take into consideration the time-dependent rate of residual activity that reflects the dynamics of tritium resulting
<table>
<thead>
<tr>
<th>Day</th>
<th>Thymus</th>
<th></th>
<th>Spleen</th>
<th></th>
<th>Lung</th>
<th></th>
<th>Liver</th>
<th></th>
<th>Brain</th>
<th></th>
<th>Kidney</th>
<th></th>
<th>All Organs</th>
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<tr>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Newborn</td>
<td>1</td>
<td>794</td>
<td>10</td>
<td>318</td>
<td>20.6</td>
<td>790</td>
<td>18</td>
<td>549</td>
<td>37.3</td>
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<td>11</td>
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<td>343</td>
<td>21.3</td>
<td>31</td>
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<td>29.1</td>
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<td>668</td>
<td>41.9</td>
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<tr>
<td>Juvenile</td>
<td>1</td>
<td>1,681</td>
<td>21</td>
<td>335</td>
<td>18.0</td>
<td>3,280</td>
<td>38</td>
<td>170</td>
<td>32.8</td>
<td>1,952</td>
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<td>832</td>
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<td></td>
<td>29</td>
<td>24</td>
<td>609</td>
<td>40.3</td>
<td>34</td>
<td>1,846</td>
<td>88.1</td>
<td>28</td>
<td>1,696</td>
<td>95.0</td>
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<tr>
<td>Adult</td>
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<td>3,759</td>
<td>15</td>
<td>951</td>
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<td>707</td>
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<td>7.1</td>
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<td>39</td>
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<td>492</td>
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<td>135</td>
<td>7.3</td>
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<tr>
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<td>449</td>
<td>21.7</td>
<td>23</td>
<td>711</td>
<td>46.6</td>
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<td>1,127</td>
<td>57.7</td>
<td>38</td>
<td>542</td>
<td>22.7</td>
</tr>
</tbody>
</table>
TABLE V. CRUDE SPECIFIC ACTIVITY (DPM/μg) FOLLOWING HTO (3 X 100 μCi/g OF BODY WEIGHT)

<table>
<thead>
<tr>
<th></th>
<th>DNA</th>
<th>RNA</th>
<th>PROTEIN</th>
</tr>
</thead>
<tbody>
<tr>
<td>Day</td>
<td>1</td>
<td>14</td>
<td>35</td>
</tr>
<tr>
<td>Thymus</td>
<td>7.8</td>
<td>1.7</td>
<td>.17</td>
</tr>
<tr>
<td>Spleen</td>
<td>14.8</td>
<td>2.1</td>
<td>.63</td>
</tr>
<tr>
<td>Testes</td>
<td>4.1</td>
<td>2.8</td>
<td>.44</td>
</tr>
<tr>
<td>Lung</td>
<td>2.8</td>
<td>1.4</td>
<td>.50</td>
</tr>
<tr>
<td>Liver</td>
<td>4.8</td>
<td>2.2</td>
<td>.58</td>
</tr>
<tr>
<td>Mean</td>
<td>6.9</td>
<td>2.0</td>
<td>.46</td>
</tr>
</tbody>
</table>

from the degree and site of incorporation, and from the rate of replacement of exchange hydrogens in labelled macromolecules and in catabolites.

The RNA values extend from 0.5 to short of 10 following administration of tritiated water, but absolute values of activity are exceedingly small. After a 28-day interval, for example, they show a thousandfold decrease.

3. INCORPORATION OF TRITIUM INTO DNA, RNA AND PROTEINS

a. Materials and methods

Groups of juvenile male mice, 15 days of age, were injected on three consecutive days with 100 μCi of HTO/g of body weight. The animals were sacrificed by cervical disruption, respectively at a 24-hour interval following the last exposure, then at 3, 5, 7, 9, 14, 21, 28 and 35-day interval. The mice were autopsied and the dissected organs were rapidly frozen on dry ice. Selected organs were saved for further analysis: thymus, spleen, testes, lung and liver. DNA was isolated by the method of Kay et al., RNA by the method of Torelli and the protein fraction was determined with the Buerstedt reaction.

Respective tissues were homogenized in ice cold buffer of 0.9% NaCl, 0.01 M Na citrate, 0.01 M EDTA pH 5.2, centrifuged for 30 minutes at 0°C and 2 500 RPM (750 x g). This step was repeated with sediment three times. The last sediment was resuspended in 0.9% NaCl, to which was added 5% SDS solution in 45% ethanol to final concentration 0.4%. The solution was stirred three hours at room temperature and solid NaCl to final concentration 1 M was added. The final solution was spun for thirty minutes at 4°C and 13 000 RPM. The supernatant was decanted, chilled, and the DNA fiber was wound on a glass rod after the addition of two volumes of ice cold absolute alcohol. Following three successive washings in 95% alcohol and two washings in ether, the DNA fiber was dissolved in distilled water. RNAase 400 μg/ml and pronase 50 μg/ml and EDTA was added to a final concentration of 0.005 M then incubated for thirty minutes at room temperature and dialysed overnight against 0.01 M NaCl at room temperature. DNA was quantitatively determined by spectrophotometer at 260 mm wave-length and aliquots were taken for determination of radioactivity in a Packard liquid scintillation spectrophotometer equipped with absolute activity analyzer. These data served for the calculation of specific activity in terms of DPM/μg of DNA. The combined supernatants were treated with 5% SDS to a final concentration of 0.5%, vigorously shaken for 30 seconds and,
after addition of one volume of phenol-m-cresol 7:9:1 containing 0.12 8-hydroxy-quinoline, incubated 3 minutes at 65°C, rapidly cooled in dry ice-alcohol mixture and centrifuged 10 minutes at 4°C and 11 000 RPM. The aqueous phase was re-extracted with hot phenol-m-cresol containing 0.5% SDS. It was spun at 11 000 RPM, for ten minutes at 4°C. The aqueous phase was washed three times with ether. Two volumes of alcohol-m-cresol 9:1 with one volume of K acetate was added. The mixture was frozen overnight, centrifuged at 15 000 RPM for 20 minutes at 0°C.

The precipitate of RNA was washed three times with 75% alcohol and dissolved in 0.01 M tris buffer at pH 7.4 DNA-ase was added 10 ug/ml, pronase 50 ug/ml and MgCl₂ to final concentration 0.001 M. The mixture was incubated for thirty minutes at room temperature and dialyzed against 0.01 M NaCl overnight. The amount of RNA was measured by spectrophotometer at 260 nm wave-length and aliquots taken for counting of radioactivity. The specific activity was assessed in terms of DPM/g of RNA.

Protein was obtained from the sediment after removal of DNA. It was washed once with methanol, twice with methanol-ether 1:1, three times with ethanol, once with ethanol-ether 1:1 and finally three times with ether. It was incubated for 1 hour in 5% TCA, washed once with cold 5% TCA, dissolved in 0.5 N NaOH. The amount of protein was measured by the Biuret reaction and the residual radioactivity counted from the aliquots.
FIG. 8. Residual tritium activity (thymus) in isolated DNA, RNA and protein following injection of tritiated water in juvenile mice.

b. Results

Data available at that time are summarized in Table V and Figures 7 and 8. It is seen that incorporation of tritium into DNA, RNA and protein is small but significant in all organs tested under our experimental conditions.

COMPARATIVE DOSE ESTIMATES FROM TRITIATED THYMIDINE OR TRITIATED WATER IN THE TESTIS

a. Materials and methods

Young adults, male and female (35 days old), C57 BL/6M mice were injected intraperitoneally with 1 pCi/g of body weight of tritiated thymidine. Animals were sacrificed after 24 hours, 7, 14, 21, 28 and 35 days. Another group of young adults, male and female (35 days old), C 57 BL/6M mice were drinking water (10 uCi/ml) for a maximum of 35 days. Animals were sacrificed after 24 hours, 7, 14, 21, 28 and 35 days. Tritium incorporation was assessed both in the aqueous and in the organic component of tissues from various organs, including the thymus, spleen, lungs, liver, brain, kidney, ovaries or testes, carcass and small intestine. The tritium assay was carried out by dehydration and subsequent oxidation of the tissue residue in a Packard Tri-Carb oxidizer according to a procedure involving several successive steps, including progressive dehydration, multiple flushing of the memory activity, and final oxidative combustion of the sample.
TABLE VI. RESIDUAL ACTIVITY IN TESTES FOLLOWING TRITIATED THYMIDINE (1 μCi/g BODY WEIGHT)

<table>
<thead>
<tr>
<th>Number of Animals</th>
<th>Survival (days)</th>
<th>MEAN ACTIVITY</th>
<th>TOTAL ACTIVITY</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>F.W. a</td>
<td>O.B. b</td>
</tr>
<tr>
<td>4</td>
<td>1</td>
<td>194</td>
<td>1046</td>
</tr>
<tr>
<td>5</td>
<td>9</td>
<td>25</td>
<td>717</td>
</tr>
<tr>
<td>5</td>
<td>18</td>
<td>4.7</td>
<td>363</td>
</tr>
<tr>
<td>3</td>
<td>27</td>
<td>3.7</td>
<td>181</td>
</tr>
<tr>
<td>4</td>
<td>35</td>
<td>3.5</td>
<td>77</td>
</tr>
</tbody>
</table>

a F.W.: Free water tritium  
b O.B.: Organically bound tritium  
R.S.A.: Relative specific activity as the ratio of specific activity of organically bound tritium to aqueous tritium, corrected for relative abundance of hydrogen in either pool.

TABLE VII. RESIDUAL ACTIVITY IN TESTES OF MICE DRINKING TRITIATED WATER (10 μCi/ml)

<table>
<thead>
<tr>
<th>Number of Animals</th>
<th>Survival (days)</th>
<th>MEAN ACTIVITY</th>
<th>TOTAL ACTIVITY</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>F.W. a</td>
<td>O.B. b</td>
</tr>
<tr>
<td>5</td>
<td>1</td>
<td>2782</td>
<td>1980</td>
</tr>
<tr>
<td>6</td>
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<td>4426</td>
</tr>
<tr>
<td>3</td>
<td>35</td>
<td>5695</td>
<td>4642</td>
</tr>
</tbody>
</table>

a F.W.: Free water tritium  
b O.B.: Organically bound tritium  
R.S.A.: Relative specific activity as the ratio of specific activity of organically bound tritium to aqueous tritium, corrected for relative abundance of hydrogen in either pool.
b. Results

Data available at that time are summarized in Table VI and VII. They were used to define our dose estimates from tritium to sperm in males exposed either to tritiated thymidine (1 μCi/g of body weight in a single injection) or tritiated water (as drinking water for 35 days at the concentration of 10 μCi/ml). The following assumptions were made in the calculation model:

- Tritiated water is homogeneously distributed in the tissue;
- Nucleus and cytoplasm contain 75 percent water and radioactivity per mass is 75 percent of total activity;
- In spermatocyte, the volume ratio cell/nucleus is about 13.4, assuming an average cellular diameter of 10.0 μ and an average nuclear diameter of 4.2 μ;
- Tritiated thymidine is predominantly incorporated into the nucleus with an edge effect of 0.8.

From our data the dose to male sperm over a 35-day period was estimated at 3.7 rad from tritiated water and at 3.9 rad from tritiated thymidine under our experimental conditions.

5. DISCUSSION

A soft beta emitter with an estimated R.B.E. equal to unity, tritium in the form of tritiated water has been assigned one of the highest maximum permissible body burden values (1,000 μCi) for any radioisotope by the I.C.R.P. When maximum permissible dose results from internal body irradiation, the I.C.R.P. recommendations imply that, whenever possible, allowance should be made for non-uniform dose distribution of radionuclides within the tissues. Organically bound tritium, however, is not acknowledged in the computation of the maximum permissible dose for tritium recommended by the I.C.R.P. Assuming a 0.85 to 1.00 value for the ratio of organic tritium to body water tritium, Evans estimated (15), in the standard man, a body burden 1.4 to 1.5 times higher than predicted by the I.C.R.P. Martin and Koranda (16) found for kangaroo rats a correction factor of 1.4 in close agreement with Evans. Using a complex mathematical model, Petzer (17) was able to compute correction factors ranging from 1.11 to 1.24 for various rat organ tissues. Our data on short exposure of mice to tritiated water confirm previously reported values for the acute (briefly delivered) dose. For commitment dose, however, we found RBE values up to about 10. The implications of this finding on late effects will be further investigated by assessing the comparative carcinogenic potency of tritiated water in relation to various schedules of exposure.

The long-term toxicity of tritium in living tissues and particularly in mammals is largely unknown. There is ample experimental evidence that following intake of tritiated water, tritium becomes organically bound in a non-labile form. In this respect, our data on tritiated water merely confirm what has been known for more than twenty-five years (Thompson, 18). Organification of tritium in a tritiated ecosystem has been demonstrated in deer tissues (Evans, 15) from the Savannah River Plant Site, and in kangaroo rats captured at Sedan Crater in the Nevada Test Site (Martin and Koranda, 16). Hatch and Mazrimas (19) found that in mice and kangaroo rats given tritiated water, the specific activity of non-aqueous tissue hydrogens was 252-40X that of hydrogen in tissue water.

After tritium has been incorporated into one of the subcellular biochemical structures, a concentration factor of the order of 10 should be applied with respect to an assumed whole body distribution (Olive and LaJtja 20). Thus, the particular nature of uptake makes it difficult to assess any maximum permissible body burden for tritiated precursors (including tritiated water inasmuch as it is organically incorporated). Furthermore, with organically bound tritium the main concern is less with tissue damage than with mutagenic and/or possibly oncogenic effects on those cells which have a long life span and several mitotic cycles in the body, i.e., in fact any stem cell population.

The carcinogenic risk has been, as a rule, estimated from an average dose dependency model. Very little is known about the relative contribution of the time
sequence of dose during the usually long latency period. Such factors may be critical in carcinogenesis from internal irradiation and conceivably, the observation of various patterns of carcinogenic potency in relation with tritiated water or precursors might shed some light on these problems. For instance, in mice with an increased tumor incidence following administration of tritiated thymidine at birth, an average residual activity of about 32 per animal was found during the last third of the life span (Mewisien, Furedi and Rust, 21). Whether this final dose level, which contributed to most of the commitment dose, was the significant one in triggering carcinogenic action or whether the initial exposure, which was by far much larger, was the initiating one, remains unsolved. Similarly, the comparative carcinogenic yield of tritium-labeled precursors of either DNA, RNA or protein might in addition generate data pertaining to the carcinogenic process at the subcellular level. Inasmuch as radiation effects observed in experimental animals can be extrapolated to man, such findings are of obvious interest in view of the role of tritium as a possible environmental pollutant resulting from the expansion of nuclear power plants.

ACKNOWLEDGEMENTS

This investigation was supported by Public Health Service Research Grant No. CA-13080 from the National Cancer Institute and by Grant No. 5242 from Commonwealth Edison Company.

REFERENCES


DISCUSSION

A.A. MOGHISSI: I have always been impressed by your concern regarding the applicability of your findings to radiation standards. The increased toxicity of tritiated thymidine could be due to an increased residence time of tritium or to specific location on DNA following the introduction of the substance. I understand that you favour the hypothesis of specific location.

It is also being suggested that a toxicity value of 10 should be assigned to tritiated thymidine as compared with tritiated water. Would you agree with this value?

D.J. MEWISSEN: I agree with you that a factor of 10 seems adequate from our available data.

V. KASCHE: First, do your data on specific activity refer to activity per μg of protein DNA or per μg of tissue?

Secondly, do you have data on the specific activities in the organic components after chronic exposure?

D.J. MEWISSEN: Tritium activity is expressed as disintegrations per minute per μg of protein or DNA or RNA as well as disintegrations per minute per mg of tissue. In reply to your second question, we have no parallel data following chronic exposure.
J.K. MIETTINEN: What is the total dose from HTO and tritiated thymidine that produces tumours in mice, and after what induction times do the tumours become observable?

D.J. MEWISSEN: I have no data, at the present time, on the total dose absorbed by our animals as a result of lifelong exposure to tritiated water or tritiated thymidine. It should be borne in mind that in either case the time sequence of irradiation is biphasic. A brief sizeable dose is delivered as an acute dose, followed by continuous irradiation (commitment dose) arising from the residual activity. Furthermore, it would be interesting to correlate the carcinogenic potency of tritium with the space and time distributional dissipation of energy in tissues.

R.V. OSBORNE: Your data for tritiated thymidine refer to injected thymidine. For radiation protection purposes any intake of thymidine would be by ingestion. In this case there may be up to a factor of 10 catabolism of the thymidine in passing through the gut, i.e. only 10–15% of the thymidine may enter the blood stream (cf. VENNART, J., Radiotoxicology of tritium and $^{14}$C compounds, Health Phys. 16 4 (1969) 429). It would thus appear that the toxicity would not then be as great as you suggest.

D.J. MEWISSEN: I agree that any potential hazard from tritiated thymidine will be lessened when exposure results from ingestion rather than injection.

K. KREJCÍ: Do you find any differences in residence time or biological half-life as between various levels of activity ingested?

D.J. MEWISSEN: Within the range of activity that we used, no statistically different residence times were observed.
INDUCTION OF SPECIFIC-LOCUS MUTATIONS IN THE MOUSE BY TRITIATED WATER*

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Tennessee,
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Abstract

INDUCTION OF SPECIFIC-LOCUS MUTATIONS IN THE MOUSE BY TRITIATED WATER.

The results reported are the first obtained on transmitted gene mutations induced by tritium in any form in any mammal. They are, therefore, of obvious practical importance in estimating the possible biological hazards of man-made tritium in the environment. Male mice were injected intraperitoneally with either 0.75 or 0.50 mCi per gram of body weight of tritiated water. They were then used in our standard specific-locus mutation test in which the treated wild-type stock of mice is mated to a stock homozygous for seven recessive marker genes. Mutations at any of the seven loci are scored in the offspring. The earlier matings provided information on the mutation frequency in germ cells irradiated in postspermatogonial stages, and the later matings gave the mutation frequency in treated spermatogonia. The spermatogonia are the important cells so far as human risks are concerned, and the mouse results for this germ-cell stage yielded a relative biological effectiveness (RBE) of approximately 2 for tritiated water compared with low-dose-rate gamma irradiation. There are various uncertainties involved in arriving at this figure, and the difference between it and 1 is probably not statistically significant. However, for risk estimation, it seems prudent to use the RBE value of 2, which is, after all, the best point estimate computed from the present data.

INTRODUCTION

Increasing concern over the possible biological impact of tritium in the environment from the various man-made sources has stimulated research in this field, including the genetic study described here and that of Carsten and Commerford on dominant lethal mutations [1]. Before the experiments reported here, there were no mammalian specific-locus mutation data or other mammalian gene mutation information available for tritium in water, in organic molecules or in any other form. In addition to the concern over the possible genetic hazards of

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tritium, there is also scientific interest in a comparison of the genetic effects of low-energy beta particles with those of higher-energy external radiation sources. Preliminary accounts of this work have appeared in the Annual Reports of the Biology Division of Oak Ridge National Laboratory [2–4].

MATERIALS AND METHODS

The genetic results reported here were obtained by our standard specific-locus test [5] and are limited to those occurring in the offspring of treated males. (101 × C3H)F1 wild-type male mice were injected intraperitoneally with a single dose of tritiated water (HTO) and mated to our T stock females, which are homozygous for seven recessive marker genes. The offspring were scored for presumed mutations at the seven loci, and the presumed mutants were bred to establish allelism of the mutations and to determine the viability of the mutations in homozygous condition.

The treated mice were kept for two weeks after injection in an isolator system developed to prevent tritium release [6]. Two types of experiments were conducted. In the first, each male was mated with from three to five untreated females immediately after injection, and with the same number of new females at weekly intervals for four weeks thereafter. Thus, in this case, the first two weekly batches of females were placed with males in the isolators. This permitted the collection of data from germ cells exposed in the post spermatogonial stages (Experiment 1a). With the doses of HTO used, a temporary sterile period ensued in the males two to four weeks after treatment. When fertility returned, each male was caged with one female. The conceptions occurring from then on came from germ cells that had received most of their dose in spermatogonial stages (Experiment 1c). In a replicate (Experiment 1b) each male was mated to five females a week for the first three weeks after injection, and then not used for later matings. In the second type of experiment (Experiment 2), the males were not mated until after their removal from the isolators and after the end of their temporary sterile period. In this experiment each male was kept with two females.

In the first series of experiments, a few males were injected with 0.75 mCi of HTO per gram of body weight. With this dose, fertility before the temporary sterile period usually lasted only two weeks, and litter sizes were greatly reduced, particularly in the second week. The length of the temporary sterile period was similar to that observed following an acute dose of 1000 R of X-rays. The injected dose was, therefore, reduced to 0.50 mCi per gram of body weight for the remainder of the males in this experimental series and for all males in the rest of the study.

There are many complications in estimating the dose of radiation actually received by the genetic material of the germ cells. Our attempts to circumvent
TABLE I. MUTATION FREQUENCY AT SEVEN SPECIFIC LOCI IN OFFSPRING OF MALE MICE INJECTED WITH TRITIATED WATER

<table>
<thead>
<tr>
<th>Cell stage exposed to radiation</th>
<th>Experiment</th>
<th>Injected dose (mCi/g of body weight)</th>
<th>No. of offspring</th>
<th>No. of mutations</th>
<th>Estimated weighted mean dose to germ cells (rad)</th>
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<tr>
<td>Post-spermatogonial</td>
<td>1a</td>
<td>0.75</td>
<td>515</td>
<td>1</td>
<td>430</td>
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<tr>
<td></td>
<td>1b</td>
<td>0.50</td>
<td>3327</td>
<td>4</td>
<td></td>
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<tr>
<td>Spermatogonial&lt;sup&gt;a&lt;/sup&gt;</td>
<td>1c</td>
<td>0.75</td>
<td>2408</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>0.50</td>
<td>18218</td>
<td>13&lt;sup&gt;b&lt;/sup&gt;</td>
<td>615</td>
</tr>
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</table>

<sup>a</sup> A negligible portion (<1%) of the dose was received in post-spermatogonial stages.

<sup>b</sup> The number includes both mutants of a cluster of two in the offspring of one male in this experiment.

as many as possible of these, and our methods for arriving at a weighted mean dose for the germ cells in the experiments reported here, are described in a paper by Cumming and co-workers [6] also presented in this Symposium.

No contemporary controls were run in this investigation. The spontaneous mutation rate, which is low in comparison with the induced mutation rates obtained here, was estimated from the cumulative controls of extensive past experiments.

RESULTS

The mutation frequencies in the offspring from matings in which the germ cells had received their radiation dose in post-spermatogonial or spermatogonial stages, are shown in Table I. The distribution of the mutations among the seven loci and their viability in homozygous condition, when this was determined, are listed in Table II. Confirmation of allelism, i.e. that a mutation actually occurred at the locus to which it had been assigned on the basis of its phenotypic appearance, was established by breeding tests for all except five of the mutations. These include the three sterile mutants and two which died before testing: the mutation at the p locus listed as 'untested', and one of the s-locus mutations listed as 'untested' in the group from irradiation of spermatogonial stages. The other two
<table>
<thead>
<tr>
<th>Cell stage exposed to radiation</th>
<th>Result of viability test</th>
<th>a</th>
<th>b</th>
<th>c</th>
<th>p</th>
<th>d</th>
<th>se</th>
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<td>1</td>
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<tr>
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<td>1</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
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<td>5</td>
<td>3</td>
<td>4</td>
<td>21</td>
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\textsuperscript{a} Defined here as: lethal after perinatal period and before breeding age.

\textsuperscript{b} Lethal before birth or perinatally.

\textsuperscript{c} Mutants themselves were sterile and therefore could not be tested.

s locus mutations listed as ‘untested’ in the viability test were, however, tested for allelism. Although the allelism of the five mutations designated above was not established by breeding tests, it is highly likely, on the basis of extensive past experience, that the phenotypes of these five were correct indications of the loci involved.

The weighted mean dose for the experiment with irradiated post spermatogonial stages was calculated by estimating the accumulated dose for each succeeding day after HTO injection, and weighting by the number of offspring conceived on that day. For the experiment with irradiated spermatogonial stages, the weighting was by weekly, rather than daily, intervals. In both cases, the limited number of offspring obtained following the 0.75 mCi/g injected dose have been included, by appropriate weighting, in the estimated mean dose.

The induced mutation rate is obtained by subtracting the spontaneous mutation frequency for males obtained in past experiments, namely 28 mutations in 531,500 offspring. For the offspring from irradiated post spermatogonial stages this gives an induced mutation rate, per locus, per rad, of 44 × 10\textsuperscript{-5}. 
For the offspring from irradiated spermatogonial stages the induced mutation rate is $15 \times 10^{-8}$ per locus, per rad. In computing this, each of the two clusters of the mutants listed in Table I has been counted as two mutations with, however, a minor correction for the fact that the males who gave the clusters had a slightly larger number of offspring than the mean number of offspring for the rest of the males. Without this correction the mutation rate is $15.4 \times 10^{-8}$; with the correction it is $15.0 \times 10^{-8}$.

Each of the two clusters has, of course, been counted as a single mutational event in Table II, where we are concerned not with mutation frequencies but with the relative frequencies of different types of mutational event.

**DISCUSSION**

Both from the scientific point of view and for the practical application to risk estimation, it is of interest to compare the results obtained here from an internal emitter with those from external radiation sources such as X- and gamma radiation. Before attempting to make a quantitative comparison, we should first examine the data to see if the mutations induced by tritium differ in any important qualitative way from those obtained with X- and gamma rays.

One way of checking for qualitative differences is to examine the frequency distribution of the mutations among the seven loci. The distribution of the mutations in offspring from spermatogonial irradiation is not significantly different from that occurring with X- and gamma irradiation, where a low frequency is always observed at the $a$ and $se$ loci. The frequency at the $s$ locus in the present data is lower than expected on the basis of results from external radiation, but the difference is not statistically significant. Furthermore, in a replication of Experiment 2 not yet completed, and not otherwise reported here, five out of the total of eight mutations being tested are apparently at the $s$ locus, bringing the total at this locus more in line with the relative frequency expected from external radiation.

The mutations in the offspring from irradiation of post-spermatogonial stages appear to be more evenly distributed among the loci, a characteristic of those obtained from external irradiation of these germ cell stages [7]. With external irradiation, the frequency of detected chromosome aberrations is higher for treated post-spermatogonial stages than for spermatogonia. For example, deficiencies involving the $d$ and $se$ loci simultaneously are rare for irradiated spermatogonia, but common for treated post-spermatogonial stages [7, 8]. No $d$ $se$ deficiencies have been found in the small total number of mutations from post-spermatogonial stages in the present data, but there is other evidence of possible chromosomal aberrations. Thus, three of the mutants were sterile, and the $s$ locus mutant listed as 'untested' had a high frequency of sterility in her offspring.
The appreciable proportion of mutations, particularly those from irradiated spermatogonia, that turned out to be viable in the homozygous condition, is also in line with results from external radiation. One viable mutation at the c locus in the offspring from irradiated spermatogonia is intermediate in phenotype between albinism and the wild type, and one viable mutation at the b locus in the offspring from irradiated post spermatogonial stages is intermediate between b and the wild type.

In summary, the present data show no evidence that the mutations induced by injection of tritiated water differ in any marked qualitative fashion from those induced by external X- or gamma radiation. In view of this, it seems reasonable to proceed with a quantitative comparison of the mutagenic efficiency of the two types of radiation.

The mutation rates given here are based on mutant animals that survive to weaning, by which time mutations at all of the seven loci have become clearly discernible. It is known that a few mutations which were detectable at an earlier age occurred in animals that died before weaning. Thus the true mutation rate must be somewhat higher than that recorded at weaning. However, since the scoring of mutations has been done in the same way for external irradiation, it is valid, for comparative purposes, to use the mutation rates reported here.

For post spermatogonial stages, the experiment with external radiation which used a dose closest to that in the experiment reported here was one in which 300 R of X-rays was given at approximately 90 R per minute. The dose rate in the tritium experiments was considerably below this figure, but mutation rate in post spermatogonial stages appears to be independent of dose rate or, at least, not markedly affected by it [9]. Since more than 95% of the offspring born in the tritium experiment on post spermatogonial stages were conceived in the first two weeks after injection, the results from the first two weeks of mating in the X-ray experiment are selected for comparison. The mutation frequency for these weeks was 25 mutations in 18693 young [10]. Subtracting the spontaneous mutation rate gives an induced rate of \(61.2 \times 10^{-8}\) per locus, per R. Dividing this into the tritium-induced mutation rate of \(44 \times 10^{-8}\) per locus, per rad, and making a conversion from R to rad, yields a relative biological effectiveness (RBE) of 0.7 for tritium, compared with X-rays, for specific-locus mutations induced in post spermatogonial stages. With the limited number of mutations scored, this estimate is clearly not statistically significantly different from 1.

For comparison of results with spermatogonial stages, the dose rate is important, since there is a marked effect of the dose rate on mutation frequency in these stages [9, 11]. In the tritium experiment the dose rate is relatively high in the first few days after injection compared with what it is later, but even in the first half-day it probably does not average much more than about 0.1 rad per minute. In extensive exploration of the dose-rate effect with gamma rays on spermatogonial stages [11], it has been shown that, although there is a marked
effect on mutation frequency as dose rate is lowered from 90 to 0.8 R/minute, there appears to be no further change in mutation frequency when the dose rate is dropped to lower levels. It is appropriate, therefore, to compare the tritium results with gamma irradiation results at dose rates below 0.8 R/minute. The induced mutation rate for all data from spermatogonia irradiated in this dose-rate range was calculated in the review paper by Searle [12] to be $6.59 \times 10^{-8}$ per locus, per R. Dividing this into the tritium-induced rate of $15 \times 10^{-10}$ per locus, per rad, and converting from R to rad, gives an RBE of 2.2 for tritium, compared with gamma rays, for specific-locus mutations induced in spermatogonia. With, again, the limited number of mutations in the sample, with uncertainty as to the number of independent events involved when clustering occurs, and with some uncertainty as to the true doses, it is unlikely that this figure differs significantly from 1, but it is not possible to put a precise probability value on the null hypothesis.

We realize that a comparison of the effects of tritium with those of X- and gamma radiation at the microdosimetric level may be more complicated than is implied in the making of the above simple calculations. However, the RBEs derived above are of the kind which, in the absence of additional information, are used in risk estimation. They are presented primarily for that purpose.

With regard to applying these data to the estimation of genetic hazards of radiation in man, the spermatogonial stage is of prime importance. It would seem prudent at this time to assume that the RBF for exposure of these stem cells in the testis to tritiated water might be approximately 2.

ACKNOWLEDGEMENTS

We are grateful to all members of the Mammalian Genetics and Development Section who assisted with the laboratory work.

REFERENCES

DISCUSSION

A.A. MOGHISSI: As your data and those of other workers indicate, tritiated water introduces relatively little tritium into nucleic acids. We and various other people have found that under environmental conditions all organs and presumably all molecules are uniformly tritiated.

H.J. BURKI: Are the differences between the RBEs for post spermatogonial and spermatogonial tritium exposure significant?

W.L. RUSSELL: A test of significance of the difference between the RBEs for spermatogonial and post spermatogonial stages would have to take into account not only the binomial variability in the tritium data but also that in the X- and gamma-ray data, as well as in the controls. I have not attempted this. My guess is that such a test might show the difference in RBEs to be on the borderline of significance. However, there is also uncertainty as to the accuracy of the dose estimates, and there is the possibility of some extrabinomial variability (e.g. fluctuation in control mutation frequency). Therefore there would be some doubt as to the reliability of the probability value obtained from a test of significance.

H.J. BURKI: If there were a significant difference in the RBEs, what in your opinion would be the genetic explanation for this?

W.L. RUSSELL: If the RBE for post spermatogonial stages does prove to be significantly different from that for spermatogonial stages, one might look for an explanation in the qualitative differences in the types of mutations scored in the two stages. The specific-locus test has revealed that the ratio of X-ray-induced chromosomal aberrations, e.g. small deficiencies, to gene mutations is higher in post spermatogonial than in spermatogonial stages.

P.J.C. DINNER: If, as you suggest, your results indicating a higher RBE for tritium are not statistically significant, is it not premature to advise the use of
an RBE of 2 for risk estimation? Secondly, in your experimental design you must
have had in mind some range of RBEs, in order to justify the size and cost of your
proposed experiment. If you expected that your findings would indicate an RBE,
say, between 1 and 3, and that you would wish to publish this value, would it not
have been possible to design the experiment to provide a greater likelihood of
yielding a statistically significant result upon which to base your recommendation?

W.L. RUSSELL: I think a distinction must be made between advice on risk
estimation and recommendations regarding standards. I have concerned myself
with only the former of these two. Since our results are the only ones so far
obtained on transmitted gene mutations induced by tritium in any mammal, I do
not regard it as premature to report them and to advise that risk estimation should
be based on the best point estimate of the mutation frequency observed in the
experiments. I am sure you would agree that it would be scientifically and morally
unsound to say that risk estimation should be based on one half of the mutation
frequency actually observed.

The application of the results to the standard setting of radiation levels is
much more complex, and I presume your question is concerned primarily with
this problem. Consideration of any change in standards would have to take account
of many factors, such as the confidence limits of the point estimate, a comparison
of the results with other information, the proportion of the total risk from tritium
that is represented by gene mutation, and so on. Operations of this sort are, in
my opinion, best handled by committees of experts. I have served on several such
committees, and I prefer that procedure to making individual pronouncements on
whether the standards for permissible levels of radiation should be changed.

The design of our experiments was based on the calculation that if the true
RBE is 1, we should be able, with our sample size, to obtain enough mutations
to show — with high probability — that the RBE cannot be very high. Even
though the results give, as the best point estimate, an RBE of 2 for spermatogonial
stages, I believe they do exclude, with high confidence, the possibility that the
true RBE for this genetic effect could be as high as 5. Raising and scoring over
40,000 mice does not constitute an experiment that can easily be doubled in size,
let alone increased by an order of magnitude, and other people besides myself
have a say regarding the extent of our budget!
TRITIUM IN MAN

Session 8 and Session 9, Part 1
BIOLOGICAL HALF-LIFE OF TRITIUM IN HUMANS

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Abstract

BIOLOGICAL HALF-LIFE OF TRITIUM IN HUMANS.
The biological half-life of tritium in two former tritium dial painters was studied. Urine samples were collected periodically and were analysed for tritium. The study began several months after these workers had left the factory. The calculated half-lives were 340 and 630 days respectively.

One of the major factors affecting environmental health aspects of a toxicant is its resident time in a human body. It is well recognized that the toxicity of a radionuclide is directly proportional to its residence time, often expressed as \( \mu \text{Ci} \cdot \text{d} \). This value is the integral of concentration versus time. It appears that the concept of \( \mu \text{Ci} \cdot \text{d} \) is also valid for most, if not all, other toxicants. In these cases this value is expressed as \( \mu \text{g} \cdot \text{d} \).

Because the retention of many toxicants, including tritium, in the human body follows an exponential function, the knowledge of the biological half-life is necessary to assess the radiation dose resulting from tritium intake under dynamic conditions.

In previous studies this author and collaborators [1–4] have proposed the following equation describing tritium retention in the human body:

\[
A_t = A_s e^{-0.693t/T_s} + A_w e^{-0.693t/T_w} + A_i e^{-0.693t/T_i} + A_l e^{-0.693t/T_l}
\]

where \( A \) is the pool size, \( t \) is the elapsed time after injection, \( T \) is the tritium half-life and subscripts \( s, w, i, \) and \( l \) relate to the pool sizes and half-times denoted as short, water, intermediate and long.

This study contains data obtained from two new subjects who worked at a small tritium dial painting operation located in the southeastern part of the United States of America.
TABLE I. TRITIUM CONCENTRATION IN URINE SAMPLES FROM TWO SUBJECTS

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EXPERIMENTAL

The two subjects, MB and JM, are middle-aged females apparently in good health. They worked infrequently at the plant, in that they were called upon when the workload was high. The exposure was presumably through inhalation and skin absorption. Although a tritiated luminous compound was used, the exposure was probably predominantly, if not exclusively, tritiated water [5].
The routine sampling started about six months after the subjects stopped working at the plant. The sampling, sampling frequency and analysis have been described previously [1, 2]. The concentration of tritium in urine was well above the minimum limit of detection of about 0.2nCi/ltr. The statistical evaluation of the data was carried out using earlier procedures except for the description of error which now appears as the correlation factor.

RESULTS AND DISCUSSION

Table I and Figs 1 and 2 show the results of the study. Because the subjects were sampled after $T_s$, $T_w$ and $T_1$ had elapsed, no data for these terms are
available. The T1 values of 340 and 630 days are in reasonable agreement with previously reported values of 350, 550, 280 and 344 days. There are, however, certain peculiarities to these two cases. Both excretion cases show a sinusoidal behaviour. This behaviour is particularly pronounced in the case of JM. For example, a peak of 55 nCi/litr is followed by a concentration of 2 nCi/litr within about a three-month period. A similar excretion pattern was observed in a previous case [2]. Obviously, temperature, water intake and certain other factors influence the excretion rate. As Pinson and Langhan have shown, a four-fold reduction in the biological half-life of tritium may be observed if a forced water regimen is maintained. The deviations from the exponential pattern are, however, too large to be attributable to these factors. It is clear that mechanisms other than temperature and water intake are involved.

REFERENCES


DISCUSSION

R.V. OSBORNE: Were all the data points used in calculating the half-time?
A.A. MOGHISI: Yes.
R.V. OSBORNE: May I now comment as follows. When the single exponential components are plotted with the data, there is a tendency for data points, on average, to lie along a curve with decreasing slope at increasing time, i.e. a single exponential component may not be the appropriate model to use. A model with a time-constant increasing with time may be more suitable.¹

However, I think that this brings up a more general point on the analysis of tritium excretion curves. If it is reasonable to consider the body as a set of compartments, as is suggested by the application of a multi-exponential model to excretion data, then the dose commitment from any organically bound tritium from an intake of tritiated water depends on the fraction of organically bound hydrogen that may be derived metabolically from hydrogen in body water. The half-life, or turnover time, in any particular pool or compartment is not important. This simple dependency applies when the body tissue is not increasing in mass, i.e. catabolism is balancing synthesis, and applies to both acute and chronic exposures. If the components of a pool are cycling quickly, then for a given exposure to HTO in body water, a large fraction of the components may be labelled but the activity is retained for only a short time. If the cycling is slow, then only a small fraction is labelled but the activity is retained for a long time. Hence, if the fraction of hydrogen that may be labelled in this manner is included with the body-water component for calculating the dose commitment from an intake of tritiated water, a reasonable estimate is obtained.

S. PORTER: Are you certain that the worker intake of $^3$H luminous paint was in the form of tritiated water?

A.A. MOGHISI: Yes, we measured the chemical form.

S. PORTER: Could the sinusoidal excretion behaviour of both curves be due to a large $^3$H intake a few years earlier? In that case we are observing the asymptotic portion of the curve where we should expect significant variations from the three-compartment model.

A.A. MOGHISI: It's not for me to say that this could not be so!

J.K. MIETTINEN: I could explain your sharp peaks after 2–3 years in terms of recontamination through, for example, a visit to the plant or — a more pleasurable alternative — consumption of a bottle of 1963 wine, but I cannot understand the occasional sharp drops at all. They would mean that the slow catabolism of the tritiated compounds in the body stopped for a while and then started again. Such things just do not take place in metabolism.

A.A. MOGHISI: I quite agree that it is most strange. We checked on various possible pathways of contamination but always drew a blank. At all events, the highest value for 1963 tritium in the United States of America was not as high as the peak of the data we have here, and therefore consumption of wine cannot have made a significant contribution!

---

K. KREJČÍ: Did you check the subject’s home and clothing for surface contamination? Let us not forget that 4 mg of average-composition luminous compound is equivalent to 1 mCi.

A.A. MOGHISSI: We did not check the home, but it seems to me most unlikely that any contamination would remain for six months — the period between the cessation of exposure and the beginning of the study — either there or on clothing. We were also sure that the subject did not use a tritiated watch. You see, some of the peaks occurred two whole years after exposure. Another mysterious feature, not accountable in terms of contamination, is the periodic fluctuation, down in summer and up in winter. We feel there must be some biological factor at work.

J.A. HETHERINGTON: With reference to the sudden peaks which you have observed in tritium in urine concentrations during the slow excretion phase, have you made any measurements on control subjects who live in the same area as your experimental subject but have never been exposed in the luminizing plant?

A.A. MOGHISSI: No, we have had no control subjects living in the same area at the appropriate time. We have, however, a large volume of data on surface waters, on precipitation and on body water from subjects in various parts of the United States of America including from areas close to that which concerns us here. Also, several years ago we ran a programme called the 'Institutional total diet sampling network', which comprised a bioassay programme. The normal body water concentration in control subjects is in the range 0.2–0.5 nCi/ltr, which is much lower than the concentrations we report here.
DIFFUSION KINETICS OF TRITIATED WATER THROUGH HUMAN SKIN

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Abstract

DIFFUSION KINETICS OF TRITIATED WATER THROUGH HUMAN SKIN.

The delay in permeation of tritiated water vapour (HTO) through human skin has been estimated from measurements in vivo. The variations with time of the desorption rates of HTO from defined areas of the volar surfaces of forearms were measured immediately after the areas had been exposed to HTO for periods in the range 0.1–40 min. The HTO absorbed from each exposure was estimated from the concentrations of HTO measured in urine samples. Estimates of the delay were obtained for each exposure in two ways; from the observed ratios of total desorbed HTO to the total HTO absorbed by the individual, and from the estimated parameters of the theoretical desorption model fitted to the observed desorption data. The estimated values were in the range 8–17 min which is similar to the range of values obtained from in vitro studies elsewhere. The methods are generally applicable to determining the delay time for passage of HTO through diffusional barriers when one side of the barrier is not readily accessible.

Introduction

The permeation of water and tritiated water (HTO) through human skin has been extensively studied and reviewed almost independently by groups interested in health physics [1,2], dermatology [3-5] and biophysical or chemical phenomena [6]. The steady-state rate of permeation is well known from in vitro experiments involving both small area and whole-body exposures to HTO and these results agree well with in vitro measurements on samples of skin. It is known that several hours after an exposure to HTO tritium is fairly uniformly distributed throughout the body water. Not well known is the delay in vivo in permeation of HTO through the skin before being transported round the body.

The major barrier to water diffusion through the skin is known to be the bulk of the stratum corneum [3], a region of close-packed cells filled with keratin filaments in a matrix of lipid and non-fibrous protein. The underlying epidermis and the uppermost part of the dermis through which any substance must diffuse to reach the blood stream are believed to provide little barrier to water diffusion. The kinetics of water transport
have been studied in vitro with excised skin samples since, clearly, in vivo measurement of the rate of appearance of activity just beneath the barrier poses severe experimental problems.

Several factors may cause the in vivo kinetics to differ from those observed in vitro [4]; the degree of hydration and swelling, the extent to which the stratum corneum folds, and the condition of the skin appendages - sweat ducts and hair follicles - which are affected by excision.

Measurement of the total absorption through the skin in vivo provides no information as to the delay in the skin as is illustrated later. In the work described here the delay time through the skin barrier is estimated from the measured desorption rates of activity from skin, previously exposed to HT0 vapour. Although applied in this instance to human skin the method is applicable in principle to any system in which the kinetics of diffusion of HT0 through a barrier are being investigated but in which one side of the barrier is inaccessible.

Theory

The simplest model for the skin barrier to water permeation is a single inert membrane (the stratum corneum) of given thickness and diffusion constant, with effectively no delay from the inner surface of the barrier through the epidermis and dermis to the vascular system and a perfusion rate in the vascular system sufficiently rapid to maintain effectively zero concentration at the inner surface of the membrane. The kinetics of diffusion in such a system may then be described by Fick's equations for diffusion in one dimension:

\[ F = -D \frac{dc}{dx} \]  

(1)

and

\[ \frac{dc}{dt} = D \frac{d^2c}{dx^2} \]

(2)

where \( F \) is the flux of water (i.e. amount per unit area and unit time), \( D \) is the diffusion constant, and \( C \) is the concentration of water in the membrane at distance \( x \) from the outer surface. Two features of the solutions to these equations for various conditions (see, for example, references [7,8], are pertinent to this work: First the total intake of water per unit area, \( Q \), after exposure to a concentration of water in air of \( C_0 \) for a time \( T \), is given by

\[ Q = PC_0 T \]

(3)

where \( P \) is the permeability constant characteristic of the membrane.

Second, the delay in transmission of water through the membrane may be characterized by a 'lag-time' \( \phi \), the intercept on the time axis of an extrapolation of the linear (steady state) portion of the graph of the amount of water passed through the membrane against time (Fig. 1). \( \phi \) is related to \( D \) and the membrane thickness, \( d \), by

\[ \phi = \frac{d^2}{6D} \]

(4)
FIG. 1. *The amount of water vapour that has permeated through a membrane at various times during and after application of a concentration of water \( C_0 \) on one of the surfaces for time \( T \).* The concentration before and after the application and on the other surface is zero. The permeation rate \( PC_0 \) is the slope of the steady-state part of the absorption curve, the intercept of which with the time axis is the 'lag-time' \( \phi \). The amount ultimately permeating the membrane \( Q \) is proportional to \( T \). The amount that desorbs from the first surface after the source of water vapour has been removed increases to \( A_d \) as shown at the right. The ratio \( A_d/Q \) is designated \( R_d \) in the text. \( A_d \) is the amount desorbed in any particular time interval.

The amount that finally passes through the membrane is independent of \( \phi \) as is apparent from elementary considerations. Hence, measurement of \( Q \) provides no indication of the membrane 'lag-time'.

The methods of Barrer [7] and Crank [8] may be extended to solve equations (1) and (2) for the amount of water desorbed through the outer surface after a given exposure.

The solution is of the form

\[
I = f(t) - f(t + T)
\]  

(5)

where \( I \) is the flux of water through the outer surface of the membrane at time \( t \) after the end of an exposure of time \( T \) to a concentration in air of \( C_0 \). The most convenient form of the function \( f(t') \) for this application is

\[
f(t') = 2PC_0 \sum_{n=1}^{\infty} \exp \left( -\frac{R^2\pi^2}{6} \frac{t'}{\phi} \right)
\]  

(6)

Observation of the variation of the rate of desorption with time of HTO from the outer surface of the membrane after an exposure to \( C_0 \) for time \( T \) allows \( P \) and \( \phi \) to be obtained by fitting equation (5) (with \( \phi^2 \)) to the experimental data.
FIG. 2. Curve 1: The ratio, $R_d$, of desorbed activity to total absorbed activity versus the ratio of exposure time to 'lag-time' ($T/\phi$). Curve 2: The desorbed activity ($A_d$) as a fraction of the activity absorbed from an exposure of length $\phi$ ($PC_0\phi$) versus the ratio of exposure time to 'lag-time' ($T/\phi$).

From equations (3), (5) and (6), the ratio ($R_d$) of the total amount desorbed after an exposure for time $T$ to the total amount ($Q$) that ultimately passes through the membrane is given by

$$R_d = \frac{2\phi}{T} \left[ 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp \left( -\frac{n^2 \pi^2}{6 \phi} T \right) \right]$$

(7)

The value of $R_d$ against the ratio $T/\phi$ is shown on Fig. 2.

Measurement of $R_d$ for a given $T$ allows $\phi$ to be determined.

For exposure times much longer than the 'lag-time' ($T \gg \phi$) the desorbed activity ($A_d$) is given by $2PC_0\phi$ (from equations (3) and (7)). Hence, if the exposure has continued so that steady-state conditions have been established, the activity absorbed is simply twice the activity absorbed from an exposure of length $\phi$. The variation of $A_d/PC_0\phi$ with $T/\phi$ is also shown on Fig. 2.

Methods

The intent in the experiments described here was to establish steady-state values of humidity, temperature, pressure and water vapour flux over a well-defined area of skin, introduce tritium-labelled water molecules ($HTO$) over the area for a measured time without changing the steady-state values and, having reintroduced non-labelled molecules ($H_2O$) above the skin, to collect the desorbing HTO under the same steady-state conditions.
Areas of the volar surfaces of forearms of volunteers were exposed to HTO in air for times ranging from 0.1 to 60 min using the inverted cup-like device shown in Fig. 3. The enclosed volume (≈ 25 cm³) could be ventilated with appropriately humidified air through the two upper connections. The cup was held on the skin by suction on the annular groove around the rim. The suction resulted from a pressure drop of 15 kPa across twelve notches in the outer edge of the rim, maintained by drawing a constant flow of air out of the connection to the annular groove. This method of attachment was reproducible, avoided the use of glues that could absorb HTO and was sufficiently comfortable for the prolonged exposures. The retractable disc thermistor allowed the skin temperature to be monitored just before an exposure.

Desorbing HTO was collected, for transfer to vapour traps, in a second cup (Fig. 3) that enclosed a slightly greater area than used for the exposure (to avoid errors from misalignment). The use of this cup
FIG. 4. The arrangement of tritiated and non-tritiated water vapour flows and the sampling devices for measuring the concentrations of HTO during exposures and the activity of HTO subsequently desorbed. In the exposure section the open arrows indicate the flows before exposure; the solid arrows during exposure. In the desorption section the arrows indicate the method of obtaining serial samples.

with an independent sampling system ensured that all tubing and devices used for collecting the desorbed HTO would not have been contaminated by HTO in the exposure. An excess of non-tritiated air at the correct humidity was introduced into the annular space and was therefore exclusively available for transferring the desorbed activity.

The arrangement of flows is shown in Fig. 4. Prior to the start of an exposure to HTO, identical steady-state conditions were established for both H₂O and HTO flows; the H₂O flow passing through the exposure cup. Both flow systems were adjusted so that the air pressure inside the exposure cup with either flow was equal to ambient. The vapour traps were sets of water bubblers [9] arranged in parallel so that sequential samples from measured volumes of air could be collected. For an exposure, electrically operated valves switched the HTO flow through the exposure cup for the desired time and re-established the H₂O flow at the end of
FIG. 5. The permeability constants (P) estimated for the skin of 6 persons. The average value for each person is shown. The measured skin temperatures were in the range 30°–33°C. (Many of these data were obtained as controls in other absorption experiments and were not associated with desorption measurements.) The unit of the permeability constant is derived from the rate of activity transmission (Bq/min, say) per unit area (m²) for unit concentration in air (Bq/L). (L is the symbol used in this paper for litre.)

the exposure. At the flow rate used (3 L/min) the time constant for the change in concentration of HTO in the cup was approximately 0.5 s. The concentration of HTO in air was measured both before and during exposures and, in the case of exposures of less than 1 min, after exposures. The concentrations used ranged from 20 – 200 MBq/L. Within less than 5 s after the end of an exposure the exposure cup was replaced by the desorption cup on the arm and sequential collection of desorbed HTO started. Samples were collected over progressively greater intervals, starting at 10 s, for up to 100 min after an exposure. Generally the desorptions were continued long enough to allow the desorption rates to be followed for three decades.

Urine samples were obtained from the volunteer subjects before an exposure, at approximately two-hour intervals for the following ten hours and less frequently in the next day. All measurements of tritium activity were with the same liquid scintillation counter. The total activity absorbed (Q) from any exposure was calculated from the empirical relation [2]

\[ Q = 0.55W^{-0.53}H^{0.4}U \]
FIG 6. The ‘lag-times’ $\phi$, estimated for the skins of three persons from the ratio ($R_d$) of desorbed activity to total absorbed activity and, for one person, from fitting the theoretical desorption expression to observed variations of desorption rate with time. The error bars in the former are standard deviations estimated from measurements of urine concentrations, exposure times and desorption times and in the latter from the least-squares fit. The arrows in the lower plot show the effect on the estimated values of $\phi$ if a two-component model is used for three of the desorptions that were followed for at least 40 min.

where $W$ was the subject’s mass (in kg), $H$, the height (in cm) and $U$, the increment in the concentration per litre of tritium in urine estimated by extrapolation from the post-exposure samples.

Incremental concentrations in urine ranged from 2.5 to 150 kBq/L with an average of 30 kBq/L.

Estimates of $\phi$ were obtained from each set of desorption data by a least-squares fit of equation (5) (integrated for the period of each sample) for samples taken after 10 s from the end of an exposure. The
FIG. 7. The variation of desorption rate ($I$) with time ($t$) after the end of exposures of various periods ($T$) for one person. The curves were obtained from the least-squares fit of the theoretical variation of desorption rate with time.

data were corrected for carry-over in the sampling system from one sample to subsequent ones.

Estimates of $P$ were obtained for the same experiments from $Q$ and alternative estimates of $\phi$ were obtained from the observed ratio of desorbed activity to $Q$. The ratio $R_A$ of equation (7) refers to the total desorbed activity which, in practice, for the shortest exposure times, was not measurable since a large fraction of the desorption occurs in the first few seconds. Hence an expression for the ratio, $R'$, of the activity desorbed between two times to the total absorbed activity (see Fig. 1) was used in an iterative routine with equation (7) (Fig. 2) to obtain an estimate of $\phi$ consistent with the observed $R'$.

Results and Discussion

Estimates of the permeability constants for the forearm of 6 individuals are shown in Fig. 5. These are derived from measured concentrations of tritium in urine and illustrate the point made earlier that there is no systematic variation in observed total absorption rate with exposure time from 6 s up. Surface adsorption does not therefore appear to extend the effective length of even very short exposures of a few seconds. The desorption data should therefore reliably reflect the barrier kinetics.

The permeabilities observed are within the range summarized earlier [2], from in vivo measurements with HTO ($2.1 - 8.4$ L min$^{-1} m^{-2}$; mean $4.3$ L min$^{-1} m^{-2}$) and lie between the values estimated as typical of the total skin ($6.5$ L min$^{-1} m^{-2}$) and of the volar surface of a forearm ($1.7$ L min$^{-1} m^{-2}$) from the summary [3] of measurements with water.
Values for $\phi$, the 'lag-time', obtained from the ratios of desorbed activity to total absorbed activity for three individuals are shown in Fig. 6. The average values from the two main series are $8.3 \pm 0.7$ min (individual A) and $13.8 \pm 0.7$ min (individual B). These values are similar to the value quoted [3] for water permeation through skin excised from the volar forearm (12 min). Although skin samples excised from various parts of the body have widely disparate delay times (e.g. 106 min for the sole of the foot at one extreme to 1 min for scrotal skin) the values for the major areas (10 - 20 min) appear to be similar to that of the forearm. The agreement of the in vivo results here with the known in vitro value for that tissue supports the idea that the delay times from in vitro studies are applicable to in vivo exposures to HTO so that the major fraction of any intake of HTO through the total skin area will have a 'lag-time' of 10 - 20 min.

The alternative method for estimating $\phi$ by fitting the theoretical desorption curve to serial measured values of desorbed activity is illustrated in Fig. 7 for some of the exposures on one individual. The values of $\phi$ so obtained for this individual are compared to the previous estimates of $\phi$ in Fig. 6. The mean value of $\phi$ for this curve-fitting method ($13.4 \pm 1.2$ min) does not differ significantly from that obtained (above) from the total desorption method but, as Fig. 6 indicates, there is a systematic variation between the two sets, the fitted values being greater for the desorption after the longer exposures and the fitting errors being greater.

The systematic variation is removed if a more complicated mathematical model is used — for example if a fraction of the barrier to permeation is
assumed to have a different (longer) 'lag-time'. Such a fraction may be attributable to skin appendages [10] or pores [11]. The fits with such a model with one barrier shunted by another (i.e. parallel pathways and hence, 4 fitted coefficients) to desorptions that have been followed for 60 min or more after long exposures generate 'lag-times' for the main component that are similar to those obtained with the simple model from desorptions after short exposures. After the short exposures the desorptions are dominated by the main component within the range of desorption rates observed. On Fig. 6 the effects of the more complicated fitting are shown for the longer exposures and desorptions. Similarly for individual A a mean 'lag-time' of 10.8 ± 0.4 min is obtained for the main component of a parallel pathway model, in fair agreement with the mean obtained above from the total desorption. Fig. 8 shows the fit with two components obtained for the data from the longest desorption and exposure followed. The minor components which, in the fits to this model, account for up to 10% of the permeations, have 'lag-times' in the range 35 - 70 min. Very fast components but with very low relative permeation rates such as have been observed [10] in vitro would not be detectable in the analysis here.

However, to account for the longer component observed, alternative refined models may be plausible - e.g. diffusion in the epidermis or dermis, in the various skin appendages, a slow pick-up of activity by the blood or diffusion laterally at the edges of the exposed area. Such a component identified with either of the first two would be consistent with the rise time of activity observed in urine [2]. Mathematical models for such processes have been outlined [12] and fitted to some of the data. Clearly it is apparent that such models are not likely to be unequivocally distinguished by the data and hence further mathematical refinement has therefore not been attempted. However, the lack of effect of blood perfusion on the intake kinetics has been demonstrated [13] experimentally by observing the desorption from skin perfused by an enhanced blood flow obtained by ingestion of nicotinic acid prior to exposure to HTO. The desorption rate is only marginally affected by the enhanced blood flow which would therefore appear to be not a major contributor to the observed longer component.

Conclusions

- Estimates of the delay in passage of tritiated water vapour (HTO) through human skin in vivo have been obtained from measurements of the activity desorbed from the skin after exposure to HTO by two methods:

  a) By fitting the theoretical expression for the desorption rate to the experimental desorption data, and

  b) By comparing the total measured desorption with the total absorption as determined from measurements of tritium in urine.

The methods are generally applicable to determining the delay times for passage of HTO through diffusional barriers when one side of the barrier is not readily accessible.

- The delay times measured by the two methods agree reasonably and are similar to values reported from measurements in vitro with samples of intact stratum corneum. Evidence of a component in the permeation barrier with a longer delay time was noted but its source was not identified.
ACKNOWLEDGEMENTS

The collaboration of W.R. Bush in designing and carrying out these experiments, the medical advice from A.A. Driedger and, above all, the willing cooperation of the volunteers are gratefully acknowledged.

REFERENCES

MODELS FOR ASSESSING THE ENVIRONMENTAL EXPOSURE BY TRITIUM RELEASED FROM NUCLEAR INSTALLATIONS

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Abstract

MODELS FOR ASSESSING THE ENVIRONMENTAL EXPOSURE BY TRITIUM RELEASED FROM NUCLEAR INSTALLATIONS.

The ingestion of food contaminated by HTO constitutes a critical exposure pathway in the environment of nuclear installations, e.g. fuel reprocessing plants, which release considerable amounts of tritium into the atmosphere. To calculate the ingestion dose for members of the public, a number of national regulations are based on the specific activity model, which assumes that the HTO/H₂O relation is identical in vegetation and in ground-level air humidity. In the present study, the specific activity model is modified so that the water supply of plants is not only taken from humidity but also from soil water and from precipitation deposited on the leaves. A comparison of the models shows that the uncorrected model underestimates the HTO contamination of plants for small source distances, particularly for major release heights. In addition, a modification is suggested for the general ingestion dose model, developed for aerosols, which can be applied to tritium.

1. INTRODUCTION

In connection with the release of radioactive substances with waste air from nuclear installations, especially in the case of reprocessing plants, tritium may constitute one of the critical nuclides for environmental exposure. The main exposure pathway in this connection is the ingestion of food contaminated by tritium-containing water (HTO). In calculating the ingestion dose to which persons are exposed in the environment of a source as a result of tritium releases, the Federal German guideline [1] proceeds on a specific activity model, according
to which the tritium content of plant water, related to the content of stable hydrogen, is the same as that in air humidity$^1$:

$$
\frac{C_{\text{HTO},L}^P}{C_{\text{H}_2\text{O}}^L} = \frac{C_{\text{HTO}}^L}{C_{\text{H}_2\text{O}}^L} \times \frac{Q}{\phi}
$$

where

- $C_{\text{HTO},L}^P$ = tritium activity concentration of the plant in equilibrium with air humidity (Ci/kg)
- $C_{\text{HTO}}^L$ = tritium activity concentration of air humidity (Ci/kg)
- $C_{\text{H}_2\text{O}}^P$ = water content of the plant (kg/kg)
- $C_{\text{H}_2\text{O}}^L$ = water content of the air (kg/kg)
- $\phi$ = absolute humidity (kg/m$^3$)
- $Q$ = average release rate (Ci/s)

The HTO-concentration in the atmosphere may be calculated using the long-time diffusion factor (s/m$^2$) for quasi-continuous releases:

$$
X = \frac{N}{\sqrt{2\pi}} \sum_j \left\{ \frac{e^{-\frac{H^2}{2\sigma_j^2}}}{\sigma_j} \right\} \sum_k \frac{p_{ijk}}{u_{jk}}
$$

where

- $H$ = effective release height (m)
- $X, y, z$ = downwind, crosswind and vertical co-ordinates (m)
- $p_{ijk}$ = 3-dimensional diffusion statistics (wind direction i, diffusion category j, wind velocity class k)
- $\sigma_j$ = vertical diffusion parameter (m)
- $u_{jk}$ = wind velocity at release height (m/s)
- $N=12$ = number of wind direction sectors

$^1$ According to the U.S. Regulatory Guide [2], an additional factor of 0.5 is introduced, which accounts for the assumption that the tritium concentration in plant water is only half the amount of that in air humidity.
Consequently, in computing the contamination of the food chain, tritium (as well as $^{14}$C) is treated differently in comparison with all the other radionuclides for which the contributions of direct deposition on the leaves and uptake via the roots from the soil contaminated by fall-out and wash-out are totalled [1]:

$$C_P^P = Q \left\{ (v_g X + fW) \frac{1 - e^{-\lambda_p t_e}}{\lambda_p} + (v_g X + W) T_P \frac{1 - 3}{p \lambda_r} e^{-\lambda_r t_b} \right\}$$

(3)

where

- $C_P^P$ = activity concentration of the plant (Ci/kg)
- $v_g$ = deposition velocity for fall-out (m/s)
- $W$ = long-time wash-out factor (cf. Eq. (6)) (m$^{-2}$)
- $f$ = retention factor for wash-out activity on plants
- $\lambda_p$ = effective removal constant of plant contamination (s$^{-1}$)
- $\lambda_r$ = radioactive decay constant
- $t_e$ = growing period (s)
- $t_b$ = accumulation time for soil activity (s)
- $t_v$ = decay time of food before consumption (s)
- $Y$ = plant density (kg/m$^2$)
- $P$ = mass per unit area of 15 cm soil (kg/m$^2$)
- $T_P$ = transfer factor from soil to plant

A direct application of this general model to tritium is not possible, some of the reasons being:

(a) The values for $v_g$ and $f$ recommended for aerosols are not applicable to HTO. Corresponding values for tritium cannot be specified. In any case, the special conditions prevailing for a plant’s uptake and discharge of tritium with water would require modifications of the model.

(b) Because of the processes of seepage and evaporation of HTO in the soil, account must be taken of the removal in the soil which is neglected in connection with other nuclides.

(c) In view of the high dilution of tritium by stable hydrogen, it will be necessary to consider the T/H ratio, which suggests application of the specific activity model.
Since the plant's water requirements are covered essentially by uptake from the soil via the roots and, to a lesser degree, by uptake from air humidity via the leaves or precipitation deposited on the leaves, the specific activity model will here be extended with the aim of determining the ingestion dose in a more realistic manner. This procedure will reveal whether it will be possible, for practical application, to obtain a sufficiently precise or at least conservative calculation of the ingestion dose by simple treatment according to Eq. (1).

2. MODIFIED SPECIFIC ACTIVITY MODEL

In analogy with the tritium uptake from air humidity, the following may be derived for the uptake from rainwater deposited on the plant according to the specific activity model:

\[
C_{\text{HTO},N}^{p} = \frac{C_{\text{H}_2\text{O}}^{N}}{C_{\text{H}_2\text{O}}^{p}} C_{\text{HTO}}^{N}
\]  

(4)

where

- \(C_{\text{HTO},N}^{p}\) = mean tritium activity concentration of the plant from the uptake of rainwater via the leaves (Ci/kg)
- \(C_{\text{H}_2\text{O}}^{N}\) = water content of precipitations

The mean HTO concentration of precipitations (Ci/kg) in the wind direction sector \(i\)

\[
C_{\text{HTO}}^{N} = \frac{Q_i W}{n_i}
\]  

(5)

is derived from the deposition of tritium with the precipitations \(Q_i W\)(Ci/m²s) in this sector, divided by the precipitation intensity \(n_i\)(kg/m²s), which is calculated from the annual volume of precipitation \(N_i\)(mm/a) in connection with the wind direction \(i\) (1 mm = 1 kg/m²).

According to Ref. [3], the long-term diffusion factor for the wash-out in sector \(i\)

\[
W = \frac{6 c N_i}{\pi u_a x}
\]  

(6)
is proportional to the annual precipitation rate \( N_i \) (mm/a) related to the wind direction \( i \) and inversely proportional to the mean wind velocity \( u_a \) and the source distance \( x \). \( c \) is the ratio of wash-out constant (s\(^{-1}\)) and precipitation intensity (mm/s) and may be assumed as constant in a first approximation.

Consequently, according to the specific activity model, the plants’ tritium concentration due to the uptake of water via the root,

\[
c_{\text{HTO,B}}^P = \frac{c_{\text{H}_2\text{O}}} {c_{\text{H}_2\text{O}}} c_{\text{HTO}}^B
\]  

(7)

should be assumed as proportional to the HTO content of the soil water,

\[
c_{\text{HTO}}^P = Q W f_i \frac{1 - e^{-\lambda t_b}} {P \lambda}
\]  

(8)

which results from the solution of the differential equation

\[
c_{\text{HTO}}^B = f_i \frac{Q W} {P} -\lambda c_{\text{HTO}}^B
\]  

(9)

The factor \( f_i = N_i / \Sigma N_i \) accounts for the dilution of the tritium activity in the soil water due to non-contaminated precipitations from other wind directions. \( P \) (kg/m\(^2\)) is the mass per unit area of soil down to a depth \( z_B \) (e.g. \( z_B = 15 \) cm), and \( \lambda \) (s\(^{-1}\)) is the removal constant for HTO in this soil layer.

A combination of the three contributions to HTO uptake from air humidity, precipitation deposited on the plant and soil water may be attained by weighted averaging:

\[
c_{\text{HTO}}^P = f_i c_{\text{HTO},L}^P + f_N c_{\text{HTO},N}^P + f_B c_{\text{HTO},B}^P
\]  

(10)

in which the weight factors \( f_i, f_N \) and \( f_B \) denote the respective shares of the plants’ water uptake from air humidity, precipitation and soil water. Substitution of Eqs (1) and (4) for Eq. (8) will lead to
\[
C_{HTO}^P = Q\left( f_L \frac{C_{H_2O}^P}{\phi} \chi + f_N \frac{C_{H_2O}^P}{n_i} \right) \\
+ f_B \frac{N_1}{\Sigma N_i} \frac{C_{H_2O}^B}{C_{H_2O}^B} \left( 1 - e^{-\lambda t_B} \right) \]  

(11)

3. FACTOR COMPARISON BETWEEN THE SPECIFIC ACTIVITY MODEL AND THE GENERAL MODEL

By means of a factor comparison of the modified specific activity model, Eq. (1), with the general model, Eq. (3), it will now be possible to determine the parameters \( v_g \) and \( f \) for the deposition of tritium and to verify the transfer factor \( T_p \) between soil and plant. For this purpose, two simple modifications of Eq. (3) are still required:

(a) In the last parenthesis addend, the radioactive decay constant \( \lambda_r \) is to be replaced by the effective removal constant \( \lambda \) to allow for the loss in the upper soil layer due to seepage and evaporation, which is significant in connection with HTO.

(b) A distinction must be made between the deposition velocity on the plant \( v_g^P \) (first addend) and on the soil \( v_g^B \) (second addend).

The exponential function at the end of Eq. (3), allowing for the decay up to consumption, will be equal to one for tritium.

A factor comparison of the expressions in parentheses in Eqs (3) and (11) will then lead to

\[
v_g^P = f_L \frac{C_{H_2O}^P}{\phi} \frac{Y \lambda_P}{1 - e^{-\lambda_P t}} \]  

(12)

\[
v_g^B = 0 \]  

(13)

\[
f = f_N \frac{C_{H_2O}^N}{n_i} \frac{Y \lambda_P}{1 - e^{-\lambda_P t}} \]  

(14)
\[ T_P = f_B \frac{N_1}{\Sigma N_1} \frac{C_P}{C_{H_2O}} \]  

(15)

If we assume, for instance, that the plant covers 80% of its water requirements from the soil water and 10% each from air humidity and leaf moisture \( f_B = 0.8, f_L = f_N = 0.1 \), that the water content amounts to 75% in the plant \( [2] \) and 20% in the soil \( [4] \) \( (C_{H_2O} = 0.75, C_{H_2O}^B = 0.2) \), that HTO is removed from the plant leaves with a half-life of 1 h \( \text{cf. Ref.} [5] \) \( (\lambda_0 = 1.9 \times 10^{-4} \text{ s}^{-1}) \), and that 200 out of a total of 700 mm/a of precipitation are linked with the main wind direction \( (N_i/\Sigma N_i = 200/700) \), the following effective values for HTO will result instead of the values \( v_g = 1 \times 10^{-3} \text{ m/s} \) and \( f = 0.2 \) recommended in Refs \( [1, 2] \) for aerosols based on a mean absolute air humidity \( \phi = 9 \times 10^{-3} \text{ g/m}^3 \) \( [1] \) and a growth density \( Y = 1 \text{ kg/m}^2 \):

\[ v_g = 1.6 \times 10^{-3} \text{ m/s} \]
\[ f = 2.3 \]

Instead of a soil-to-plant transfer factor of 4.8 recommended in Ref. \( [2] \) for tritium, a value of

\[ T_P = 0.86 \]

is calculated. Naturally, the terms \( v_g \) and \( f \) do not have the physical significance of a deposition velocity or a retention factor for HTO, but they constitute 'effective' values of these quantities, determined by adaptation to the specific activity model, thus offering the possibility of applying Eq. (3) to tritium as well and, consequently, to proceed on the same general formula for determining the ingestion dose for tritium in computer programs (as well as for \( ^{14} \text{C} \), which still remains to be implemented in detail).

4. INGESTION DOSE CALCULATION FOR TRITIUM

For calculation of the annual dose from the ingestion of contaminated food, the activity concentration of the harvested products must be multiplied by the annual rate of consumption (kg) and the dose factor for the respective nuclide and organ.

For calculation of the tritium concentration in milk and meat from the activity concentration in fodder (grass) according to the equation

\[ C_{M,F}^F = T_{M,F} \]

(16)
where

\[ f_p = 0.5 \quad = \text{share of the grazing period} \]
\[ I = 55 \quad = \text{grass consumption of a cow (kg/d)} \]
\[ T_{M,F} \quad = \text{transfer factor grass to milk or grass to meat (d/kg)} \]

the specific activity model is abandoned, according to which \( I \times T_{M,F} \) would have to be equal to one. This may be justified by the fact that a cow does not cover its entire water requirements from grass (55 kg freshweight daily corresponding to about 44 kg water daily), but that about one third is covered by non-contaminated drinking water. Allowing for this dilution, \( T_{M,F} = 1.2 \times 10^{-2} \text{d/ltr} \) is obtained according to the specific activity model. The values recommended in Ref. [2] are \( T_M = 1 \times 10^{-2}, T_F = 1.2 \times 10^{-2} \text{d/ltr} \).

Since, therefore, the ingestion dose for each individual food is proportional to the activity concentration in the plants, the following calculated examples will be based on a comparison between the tritium concentration of plants in connection with a unit release rate.

5. COMPARISON BETWEEN THE SIMPLE AND THE MODIFIED SPECIFIC ACTIVITY MODEL FOR TRITIUM

For evaluation of Eqs (1) and (11), the parameter values already used in Section 3 were again utilized. These values are merely of an exemplary character and will, of course, depend on the meteorological conditions and soil properties at the site as well as on the type of plant involved in each specific case. However, they correspond to realistic values as may actually be found at sites.

The long-term diffusion factor was calculated for release heights of 50, 100 and 200 m using the Jülich diffusion statistics and the diffusion parameters determined experimentally [6].

For the long-term wash-out factor according to Eq. (6), a precipitation constant \( c = 1 \times 10^{-4} / 8800 = 1.14 \times 10^{-8} \text{a/(mm/s)} \) according to Ref. [7] and a precipitation volume in the main wind direction sector \( N_t = 200 \text{ mm/a} \) (corresponding to \( n_1 = 6.3 \times 10^{-8} \text{ mm/s} \)) were used. The product \( P \cdot \lambda \) in Eq. (8)

\[
P \lambda = \left( \rho z_o \right) \left( \frac{1}{T} \right) = (\rho z_o) \left( \frac{u_B}{z_o} \right) = \rho u_B
\]

was replaced by the product of the soil density \((\rho = 1600 \text{ kg/m}^3)\) and the vertical migration velocity of an HTO-layer in the soil \( u_B = 1 \text{ m/a} = 3.2 \times 10^{-8} \text{ m/s} \) [8].

(\(T\) is the mean residence time of an HTO molecule in the soil layer of thickness \( z_o \).)
Fig. 1. Tritium concentration in plants calculated by the specific activity model (Eq. (1)) and the modified specific activity model (Eq. (11)) for 100-m release height.
FIG. 2. Release height dependence of tritium concentrations in plants.
The exponential function in Eq. (11) will disappear, since the effective half-life of HTO in the soil is short compared with the time of accumulation (e.g. $t_b = 50$ a).

The results of the comparative calculation are outlined in Figs 1 and 2, where the tritium concentration of plants is represented as a function of the source distance for a release rate of 1 Ci/s. For a release height of 100 m, Fig. 1 shows, on the one hand, the course of the tritium activity concentration according to the simple specific activity model, and on the other hand, the plant’s total tritium activity concentration according to the modified model as well as its composition from the contributions by the uptake of HTO from the soil water, air humidity and precipitation moisture. Whereas the plant’s tritium concentration according to the simple model of HTO concentration follows the air near ground level, and thus disappears in close proximity to the source similar to the long-time diffusion factor, its course in source proximity according to the modified model is essentially that of the long-term wash-out factor, i.e. reciprocal to the source distance. In this case, the simple model will only lead to a conservative assessment of plant contamination at source distances exceeding the distance of the maximum of the air concentration near ground level.

In Fig. 2, the two models are compared for various release heights. Since the wash-out contributions (soil water, precipitation moisture) are independent of the release height, whereas the maximum of the contribution via air humidity appears at an ever increasing distance with growing source height while decreasing in value, the extent of underestimation of the environmental impact observed in connection with Fig. 1, computed according to the simple model, is found to be dependent on the release height. Moreover, the possible underestimation also depends on the source distance of the fence, i.e. on the distance of the most unfavourable place of exposure outside the plant premises. Whereas (at least for the parameter combination taken as a basis in this connection) the simple model leads to conservative values for a 50-m release height beyond a fence distance of little more than 100 m, an underestimation of the environmental exposure is obtained with this model for higher levels of release. In connection with a source distance of the fence of 100 m, this underestimation of the maximum value at the most unfavourable point of impact amounts to a factor of 10 for a 100-m release height, and to a factor of 57 for a 200-m release height. For 2.5 km source distance of the fence, conservative values are obtained for a 100-m release height even by means of the simple model, whereas for a 200-m release height, the environmental impact in comparison with the modified model is still underestimated by a factor of 2.4.

A safe assessment of the tritium contamination of plants may also be attained when the tritium concentration of the air computed at release level instead of near the ground is substituted for the specific activity model, i.e. when the exponential function in Eq. (2) is made equal to one (Fig. 3). However, this
FIG. 3. Comparison of different specific activity models for calculation of HTO contamination of vegetation for a release height of 100 m.
method will lead to an overconservative assessment of the environmental impact in source proximity.

6. SUMMARY

The concept provided for by national regulations [1, 2] for the purpose of calculating the tritium ingestion dose according to the simple specific activity model, Eq. (1), will lead only to a conservative assessment in source proximity for low levels of release. Particularly for release heights above 50 m, it should be replaced by the modified specific activity model, Eq. [11], which provides a more realistic description of the contributions of a plant's HTO uptake with the soil water, from air humidity and via precipitation moisture, taking into account the local conditions of precipitation as a function of the wind direction sector.

A conservative alternative based on the simple specific activity model is also provided by application of the air activity concentration computed at release level instead of using the values near ground level, although this may lead to an overestimation of more than one order of magnitude in source proximity.

The general model, Eq. (3), (conceived for aerosols) may also be used, provided that, after minor variation of the model (cf. Section 3) 'effective' values for the deposition velocity $v_g$ and retention factor $f$ are used, adapted to the modified specific activity model.

It is evident that all these models, which are based on constant release rates and radio-ecological equilibrium conditions, can only give an approximate description of the true circumstances in each specific case. Determination of the environmental impact from tritium (and other radionuclides) according to a dynamic model will not only require much more attention to models but is also problematic because of lack of knowledge regarding the input parameters required, which, in many instances, are dependent on time.

REFERENCES

DISCUSSION

R. KIRCHMANN: I should just like to point out to users of models that whereas cattle may be grazing for six months of the year, during the other six months they are ingesting silage, particularly hay and maize, which contains tritiated organic matter.
ENVIRONMENTAL TRANSPORT AND LONG-TERM EXPOSURE FOR TRITIUM RELEASED IN THE BIOSPHERE

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Abstract

ENVIRONMENTAL TRANSPORT AND LONG-TERM EXPOSURE FOR TRITIUM RELEASED IN THE BIOSPHERE.

Global cycling of tritium is studied with regard to long-term exposure and dose. Dose and dose commitment are calculated for releases at different latitudes to the troposphere, land and upper ocean layer, with particular regard to effects from release into recipients of intermediate size as, for example, the Baltic Sea. The global transport of tritium appears to be governed by first order kinetics. Compartment models based on linear differential equation systems, as used in this study, should therefore be adequate. The realism and applicability of ecological compartment models are analysed with respect to completeness of the systems of reservoirs and pathways as well as accuracy in assumed reservoir sizes and exchange rates. By introducing different biospheric reservoirs and transfer mechanisms, important carriers and recipients are identified for the analysis of tritium released to air, land and water. Terrestrial biota and groundwater are shown to be significant both with regard to reservoir sizes and influence on the land-troposphere and land-sea exchange of tritium. Model studies regarding the conversion of HT to HTO in different biospheric reservoirs indicate that an atmospheric release of HT may yield up to 1.7 times the dose commitment obtained after release of the same amount of tritium as HTO. The global collective dose commitment from a tropospheric release of tritium is $0.002 - 0.004$ man-rem per Ci depending on the latitude at the release point. Release to the surface ocean layers gives a ten times lower collective dose.

1. INTRODUCTION

The nuclear industry gives rise to man-made sources of tritium. Nuclear weapons testing and cosmogenic radiation also contribute to the global inventory. At present tritium from nuclear explosions dominates the exposure of man. It is expected, however, that the nuclear industry will dominate the global tritium inventory shortly after the year 2000. In this work cycling of tritium in the biosphere is studied with regard to the long-term exposure and doses to global populations. As the biospheric transport of tritium appears to be governed by first order kinetics, compartment models based on linear differential equation systems should be adequate.
The completeness of the systems of reservoirs and pathways, as well as accuracy in assumed reservoir sizes and exchange rates, has a bearing on the 'realism' and applicability of ecological compartment models. We have analysed such factors using models developed from two different strategies based on separation of the biosphere into four latitudinal zones or linking of a sequence of regional (i.e. covering the source region), and intermediate (Baltic) ecosystems to the rest of the biosphere.

The use of the biospheric compartment models is discussed in the light of sensitivity tests for evaluating the consequences of uncertainties in the transfer parameters and comparisons of predicted and measured environmental levels of tritium.

2. NATURAL AND ANTHROPOGENIC SOURCES OF TRITIUM

The present biospheric inventory of tritium results primarily from natural atmospheric sources, nuclear explosions and the nuclear industry.

2.1. Natural sources

Nuclear reactions induced by cosmogenic radiation in the atmosphere are the main source of natural tritium generation. Calculated production rates are still rather uncertain but indicate a global inventory of about 20 – 40 MCI [1, 2]. The average concentrations of tritium of natural origin are about 3 pCi/ltr in ocean surface waters [3], 4 pCi/ltr in precipitation [4] and 6 – 24 pCi/ltr in continental waters [3].

2.2. Nuclear explosions

From nuclear explosions about 300 MCI have been injected into the atmosphere [5]. Residual tritium from nuclear weapons testing thus still dominates with more than one order of magnitude over the inventory from natural sources.

2.3. Nuclear power

The tritium content in spent light water reactor (LWR) fuel is estimated to be 23 Ci/MW(e)·a [5]. A fraction of this is discharged as airborne or liquid effluents from the power plants (typically 1.6 Ci/MW(e)·a and 0.12 Ci/MW(e)·a for pressurized water reactors (PWR) and boiling water reactors (BWR) respectively). At reprocessing all but 10% of the remaining tritium seems to be released [6].
With present forecasts for nuclear power growth [7], tritium production in LWR will dominate about the year 2000 over the total contribution from other sources.

Nuclear fusion, as discussed for future applications in controlled thermonuclear reactors (CTR), involves considerable amounts of tritium. Depending on reactor design, inventories will fall within the interval 0.02 – 0.2 MCi/MW(e) · a [8] and the release has been estimated to be about 2 Ci/MW(e) · a.

3. TRITIUM IN THE BIOSPHERE

Cycling of hydrogen in the biosphere, i.e. that part of the earth inhabited by living species, is governed by exchange and transport of water, which is its main carrier. The turnover of water in systems of land, sea and biota thus affects the distribution and time of exposure of man in a global perspective.

In regard to long-term exposure from practically continuous sources, the levels established at the steady-state conditions prevalent for tritium of natural origin are of interest.

Information about tritium concentration and the biospheric tritium inventory before nuclear weapons testing is limited, however. Because of the present dominating anthropogenic sources of tritium, precise estimates of global averages are still lacking. Furthermore, there is a significant spread in estimated rates of production in the atmospheres from cosmogenic radiation. Dose calculations based on extrapolations from measured natural tritium levels may therefore yield unsatisfactorily low precision in results. Steady-state levels are also insufficient for estimations of concentrations in the transient state lasting some decades after a release in the biosphere. For the study of biospheric turnovers and exposure to tritium during that period, theoretical models based on experimental rates for large-scale tritium exchange are useful.

3.1. Mathematical model

A model in which the rate of removal of matter from a reservoir is only dependent on the quantity Y present in the reservoir is described by first order kinetics. The outgoing flux is then the product of Y and the transfer rate coefficient k. Global cycling of tritium appears to be governed mainly by first order kinetics. Compartment models based on linear differential equation systems with constant transfer coefficients, as used in this study, should therefore be adequate.

Compartment principles and first order kinetics means that:

(a) The outflow for a reservoir 'j' is only dependent on the quantity y_j of radionuclides.
(b) The reservoir is momentarily well mixed.
(c) Each atom, molecule or other elementary particle has the same probability of leaving the reservoir.
(d) The rate of transfer is independent of time.

The model is adapted for calculation of a simple mother-to-daughter decay chain [9] or a chemical conversion as for the processes converting HT to HTO.

The relationship between the quantities of tritium activity in the reservoir system is expressed mathematically in vector form as

$$\dot{y}_M(t) = K_M y_M(t) + Q_M(t) - \lambda_M y_M(t) - C_M y_M(t)$$

for the primary or mother species and

$$\dot{y}_D(t) = K_D y_D(t) + C_M \cdot y_M(t) - \lambda_D y_D(t)$$

for the secondary or daughter species.

The vectors $y$ and $\dot{y}$ refer to the activity and activity change per unit of time in the separate reservoir parts of the system at time $t$.

The coefficient matrices $K$ (year$^{-1}$) and $Q(t)$ (activity/year) describe the rates of transfer between respectively productions and releases inside the reservoir. For the daughter species, the source strength (activity/year) in each reservoir is a function of the quantity of mother species in that reservoir. The decay constant $\lambda = \ln 2/T_{1/2}$ where $T_{1/2}$ is the physical half-life, 12.3 years, of tritium. $C_M$ is the chemical conversion rate, e.g. for the HT to HTO transfer in the atmosphere.

3.2. The biospheric system

Comparisons of exposure and dose calculations based on models of varying complexity allow evaluation of the importance of the different reservoirs and networks introduced. Such comparisons of results from more or less complete systems indicate qualitatively the relevance of a given model for a given purpose. In this work a set of compartment models has been developed in order to study the significance of models of different ecological complexity and global subdivisions.

The following aspects are considered with regard to population doses:

(a) A ground- or soil-water reservoir introduced in a global compartment system comprising reservoirs for atmosphere, hydrosphere and terrestrial biota.
(b) Separate reservoirs for long- and short-lived terrestrial biota.
(c) Organically bound tritium in the biota reservoirs and tissue water fraction.
(d) An integral global ecosystem or subdivision in four zones for the $0^\circ - 30^\circ$ and $30^\circ - 90^\circ$ latitude bands on both hemispheres.

(e) A sequence of three ecosystems covering the source region, intermediate system (the Baltic) and the global biosphere.

(f) Conversion of HT to HTO and its bearing on long-term exposure.

3.3. Rates of turnover and reservoir sizes

Transfer rates (Tables I and II) have been calculated for exchange of HTO between reservoirs in the global system and the regional and intermediate systems respectively. Estimated values of the masses of the different reservoirs in terms of kilograms of water are listed in Tables III and IV. Transfer coefficients for the atmospheric carrier system as well as the global sea system are the same as used in a previous study of global carbon-14 [10] and iodine-129 [11].

3.3.1. Atmosphere

The water content of the troposphere decreases with altitude [12]. The inhomogeneous distribution with height is considered in relation to the amount of HTO and specific activity in the lower $0 - 2$ km of the troposphere. Atmospheric HT, however, has a practically constant mixing ratio invariant with height [13, 14]. The residence time for HT in the atmosphere, which appears to be $4 - 10$ years [13, 14], is governed by chemical conversion in the atmosphere as well as microbiological processes in the atmosphere-earth interface.

3.3.2. Exchange between atmosphere and sea

HTO is transferred from the troposphere to the sea mainly by precipitation and vapour exchange, and returns to the troposphere by evaporation.

There are different opinions about the importance of vapour exchange in comparison with precipitation [15 - 17]. However, calculated transfer rates based, as in this work, on the global water balance [18] implicitly cover transfer by both mechanisms. The estimated average tropospheric residence time of about 10 days (Table I) for HTO in the troposphere is in good agreement with other published data [15 - 17, 19, 20].

The variation in transfer rates between the four latitude bands (Table I) reflects the precipitation and evaporation data for the particular zone.

Mass balance calculations indicate a certain transfer of molecular hydrogen from the oceans to the atmosphere [14]. We have assumed that the transfer in the opposite direction is governed by solubility and rate of gas exchange due to wind speed as in the previous study of carbon-14 [10]. The transfer of molecular hydrogen from the troposphere to the well-mixed sea is thus derived from the corresponding transfer rate for carbon dioxide weighted with the ratio of the respective solubilities.
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<td>Troposphere – Short-Lived Biota</td>
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<td>Land – Long-Lived Biota</td>
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<td>G\textsuperscript{b}</td>
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<td>SEA AND SEA-LAND INTERFACE</td>
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<td>Well-Mixed Layer – Well-Mixed Layer</td>
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<tr>
<td>Deep Sea – Well-Mixed Layer</td>
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\textsuperscript{a} These values were selected so as to comply with the range of values given in the references and to conserve mass balance within the compartments.

\textsuperscript{b} Global average.
### TABLE II. TRITIUM TRANSFER COEFFICIENTS IN THE REGIONAL AND INTERMEDIATE MODEL

<table>
<thead>
<tr>
<th>Zone</th>
<th>$K_{ij} \text{ (a}^{-2})$ (assumed value)$^a$</th>
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</thead>
<tbody>
<tr>
<td><strong>ATMOSPHERE</strong></td>
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<td>Atmosphere $\rightarrow$ Global troposphere</td>
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<td>Global troposphere $\rightarrow$ Atmosphere</td>
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<td>Atmosphere $\rightarrow$ Short-lived biota</td>
<td>$2.4 \times 10^6$</td>
</tr>
<tr>
<td>Short-lived biota $\rightarrow$ Atmosphere</td>
<td>42</td>
</tr>
<tr>
<td>Atmosphere $\rightarrow$ Land</td>
<td>$5.3 \times 10^{-2}$</td>
</tr>
<tr>
<td>Land $\rightarrow$ Atmosphere</td>
<td>$3.3 \times 10^1$</td>
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<tr>
<td>Atmosphere $\rightarrow$ Baltic Sea</td>
<td>$2.5 \times 10^{-1}$</td>
</tr>
<tr>
<td>Baltic Sea $\rightarrow$ Atmosphere</td>
<td>$1.2 \times 10^3$</td>
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<td><strong>REGIONAL BALTIC INTERFACE</strong></td>
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<td>Land $\rightarrow$ Short-lived biota</td>
<td>$6.1 \times 10^{-5}$</td>
</tr>
<tr>
<td>Land $\rightarrow$ Baltic Sea</td>
<td>$8 \times 10^{-4}$</td>
</tr>
<tr>
<td>Baltic Sea $\rightarrow$ Well-mixed sea</td>
<td>$4.3 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

$^a$ These values were selected so as to comply with the range of values given in the references and to conserve mass balance within the compartments.
TABLE III. GLOBAL COMPARTMENT MASSES

<table>
<thead>
<tr>
<th>Biospheric unit</th>
<th>Compartment masses (kilogram water)</th>
<th></th>
<th></th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Zone</td>
<td>30°N–90°N</td>
<td>0°N–30°N</td>
<td>0°S–30°S</td>
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<td>4.4 × 10^{15}</td>
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</tr>
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<td>6.0 × 10^{18}</td>
<td>5.0 × 10^{18}</td>
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<td>5.5 × 10^{16}</td>
<td>3.6 × 10^{16}</td>
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<td>6.2 × 10^{14}</td>
<td>6.2 × 10^{14}</td>
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<tr>
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<td>3.0 × 10^{20}</td>
<td>3.0 × 10^{20}</td>
<td>5.0 × 10^{20}</td>
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</table>

TABLE IV. REGIONAL COMPARTMENT MASSES

<table>
<thead>
<tr>
<th>Biospheric unit</th>
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<td>Regional area</td>
<td>Baltic area</td>
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<td>Atmosphere (&lt;1 km)</td>
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<td>Common</td>
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<tr>
<td>Short-lived biota</td>
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<td>6.2 × 10^{18}</td>
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<td></td>
</tr>
<tr>
<td>Land</td>
<td></td>
<td>2.2 × 10^{13}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Baltic Sea</td>
<td></td>
<td>—</td>
<td>2.2 × 10^{16}</td>
<td></td>
</tr>
</tbody>
</table>

3.3.3. Sea

The mean residence time for hydrogen in the form of HT, HTO or organically bound is assumed to be less than ten years (Table I) in the well-mixed layer. Feedback to the atmosphere is about one-fifth of the transfer to deep seas constituting essentially a sink for tritium.

3.3.4. Biota and land

The world biomass [21, 22] corresponds to about 3 × 10^{15} kg tissue-bound H₂O in the dry matter fraction. Of this all but two parts per million is concentrated
on land. We have used a single compartment for short-lived plant and animal biota where tissue-bound water resides about two years. More than nine-tenths of the hydrogen incorporated in terrestrial living matter is assimilated by long-lived biota. The mean turnover time of this biota and its decomposition products in the humus may be about one hundred years [10, 21].

Root uptake and transpiration of water from terrestrial plants constitute an important vector for transport of soil- and groundwater to the atmosphere. The net primary production (NPP) binds by photosynthesis about 0.15% of the water molecules drawn from the soil [18, 21]. Because of transpiration, thus 700 times as much water is transferred to the atmosphere as is bound organically in plant tissue. Long-lived biota gives rise to about two-thirds of the total amount of water transpired by terrestrial plants. In fact evaporation from soil contributes slightly less than the transpiration from biota to the transfer rate calculated for the land-troposphere (Table 1).

In-transit water constitutes about 80% of the plant’s fresh weight. Only hydrogen incorporated in the dry-weight fraction, i.e. the remaining 20%, belongs to the respective reservoirs for short- and long-lived biota. By this assumption it is possible to consider the significantly longer half-life of incorporated tritium compared with that in the transit water component [23]. In the dose calculations, the in-transit water fraction of the plant is assumed to have the same specific tritium activity as soil water in the land compartment.

Transfer of organically bound tritium to the atmosphere, i.e. the reverse of photosynthesis, depends on respiration and decomposition yielding as much carbon atoms as water molecules [21]. Thus the same transfer rates are used as in the study of environmental carbon-14 [10].

Groundwater run-off (Table 1) according to water mass balance studies [21, 24] affects only slightly the time water resides in the root zone layer of about 1.5 m before transfer to the sea. According to our calculations, the total effect of evapotranspiration and run-off corresponds to a global residence time of 2.2 years for water in the land compartment compared with 3 years for transfer by evapotranspiration only.

Molecular hydrogen is consumed at the soil surface by microbiological activity characterized by first order kinetics [14], thereby atmospheric HT may be converted to HTO. The present atmospheric inventory of molecular hydrogen is $1.6 \times 10^{11}$ kg. At the soil surface $1.2 \times 10^{10}$ kg is annually metabolized transferring hydrogen atoms from the hydrogen gas to the water pool at a rate of 0.08 a$^{-1}$.

This biological mechanism appears to be an important link in the ecological pathways to man of tritium released as HT as it transfers hydrogen into the system of soil, water and biota.
FIG. 1. Ecological pathways for exposure to man.

FIG. 2. Global compartment model — typical zone.
4. EXPOSURE PATHWAYS

The significant pathways for intake of water are (Fig.1)

(a) inhalation;
(b) absorption through skin;
(c) food consumption;
(d) drinking water.

Inhalation of 20 litres per minute is the working day average according to the ICRP [25]. As the resting rate 10 litres per minute appears reasonable [26, 27]. We have therefore used 15 litres per minute as an average inhalation rate, i.e. approximately 20 m³ per 24 hours. The uptake of water from inhaled air is assumed to be 100% effective. Water vapour passes through the skin at a rate corresponding to absorption of the vapour in 10 litres of air per minute.

Inhalation and skin absorption thus yield an intake rate corresponding to the water content in 25 litres of air per minute. The water vapour concentration is calculated from the content in reservoir troposphere and atmosphere of the global (Fig.2) and regional model (Fig.3) respectively.

Food contains hydrogen both in tissue water and in organic molecules. The latter may be catabolized yielding water by oxidation. The daily intake by food and water is [26, 27]

<p>| | |</p>
<table>
<thead>
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<tbody>
<tr>
<td>as liquid</td>
<td>1200 cm³</td>
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<tr>
<td>in food</td>
<td>1000</td>
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<tr>
<td>oxidation of food</td>
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<tr>
<td><strong>Total</strong></td>
<td><strong>2.5 litres</strong></td>
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</tbody>
</table>

Drinking and tissue water contain the same tritium concentration as soil water in the land reservoir. Without the land reservoir all the tritium in plants is
taken directly from the atmosphere, and drinking water contains the same specific activity as water vapour. Tritium in the water derived from oxidation of food is taken in with a specific activity equal to that in the short-lived biota reservoir.

Exposure for tritium gas (HT) may occur by inhalation, but intake of tritium gas through the skin is negligible [8]. Only 1.6% of the tritium gas inhaled enters the bloodstream [27], the remainder is exhaled directly. HT is to some extent oxidized to HTO in living tissue. However, less than 0.04% of the tritium gas inhaled is retained in man as tritiated water [23, 27]. Other studies indicate that equivalent rates of tritium appear in the body fluids provided the specific activity of HTO in ambient air is about 15,000 times that of HTO. We have assumed an uptake in tissue of tritium gas corresponding to the fraction $4 \times 10^{-4}$ of the amount in the inhaled air.

5. RADIOLOGICAL ASPECTS

Tritium decays to helium-3 by emission of a beta particle of maximum 18 keV kinetic energy and maximum 0.01 mm range in tissue. A single cell layer constitutes an efficient barrier for the emitted beta particle. Thus external radiation from tritium is a negligible hazard to man.

The weight of body water is 43 kg. At a daily intake of 2.5 kg of water the mean residence time of a water molecule in man is 17 days, i.e. corresponding to a biological half-life of 12 days. With regard to the different behaviour of hydrogen in tissue fluids and organic molecules a single compartment will not give a satisfactory description of tritium turnover in man. Other models based on three compartments have been suggested [28, 29] with half-lives of about 9, 30 and 450 days. Various dose estimations fall within the interval of 84 – 167 mrad per 1 mCi of intake [25, 27 –29]. We have assumed the upper limit of this interval which is the dose according to the ICRP.

The dose from tritium gas has been based on the same dose factor as for tritiated water. However, only that fraction, $4 \times 10^{-4}$, of the inhaled tritium gas taken up and retained in the body as tritiated water or organic molecules is considered.

6. DOSE AND DOSE COMMITMENT

The average dose and dose commitment on a global scale have been calculated using alternative models in order to study the long-term exposure and dose distribution from different aspects. The completeness of the model with regard to the long-term exposure is studied by the use of reservoir systems with and without a reservoir (i.e. land in Fig.2) for ground- and soil-water.
FIG. 4. Dose commitment with and without land compartment for release of tritium in the stratosphere, troposphere or well-mixed layer in the zone 30°–90°N. Values are in percentage of dose with release in the troposphere in the model containing land compartment.

FIG. 5. Dose commitment for release of tritium in various biospheric compartments in the zone 30°–90°N. Values are in percentage of dose with release in the land compartment.
The global dose distribution is calculated based on (a) either a global model comprising the four zones 90° – 30°N, 30° – 0°N, 0° – 30°S and 30° – 90°S, according to Fig.2, or (b) a model sequence covering the source region and the Baltic area, according to Fig.3, as a complement to the global biosphere. In this model sequence the global biosphere is not separated into different zones but is based on the same reservoir system as in Fig.2.

The global dose commitment from a release of tritiated water in the stratosphere, troposphere or well-mixed sea layer in zone 30° – 90°N has been calculated using the model of type (a) with and without land compartments. The dose commitment in percentage of the dose with release in the troposphere in the model containing the land compartment is illustrated in Fig.4.

The global dose commitment is twice as high with the assumed transfer rates (Table 1) and a system with land reservoirs (implying a half-time of water of about 2.5 years in all but the long-lived biota of the terrestrial ecosystem), than when terrestrial ground- and soil-water is neglected. It is notable that the same is true also for release to the sea. This indicates the significance of feed-back to the terrestrial environment regarding the long-term exposure.

The importance of a land reservoir is also apparent from Fig.5, illustrating the average dose commitment for release in zone 30° – 90°N in the stratosphere, troposphere, land or well-mixed layer with values in percentages of dose with release in the land reservoir.

About a three times higher dose is obtained with release to the land instead of release to the atmosphere. With regard to the low dose following release into the sea, the ratio between doses with release in the troposphere and land respectively should approximately reflect the fraction of atmospheric tritium washed out over the land surface.

The residence time for a water molecule in the troposphere is about 10 days. Thus the vertical transfer to the terrestrial and aquatic ecosystems at the same latitudes as the release occurs before substantial atmospheric mixing is possible.

The very skewed distribution of the tritium inventory following a release from confined sources is seen from measurement of the latitudinal distribution of tritium in surface waters of the Pacific from nuclear weapons testing [5].

The concentrated load over the same latitude where release occurs is also apparent from our calculations with the model of type (a) using zonal separations. Figure 6 illustrates the global distribution of dose commitment from a release at 30° – 90° N in the troposphere, land or well-mixed sea.

The global dose commitment from a one-year one-Ci release in various reservoirs using a model of type (a) with a land reservoir is portrayed in Fig.7. Maximum dose commitment refers to the average dose in any of the four latitudinal zones. The global average is calculated with due weight given to all four zones.

The global collective dose commitment has been calculated with regard to the different population growth rates in the four latitude bands. Depending on
FIG. 6. Dose commitment in different zones for release of tritium in the zone 30° – 90°N. Values are in percentage of the maximum dose with release in the land compartment.

FIG. 7. Dose commitment ($10^{14}$ rem) from 1 Ci releases (at 1 Ci/a) of tritium in various reservoirs and latitudinal zones.
the release latitude, the global collective dose commitment ranges from 0.0024 to
0.0035 man·rem per Ci released to the troposphere (Fig. 7) as HTO. With the
tritium source in the well-mixed layer of the sea, the collective dose commitment
is $1.6 \times 10^4$ man·rem per Ci.

Preliminary studies have been made using model type (b). Release of HTO
into the Baltic sea yields a collective dose commitment of $2.1 \times 10^5$ man·rem
per Ci from consumption of fish from the Baltic area.

The water exchange with the Atlantic Ocean gives rise to another
$7.0 \times 10^4$ man·rem due to the tritium release entering the ocean from the
Baltic. Evaporation from the Baltic sea surface leads to the dominating exposure
yielding $1.0 \times 10^4$ man·rem due to the one-Ci release to the Baltic. Thus the
intermediate recipient in this case increases the global dose by 12% to a total of
$1.9 \times 10^4$ man·rem/Ci.

For airborne releases, the contribution from the regional to the global
collective dose commitment is about 1%.

HT released to the atmosphere is slowly transferred to HTO by processes
in the atmosphere and at the continental soil surface (see Section 3.3). Provided
the microbiological process at the air soil interface has not an equally efficient
counterpart at the sea surface, a selective transfer of tritium into the terrestrial
environment is the consequence of the microbiological activity. In fact, by use of
the 0.08 a⁻¹ transfer rate for conversion of HT to HTO or organic compounds
in surface soil, and a net residence time in the troposphere of 4 – 10 years, the
dose per Ci tritium released as HT to the atmosphere should be from 1.3 to
1.7 times that of the release as HTO. This should be of considerable interest
with respect to health hazard evaluations of nuclear fusion power.

7. ACCURACY AND CONTROL OF MODEL CALCULATIONS

The main uncertainties in the calculated global exposure are probably caused
by the half-time and reservoir sizes for tritium in the terrestrial environment.
In comparison the hydrological cycles in the aquatic and atmospheric environments
are fairly well known.

Estimations of the mass, NPP and transpiration with respect to terrestrial
biota affect the specific activity of tritium in the assumed organic hydrogen pool,
rate of incorporation and transport of tritium from the soil to the atmosphere.
However, provided that water does not constitute a limiting factor, mass, NPP and
transpiration are strongly correlated. Thus on a global scale it is reasonable to
expect the uncertainties introduced from all three factors to be predictable from
the uncertainties in, for example, the mass of terrestrial biota. This mass in terms
of water weight is expected to be within the range $(1 - 10) \times 10^{15}$ kg, i.e. within
a factor three of the value used in this study. The contribution (0.3 kg per day)
from oxidation of food is slight in comparison with the amount of drinking water
(1.2 kg per day) and in-transit water in food (1.0 kg per day). The factor of three uncertainty in biota reservoir size and NPP, which both affect the assumed tritium intake from oxidation of food, should thus be of minor importance for precision in dose.

The land reservoir, on the other hand, significantly influences the calculated dose and dose commitment (Fig.4). Apart from the factor of two increase in dose when the land reservoir is included (Fig.3), there are other potentially important variations. We have assumed water in the land reservoir to belong to the upper 1.5 m in soil. Although tritium will be found much deeper below the soil surface, the specific activity decreases considerably with depth. Furthermore, the product of reservoir size and residence time, which is the important quantity regarding the integral exposure, probably varies relatively little. A smaller reservoir size, associated with higher specific tritium activity, should imply a faster turnover which decreases the time over which significant exposure may occur. We do not know, however, the effect of these uncertainties.

The biospheric tritium inventory at steady-state conditions with constant release in the troposphere is as follows, using the model with the land reservoir according to Fig.2:

<table>
<thead>
<tr>
<th>Reservoir</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmosphere</td>
<td>3.8%</td>
</tr>
<tr>
<td>Land</td>
<td>29</td>
</tr>
<tr>
<td>Sea</td>
<td>67</td>
</tr>
<tr>
<td>Biota</td>
<td>0.03</td>
</tr>
</tbody>
</table>

These values conform well with the results from Lal and Peters [1] estimating that 27 and 65% of the tritium inventory is to be found in the land surface and oceans respectively from the natural production of tritium in the atmosphere.

In order to compare model predictions with measurements, we have simulated the natural production of tritium by using sources of equal strength in each of the four stratospheric reservoirs in the model system in Fig.2. The average dose according to our calculations is $2.9 \times 10^{-13}$ rem per Ci released in the stratosphere. Assuming that the tritium concentration in the body is the same as in continental surface waters before anthropogenic sources of tritium became significant (see Section 2), the dose should be in the range of 1.3 to 5.3 μrad. Based on a global inventory of 20 – 40 MCi from natural sources, our estimate of this dose is 5.8 to 12 μrad.

An estimation has also been made of the global tritium inventory in the sea, land, biota and atmosphere reservoirs using the reservoir sizes in Table II and the measured tritium levels in sea, continental surface waters (for land and biota), and precipitation respectively. Normalized to a total biospheric inventory of 30 MCi tritium the levels are: sea 200,%; land 5 – 20,%; biota 0.04 – 0.14,%; and atmosphere 0.2,%. 
With due regard to the uncertainties in the natural tritium inventory and pre-nuclear weapons testing concentrations in continental surface waters, there is a satisfactory fit between predicted and measured levels.

REFERENCES

METHODOLOGY FOR HAZARD ASSESSMENT OF ENVIRONMENTAL TRITIUM

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Abstract

METHODOLOGY FOR HAZARD ASSESSMENT OF ENVIRONMENTAL TRITIUM.
The assumption is made that the ecological system immediately follows the variations of the tritium content in atmospheric water vapour. Tritium concentrations in the ecological subsystems (plant, animal and human) are calculated using balance equations for tritium intake and elimination. Due to consumption of non-contaminated food and water the tritium concentrations in animals and man are reduced by about the ratio of contaminated to non-contaminated food. The main exposure pathways are shown for several consumption habits. Since West Berlin imports a considerable amount of its food (more than 30%) from Niedersachsen (Lower Saxony), the proposed area for the West German nuclear fuel reprocessing plant, the exposure of the population of this city is used as an example.

1. INTRODUCTION

Tritium dose-rate calculations have to specify the concentration \( c_h \) (unit: nCi/litr of body water, both as tissue water and non-aqueous tritium expressed as water) of tritium in human tissue. The dose rate \( H \) (mrem/a) is then related to \( c_h \) by the well-known relationship

\[
H = 0.1 \times Q \times c_h
\]  

(1)

\( Q \) is the quality factor for the beta radiation from tritium. For exposure to tritiated water vapour, Anspaugh and co-workers [1] have developed a dynamic model describing the tritium pathways among several compartments. Their model includes transfer rates and elimination rates (time constants).
In this study this dynamic method is reduced to a dynamic equilibrium approach which consequently does not need the time constants mentioned. It takes into account the mixing of contaminated and non-contaminated food (including water intake and water vapour absorption by the skin).

2. SYSTEM OF PATHWAYS

Figure 1 presents the well-known system of pathways of hydrogen. Each compartment may have to be divided into several subcompartments, i.e. may have several hydrogen pools with separate hydrogen pathways connected. The compartment system is assumed to be a linear time-invariant system, i.e.

(a) the superposition principle holds (e.g. doubling an input will double the pool content);

(b) the coefficients describing the amounts of input and output are independent of seasonal changes or weather influences.

The system has a tritium source in the air compartment. The tritium follows the non-radioactive hydrogen (protium) on all pathways into all compartments, and no process will enrich tritium with respect to protium. The behaviour of the tritium is then described on the basis of the following equations:

\[
\begin{pmatrix}
\dot{c}_p(t) \\
\dot{c}_a(t) \\
\dot{c}_h(t)
\end{pmatrix} =
\begin{pmatrix}
-\lambda_p & 0 & 0 \\
\alpha_a/M_a & -\lambda_a & 0 \\
\alpha_h/M_h & \beta_h/M_h & -\lambda_h
\end{pmatrix}
\begin{pmatrix}
c_p(t) \\
c_a(t) \\
c_h(t)
\end{pmatrix} +
\begin{pmatrix}
R/M_p \\
(\beta_a + 2\theta_a f)/M_a \\
(\gamma_h + 2\theta_h f)/M_h
\end{pmatrix} c_w(t)
\]

\[
d/dt \vec{c}(t) = A \vec{c}(t) + B \vec{c}_w(t)
\]

where

- \( c \) = concentration of tritium in the water of compartment i (nCi/ltr)
- \( R \) = rate of rainwater consumption by plant (litre/a)
- \( I \) = rate of consumption of irrigation water (litre/a)
- \( M \) = mass of body water of animal (subscript a) or man (h) (litre)
- \( f \) = absolute humidity of air = \( 5 \times 10^{-3} \) litre/m\(^3\)

Greek letters mean intake rates (kg/a or kg/d) into the compartments denoted as subscripts of the letters. For further explanation see Fig.1. The elements \( \lambda_i \) in the diagonal of the matrix \( A \) give the time behaviour of the free system, i.e. the time constants of the system. Vector \( B \) specifies the coupling of the driving force \( c_w(t) \) to the system of food pathways.
Since tritium concentrations in air and rain vary through varying emissions and weather, it is important to know the response of the system $A$ to periodic disturbances. The response of a single unconnected compartment $i$ to a force $c_\omega(t) = \sin(\omega t)$ can be described by a transfer function $g$, which relates the concentration of the compartment $i$ to the concentration in the environment:

$$c_i(t) = b_i / |g_i(\omega)| \sin(\omega t - \phi)$$

$$|g_i(\omega)| = 1 / \sqrt{\lambda_i^2 + \omega^2} = \begin{cases} 
1 & \text{for } \omega / \lambda_i = 1 \\
0.7 & \text{for } \omega / \lambda_i = 0.1 \\
0.1 & \text{for } \omega / \lambda_i = 0 
\end{cases}$$

$$\tan \phi = \omega / \lambda_i$$

One can see that if tritium concentrations in the air change slowly ($\omega < 10\lambda_i$), the compartment concentrations follow those changes closely. The amplitudes of the system remain of the same order of magnitude for $0 < \omega < 10\lambda_i$.

Anspaugh and co-workers [1, 2] report time constants $1/\lambda_p$ for the system: tritiated water vapour-plant-soil ranging from 1 hour for tritium in tissue water to several days for tissue-bound tritium, the tritium reaching the plants both
from the air and the soil. The impulse responses measured for impulses of height $c_w^0$ and $\frac{1}{2}$ to 1 hour duration were

$$c_p(t = 0) \approx |g_p(\omega)|/c_w^0$$

(5)

for $\omega = 1 \ldots 2$ h$^{-1}$, $t =$ post-irradiation time and with

$$0.2 < |g_p(\omega)| < 0.8$$

(6)

which is not in contradiction with their time constants (see Ref.(3)).

Rosinski and Winter [3], and Lee and co-workers [4] recorded tritium concentrations in air, soil and in tissue water of several plants in the vicinity of the Karlsruhe Nuclear Research Center during seasons of late summer to winter. Although a thorough Fourier analysis of their measurements has not been performed, their data seem to point towards $|g_p(\omega)|$-values above 0.5 for very small $\omega$, $1/d < 1/\omega < 2$ m.

3. STATE OF DYNAMIC EQUILIBRIUM

In this study we will take $|g_p(\omega)| = 1$, which will be undoubtedly conservative depending upon how far the system is from stationariness, i.e. from the case $\omega < \lambda_1$ ('state of dynamic equilibrium').

There is another overestimation in this work, namely the omission of the subdivision of the plant and animal compartments. Although some transfer functions $g(\omega)$ of this system of subcompartments are small, their contribution to the food, however, is included with unreduced strength in our calculations, i.e. it is supposed that $g = 1$ for all subcompartments.

The concentrations $c_t$ will be calculated for a stationary system. That means that the equation of state (2) will reduce to a balance equation of the type:

$$\text{rate of intake of tritium} = \text{rate of elimination of tritium}$$

(7)

For plants the following relation is used ($M_p$ disappears for the stationary system):

$$R \ c_w = (R + 1) \ c_p$$

(8)

c$ _p$ is smaller than $c$ $w$ whenever a non-contaminated source of water is used, because the plant is not supposed to discern between tritium and protium.
TABLE I. POSSIBLE MAXIMUM CONTRIBUTIONS OF THE FOOD PATHWAYS TO THE DOSE RATE
*Numbers do not add up to 1 because of rounding errors*

<table>
<thead>
<tr>
<th>Consumption habit of</th>
<th>Plant</th>
<th>Animal</th>
<th>Water</th>
<th>Inhalation and skin absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Child [5]</td>
<td>0.37</td>
<td>0.25</td>
<td>0.34</td>
<td>0.05</td>
</tr>
<tr>
<td>Average</td>
<td>0.27</td>
<td>0.28</td>
<td>0.35</td>
<td>0.09</td>
</tr>
<tr>
<td>Maximum Teenager [5]</td>
<td>0.39</td>
<td>0.27</td>
<td>0.30</td>
<td>0.04</td>
</tr>
<tr>
<td>Average</td>
<td>0.29</td>
<td>0.31</td>
<td>0.31</td>
<td>0.08</td>
</tr>
<tr>
<td>Maximum Adult [5]</td>
<td>0.32</td>
<td>0.23</td>
<td>0.41</td>
<td>0.04</td>
</tr>
<tr>
<td>Average</td>
<td>0.23</td>
<td>0.24</td>
<td>0.45</td>
<td>0.08</td>
</tr>
<tr>
<td>Anspaugh [1]</td>
<td>0.13</td>
<td>0.18</td>
<td>bal.</td>
<td>0.06</td>
</tr>
<tr>
<td>2.5SVO [6]</td>
<td>0.22</td>
<td>0.18</td>
<td>bal.</td>
<td>0.08</td>
</tr>
</tbody>
</table>

TABLE II. CONSUMPTION RATES FOR MAN (AFTER REFS [5, 7])
*Meat and milk are calculated together: the values for West Berlin are averages over the whole population of 2 million inhabitants*

<table>
<thead>
<tr>
<th></th>
<th>$\sigma_h + \delta_h$</th>
<th>$\phi_h + \epsilon_h$</th>
<th>$\gamma_h + \xi_h$</th>
<th>$2\theta_h$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Child</td>
<td>550 (200)</td>
<td>370 (210)</td>
<td>510 (260)</td>
<td>70 (70)</td>
</tr>
<tr>
<td>Teenager</td>
<td>670 (240)</td>
<td>470 (260)</td>
<td>510 (260)</td>
<td>70 (70)</td>
</tr>
<tr>
<td>Adult</td>
<td>580 (190)</td>
<td>420 (200)</td>
<td>730 (370)</td>
<td>70 (70)</td>
</tr>
<tr>
<td>West Berlin</td>
<td>(225)</td>
<td>(127)</td>
<td>(175)</td>
<td>70</td>
</tr>
</tbody>
</table>
TABLE III. CONSUMPTION RATES (kg/d) FOR ANIMALS (AFTER REF. [5])

Only beef/cattle considered

<table>
<thead>
<tr>
<th>$\alpha_a + \gamma_a$</th>
<th>$\beta_a + \delta_a$</th>
<th>$2\theta_a f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>60</td>
<td>5</td>
</tr>
</tbody>
</table>

The balance equations for the other compartments will be obtained in the same way and may be interpreted similarly.

\[
c_a/c_w = \left(\alpha_a(c_p/c_w) + \beta_a + 2\theta_a f\right)/S_a
\]

(9)

\[
c_h/c_w = \left(\alpha_h(c_p/c_w) + \beta_h(c_a/c_w) + \gamma_h + 2\theta_h f\right)/S_h
\]

(10)

The total intake of food (including water) is called $S_i$ here. It corresponds to $R + 1$ in the case of plants:

\[
S_a = \alpha_a + \beta_a + 2\theta_a + \gamma_a + \delta_a
\]

(11)

\[
S_h = \alpha_h + \beta_h + \gamma_h + 2\theta_h + \delta_h + \epsilon_h + \xi_h
\]

(12)

Since $c_h$ is related to the dose rate via Eq.(1), the dose rate consists of four contributions: each term in the sum (Eq.(10)) gives the contribution of one food pathway.

The amount of tritium entering via a pathway depends on the degree of contamination $c_k$ of the species $k$ of food considered, and on the consumption habits, $\alpha_i/S_i, \beta_i/S_i, \gamma_i/S_i$, etc.

4. RESULTS

The worst case is the complete contamination of the food chain, i.e. when only contaminated food enters the chain ($c_i/c_w = 1$ for all compartments $i$). The four columns of Table I give the fraction of the dose rate for each of the four food pathways. Six different consumption habits are considered: consumption rates for children, teenagers and adults, values both for maximum and average intake used, as they are specified for 10CFR Part 50, App. I [5] (see Tables II and III). Since $c_i/c_w = 1$, the fractions in Table I are from left to right: $\alpha_h/S_h, \beta_h/S_h, \gamma_h/S_h, \text{and } 2\theta_h f/S_h$. 
FIG. 2. Adult's maximum consumption. Integers give the relative flow rates of contaminated and non-contaminated food. Fractions to the left are the first 3 summands of the sum (Eq. (10)). The summands for inhalation (0.04) are given on the inhalation pathways. The numbers in the compartments are $c_w/c_w$ and $c_w/c_w$ respectively.

FIG. 3. Adult's average consumption. Ratios of contaminated to non-contaminated food as given in Fig. 2.
FIG. 4. Adult's maximum consumption: 50% of the food is contaminated.

FIG. 5. Adult's maximum consumption. Here inhalation contributes most of the contamination to man.
FIG. 6. Adult's maximum consumption. Man lives outside the contaminated area (no inhalation of contaminated air by man), and only his milk and meat (beef) stems from the contaminated area. Cattle are fed non-contaminated food only, but breathe contaminated air.

FIG. 7. West Berlin's consumption habits. Transfer of tritium by food imported from Niedersachsen (Ns) to West Berlin causes the build-up of a tritium concentration equal to 15% of the average concentration in atmospheric water in Ns.
For comparison Table I also shows the values calculated on the basis of Anspaugh’s data and those recommended by the Federal German Radiation Protection Commission. Both do not specify the pathway of drinking water. To be consistent with the assumptions underlying the data given in the first six lines, consumption of contaminated drinking water only is assumed here, and the fraction of the dose rate due to that pathway is taken to be 1 (fraction due to plant- and animal-product intake and inhalation).

The values in the line ‘Anspaugh’ were calculated using $S_h = \lambda_h M_h$.

In the last line of Table I the dose rates were first calculated according to the recommendations from Ref. [6]. Then the dose rate was converted to $c_h$ with the help of Eq.(1) and using $c_i/c_w = 1$. Also $c_i/c_w$ was taken to be unity even though the recommendations assign the values $0.75 \times 55/100$ (for milk) and $0.75 \times 55/120$ (for meat).

In the remaining figures various consumption situations of plants, animals and man are considered in more detail, giving more realistic values for $c_i/c_w$, which are denoted in the compartments $i$. The values for the total consumption rates (e.g. $\alpha_h + \delta_h$) again are taken from Ref.[5] and are listed in Tables II and III. They will be referred to as ‘adult’s maximum’ or ‘adult’s average’ in the figure captions.
The compartment system of Figs 2–8 is mainly the same as in Fig.1. The description of the pathways and the compartments could therefore be left out.

Again each food pathway (except inhalation) consists of two parts, one contaminated and one not. The integers L in the figures give the relative contribution of one part compared with the other. That means, for example (see Fig.2),

\[ L^p_h = \alpha_h / \delta_h = 100/1 \]  \hspace{1cm} (13)

and obviously

\[ \alpha_h = (\alpha_h + \delta_h)(1 + (1/L^p_h)) \]  \hspace{1cm} (14)

A fraction denoted at the inhalation pathway and to the left of the plant-, animal- and water-pathway to man is the contribution of the pathway specified to the dose rate (for further explanation see, for example, caption of Fig.2).

The pathways contributing more than 50% of the dose rate are marked by heavy lines.

Figures 2 and 3 stand for consumption of contaminated food only, both by animals and man, except for drinking water for man, which is supposed to come mainly from fountains. The relative contributions of the plant- and animal-pathway have already been given in Table I. The slightly changed consumption habits of the average adult compared with the 'maximum adult' cause the slight change in the ratio \( \alpha_h / \alpha_w \).

In Fig.2 the dose rate is 59% of the value it would be if only contaminated food were consumed, i.e., if the integers at all contaminated pathways were infinite; 32 of those 59% come via the vegetarian pathway.

In Fig.4 an equal mixture of contaminated and non-contaminated food is considered. The dose rate is reduced by about a factor of 2 in this case compared with the case of contaminated food only (Table I).

Figures 5 and 6 show that the inhalation begins to dominate the dose rate when the contaminated food fraction becomes less than 1%.

Figures 7 and 8 have been drawn to show the situation for West Berlin, which obtains about one third of its vegetarian and two thirds of its non-vegetarian food from the province of Niedersachsen (Lower Saxony), where a reprocessing plant for nuclear fuel is planned.

People in West Berlin breathe non-contaminated air. Because of the contaminated air breathed by cattle, a Berliner would (according to these calculations) obtain about 10% of the tritium concentration he would get if he inhaled the tritium while living in Niedersachsen, but without eating contaminated food (see Figs 5 and 8).
If the plants in Niedersachsen grow without irrigation because of a wet summer (Fig. 7), a tritium concentration of 15% of the 'all contaminated food chain value' would be built up in the Berlin population.

During very dry summers the dose rate of the Berlin population is one tenth of the value of wet summers (Fig. 8).

As with any model our model requires measured data throughout its various compartments to confirm its validity. Several assumptions have made this model simple. However, the final test is the confirmation in the environment.

REFERENCES


DISCUSSION

H.J. BURKI: It appears that this model applies primarily to a 'local' source of tritium contamination. It does not seem easily applicable to global contamination. Could you comment on this aspect? For example, could the model be applied to fusion reactor contamination?

A.A. MOGHISSI: As a co-author of the paper I should like to stress that the model does have global applicability because it is based on a specific activity concept and therefore does not require biological half-life data. It considers two compartments, one associated with the tritium source and the other with the environment. The entire system is subsequently homogeneously mixed.
TRITIUM DISTRIBUTION
IN MAN AND HIS ENVIRONMENT

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Abstract

TRITIUM DISTRIBUTION IN MAN AND HIS ENVIRONMENT.

The tritium distribution between 'loose' water tritium and 'organically bound' tritium has been measured in soil, vegetation, food and man. Composited monthly tap water samples have been analysed from 1970 to 1978 inclusive. This study has confirmed that the ratio of specific activity of tritium in the 'bound' fraction to specific activity of tritium in the 'loose' fraction decreases in ascending the trophic levels as previously reported. There appears to be a reduction of a factor of 2-4 in each step from soil, ~12, to vegetation, ~4, to tissue, ~2. Soil, vegetation, food and tissue 'loose' tritium concentrations are similar to values for tap water. This would indicate that equilibrium is established rapidly between the 'loose' compartment and environmental water sources. The 'bound' tritium specific activity in tissue is about the same for each organ within a human subject, suggesting homogeneity of labelling of the 'bound' tritium compartment. It is estimated that the apparent residence time of tritium in surface waters has increased from three to five years. This increase in half-time is probably due to injection of tritium into the atmosphere from nuclear weapons tests.

INTRODUCTION

Recent measurements of tritium distribution in environmental samples have been completed to update our previous studies [1,2]. We have analyzed the tritiated water content, available by freeze drying ('loose' fraction), and the tritium available by combustion ('organically bound' fraction) in soil, vegetation, food and human tissue [3]. Monthly composited tap water samples have been measured from 1970 through 1978.

Studies of tritium contamination at Bikini Atoll conducted in 1963 revealed for the first time that higher specific activities for the 'bound' tritium than the 'loose' tritium were found in various environmental and biological samples [4]. Similar results were observed by Marko et al. [5] for 'easily exchangeable' tritium levels as well as by Koranda [6] and Cohen and Kneip [7]. It was suggested that tritium was incorporated uniformly into biological materials during the high specific activity period of nuclear weapon fallout of the early sixties. Due to longer biological half-residence times of the 'bound' tritium fraction compared to the 'loose' tritium fraction, differences in specific activity were observed.
TABLE I. TRITIUM IN NEW YORK CITY TAP WATER

<table>
<thead>
<tr>
<th>Year</th>
<th>pCi/l</th>
</tr>
</thead>
<tbody>
<tr>
<td>1970</td>
<td>480 ± 70</td>
</tr>
<tr>
<td>1971</td>
<td>350 ± 50</td>
</tr>
<tr>
<td>1972</td>
<td>270 ± 50</td>
</tr>
<tr>
<td>1973</td>
<td>230 ± 50</td>
</tr>
<tr>
<td>1974</td>
<td>190 ± 50</td>
</tr>
<tr>
<td>1975</td>
<td>190 ± 50</td>
</tr>
<tr>
<td>1976</td>
<td>180 ± 50</td>
</tr>
<tr>
<td>1977</td>
<td>190 ± 50</td>
</tr>
<tr>
<td>1978 (½)</td>
<td>200 ± 50</td>
</tr>
</tbody>
</table>

In addition, it was postulated [8] that vegetation may incorporate higher specific activity tritiated water during the accelerated growth period when the spring peak of fallout tritium occurs.

We have reported, previously, the tritium concentrations observed for 'loose water tritium' and 'organically bound tritium' for a variety of environmental and biological samples. Additional measurements have been performed to update the fallout tritium distribution in man and his environment.

RESULTS

Water

New York City tap water has been measured continuously since 1970 and the average annual values are reported in Table I. There is a decrease in the specific activity with time until 1974-1975 when additional nuclear weapons tests added to the atmospheric inventory of tritium, leading to a steady-state tritium concentration at the present time. These results are similar to values reported by others for precipitation and surface water tritium results.

Figure 1 shows the tritium concentration in surface water measured in the Northeastern United States as a function of time. The results from 1961 to 1969 were obtained from the summary by Bennett [9] and our tap water results continued from 1970-1978. For the period from 1967 through 1974, there is a decrease in the tritium concentration with a half-time of about 3.2 years when uncorrected for radioactive decay. If the values are corrected for radioactive decay to 1967, a half-time of 4.3 years is computed.

If our tap water values for 1970 and 1977 are used to estimate a half-time, after decay correction of 1977 results to 1970, a value of 5.2 years is obtained. From 1974 to the present time, the tritium concentrations have remained constant due to the injection of tritium into the atmosphere from nuclear weapons tests. This will increase the apparent half-time.
Soil Samples

Soil samples collected in the Northeastern United States have been measured for 'loose' and 'bound' tritium. The samples were collected to a depth of 25 cm and a representative aliquot was taken for analysis. The results are reported in Table II. Results for a sediment sample collected at Sagamore Lake in upper New York State show similar concentrations of 'loose' and 'bound' tritium compared to the soil measurements.

The results indicate that the 'loose' tritium values are similar in concentration to the present surface water concentration as we have
### TABLE II. TRITIUM IN SOIL AND SEDIMENT

<table>
<thead>
<tr>
<th>Location</th>
<th>Year</th>
<th>‘Loose’ tritium (pCi/l)</th>
<th>‘Bound’ tritium (pCi/l)</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soil</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chester, N.J.</td>
<td>1976</td>
<td>210 ± 40</td>
<td>2900 ± 100</td>
<td>13.8</td>
</tr>
<tr>
<td></td>
<td>1977</td>
<td>200 ± 40</td>
<td>2650 ± 100</td>
<td>13.3</td>
</tr>
<tr>
<td>Lloyd, N.Y.</td>
<td>1976</td>
<td>190 ± 40</td>
<td>2800 ± 100</td>
<td>14.7</td>
</tr>
<tr>
<td>New York, N.Y.</td>
<td>1975</td>
<td>210 ± 40</td>
<td>2500 ± 100</td>
<td>11.9</td>
</tr>
<tr>
<td></td>
<td>1976</td>
<td>180 ± 40</td>
<td>2600 ± 100</td>
<td>14.4</td>
</tr>
<tr>
<td>Sediment</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sagamore Lake, N.Y.</td>
<td>1978</td>
<td>220 ± 40</td>
<td>3500 ± 400</td>
<td>15.9</td>
</tr>
</tbody>
</table>

### TABLE III. TRITIUM IN FOOD

<table>
<thead>
<tr>
<th>Food type</th>
<th>‘Loose’ tritium (pCi/l)</th>
<th>‘Bound’ tritium (pCi/l)</th>
<th>Ratio ‘Bound’/‘Loose’</th>
</tr>
</thead>
<tbody>
<tr>
<td>Milk</td>
<td>250 ± 40</td>
<td>460 ± 50</td>
<td>1.8</td>
</tr>
<tr>
<td>Bread</td>
<td>190 ± 40</td>
<td>800 ± 60</td>
<td>4.2</td>
</tr>
<tr>
<td>Lettuce</td>
<td>200 ± 40</td>
<td>680 ± 50</td>
<td>3.4</td>
</tr>
<tr>
<td>Root vegetable</td>
<td>190 ± 40</td>
<td>750 ± 50</td>
<td>3.9</td>
</tr>
<tr>
<td>Meat</td>
<td>180 ± 30</td>
<td>520 ± 40</td>
<td>2.9</td>
</tr>
<tr>
<td>Fish</td>
<td>160 ± 30</td>
<td>630 ± 50</td>
<td>3.9</td>
</tr>
</tbody>
</table>

### TABLE IV. DIETARY TRITIUM INTAKE BY FOOD CATEGORIES

<table>
<thead>
<tr>
<th>Category</th>
<th>Tritium intake (pCi/year)</th>
<th>‘Loose’</th>
<th>‘Bound’</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbohydrates</td>
<td></td>
<td>0.030</td>
<td>0.028</td>
</tr>
<tr>
<td>Proteins</td>
<td></td>
<td>0.012</td>
<td>0.015</td>
</tr>
<tr>
<td>Dairy (milk)</td>
<td></td>
<td>0.048</td>
<td>0.007</td>
</tr>
<tr>
<td>Dry foods</td>
<td></td>
<td></td>
<td>0.019</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>0.098</td>
<td>0.069</td>
</tr>
</tbody>
</table>
previously reported. We found an average concentration of 2500 pCi/l for the ‘bound’ tritium in the Northeastern United States in 1970. The concentration reported for 1977, about 2600 pCi/l, after decay correction to 1970, would lead to a half-time of about 9 years for ‘bound’ tritium in soil in the Northeastern United States. We have previously reported a half-time of 3-4 years for a soil sample collected in Tulsa, Oklahoma.

The ratio of the specific activities, ‘bound’ to ‘loose’ tritium in soil, indicates that there is an increase compared to our previously reported value. In 1970 the ratio was 8-10 and now in 1977 it is about 13. The increase may be due to differences in the relative half-times in the ‘loose’ and ‘bound’ tritium compartments.

Food Analyses

Selected food categories which comprise the significant portion of the U.S. diet have been analyzed to estimate the change in tritium dietary intake compared to our previous measurements. The results are reported in Table III. In 1970-1971, the estimated dietary intake was 1700 pCi/d or 0.63 μCi/y. The results presented in Table IV for 1978 provide an estimated dietary intake of 470 pCi/d or 0.17 μCi/y. This represents a 73% reduction in tritium dietary intake over a period of eight years. The contributions of ‘loose’ and ‘bound’ tritium to the dietary intake remained the same as we reported for the 1970-1971 sampling, with 70% of the dietary intake contributed by the ‘loose’ tritium fraction. The ratio of ‘loose’ to ‘bound’ tritium specific activity in food is about 4, which is consistent with our previous observation.

An estimate of the tritium half-time for the composite ‘loose’ and ‘bound’ tritium in the diet is 5.2 years. This value was obtained by decay correcting the 1978 dietary result to 1970 and plotting the two values with respect to time. This half-time is the same as was calculated for tap water.

Tissue Samples

Autopsy tissue samples (lung, liver and kidney) collected in 1973 from two subjects have been analyzed and the results are reported in Table V. The results are similar to data for one group of human organs that we analyzed in 1972. The ‘loose’ tritium concentrations in 1973 are in agreement with the surface water concentration, however, there has been no significant decrease in the ‘bound’ tritium concentration. Therefore, the ratio (‘bound’/‘loose’) has increased from 1.9 (1972) to 2.0 (1973). This apparent increase in the ratio may reflect only individual variability in tritium exposure of the general population.

DISCUSSION

The lyophilization method for analysis of ‘loose’ and ‘bound’ tritium in environmental and biological samples has been reported [3]. This removal procedure does not preclude that some tritiated water may remain in the dried samples as water of hydration or constitution.

The specific activity ratio (‘bound’/‘loose’) decreases by a factor of 2-4 in ascending the trophic levels. Soils from the Northeastern United States presently exhibit a ratio of about 13 compared to a ratio of 8-10 in 1970. The increase in the ratio appears to be due to the increased half-time of ‘bound’ tritium compared to ‘loose’ tritium in soil. The ratio for vegetation (food) has remained constant since 1970 to
TABLE V. HUMAN TISSUE TRITIUM VALUES

<table>
<thead>
<tr>
<th>Organ type</th>
<th>‘Loose’ tritium</th>
<th>‘Bound’ tritium</th>
<th>Ratio ‘Bound’/‘Loose’</th>
</tr>
</thead>
<tbody>
<tr>
<td>Subject 1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung</td>
<td>250 ± 30</td>
<td>530 ± 40</td>
<td>2.1</td>
</tr>
<tr>
<td>Liver</td>
<td>220 ± 30</td>
<td>480 ± 40</td>
<td>2.2</td>
</tr>
<tr>
<td>Kidney</td>
<td>260 ± 30</td>
<td>520 ± 40</td>
<td>2.0</td>
</tr>
<tr>
<td>Subject 2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lung</td>
<td>220 ± 30</td>
<td>420 ± 30</td>
<td>1.9</td>
</tr>
<tr>
<td>Liver</td>
<td>250 ± 30</td>
<td>400 ± 40</td>
<td>1.6</td>
</tr>
<tr>
<td>Kidney</td>
<td>190 ± 30</td>
<td>450 ± 40</td>
<td>2.4</td>
</tr>
</tbody>
</table>

1976 at about 4. Tissue ratios have shown a slight increase from 1.5 in 1972 to 2.0 in 1975. This increase may be due to variation in tritium exposure of the general population.

Soil, vegetation and tissue ‘loose’ tritium concentrations are similar to present values for surface and tap water. This would suggest that equilibrium is established rapidly between the ‘loose’ fraction and environmental water sources. Human tissue ‘bound’ tritium concentrations are about the same for each organ within a particular subject, again, suggesting homogeneous labelling of the ‘bound’ tritium fraction.

Previously, we had reported tritium half-times of 3.2 years in the hydrological cycle and 3.5 years for ‘bound’ tritium in soil. The present results indicate a half-time of 5.2 years in the hydrological cycle. The increase in the half-time is probably due to the injection of additional tritium into the atmosphere from thermonuclear weapons tests. In fact, there has been little change in the surface water tritium concentration for the past 3-4 years, suggesting that steady state conditions exist. Present data indicates a longer half-time for ‘bound’ tritium in soil of about 9 years, but further studies would be required to confirm this.

It is evident that there has been a decrease in the tritium concentration in the environment from 1970 to 1977. The dietary tritium intake has decreased by a factor of 3 over this period. In recent years, tritium injections in the atmosphere from thermonuclear weapons tests have maintained the ambient surface water tritium concentration at 150-200 pCi/l. As a result of these injections, apparent half-times for tritium have increased from about 3 to 5 years.

REFERENCES


**DISCUSSION**

S. PORTER: Could you please explain how you prepare your 'bound' $^3$H samples, and what is the resultant dose to man from 'loose' and 'bound' fractions of the diet as you define them?

D.C. BOGEN: The dried sample remaining after lyophilization is combusted to convert organic hydrogen to water. The water collected is measured for its tritium content. Laboratory experiments have been performed to evaluate the efficiency of lyophilization and combustion, and other operational parameters; these were found to be satisfactory.

Regarding your second question, no dose calculations have been prepared so far.

A.A. MOGHISSI: Your work is a valuable addition to the cycling data available for $^{14}C$, as determined by the teams of Nydal and Vogel, to mention only two groups of investigators. The agreement among these data is indicative of their overall validity.

However, I do have some reservations regarding your terminology. There are aqueous, non-aqueous, organic and non-aqueous inorganic fractions. The words 'loose' and 'bound' may be misleading. It might be better to say 'aqueous' and 'non-aqueous'.

Y. UENO: I also am worried about the use of the terms 'loose' tritium or 'bound' tritium.

D.C. BOGEN: Possibly a better term for the aqueous fraction would be 'free water tritium', available by lyophilization, and the second fraction might be more appropriately called 'organic bound tritium'.

L. MERLIVAT: You have shown that the tritium content of tap water in New York has been more or less constant since 1974. You attribute this to
nuclear weapons test fall-out. But since the area in question is heavily industrial, do you not think that some other sources could be involved, as for instance nuclear reactors?

D.C. BOGEN: We did not consider any other source of tritium except nuclear weapons tests. The water catchment areas and reservoirs are located at least 100 km from any nuclear power plant, and therefore only atmospheric discharges could be involved; these are small in the case of the power plants concerned.
WORLD-WIDE POPULATION DOSES FROM TRITIUM RELEASED FROM NUCLEAR FACILITIES

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Battelle,
Pacific Northwest Laboratories*,
Richland,
Washington,
United States of America

Abstract

WORLD-WIDE POPULATION DOSES FROM TRITIUM RELEASED FROM NUCLEAR FACILITIES.

Radiation doses to the world population are estimated from tritium oxide (HTO) concentrations in the air and surface water derived from a latitudinal band model. The HTO is assumed to be continuously released to the atmosphere from a source located in the 30°–50° north latitudinal band until equilibrium levels are reached. The doses from inhalation and skin absorption (transpiration) are calculated from the resulting estimated average air concentration for a particular latitudinal band. The doses from the ingestion of drinking water and food products are calculated assuming that all the food and half of the drinking water consumed in a given latitudinal band contains HTO at the same specific activity as the average surface water over land areas estimated for that band. In addition, local and U.S. population doses were calculated from more conventional dispersion models. For a continuous release rate of 1 Ci of tritium as HTO per year to the atmosphere, the total annual dose to the world population in the year 2000 is calculated to be 0.024 man-rem. This value is higher than would have been calculated from a simple model which assumed dilution of the HTO in the circulating waters of the Northern Hemisphere. Dose rate, dose commitment and accumulated dose have been estimated from release rates of HTO from nuclear facilities having a finite lifetime. For example, a release rate of one million Ci/y of tritium in the form of HTO to the atmosphere for 40 years leads to an estimated worldwide collective dose of \(2 \times 10^4\) man-rem in the 40th year and a total accumulated dose commitment of \(1 \times 10^6\) man-rem.

INTRODUCTION

Tritium, principally in the form of tritium oxide, is currently being released to the environment from nuclear power reactors, fuels separation plants, and has been identified as a principle radionuclide in effluents from future fusion reactor power plants.

* Operated by Battelle Memorial Institute for the U.S. Department of Energy.
As part of a major study sponsored by the U.S. AEC (now U.S. DOE) on the potential environmental impact of fusion reactors, radiation doses to various population groups from tritium released at a hypothetical fusion reactor located in the midwestern U.S. were estimated for three population groups: the regional population residing within 80 km of the plant, the population of the eastern U.S., and the world population. In addition, the worldwide population dose and dose commitment from tritium released to the atmosphere during operation of a nuclear facility having a finite lifetime were estimated.

TRITIUM DOSE FACTORS

For the calculation of annual doses to individuals and population from tritium, dose factors were derived using the method of ICRP [1,2]. An average energy for tritium beta particles was taken to be 5.7 keV/disintegration [3]. The effective half-life in the body was taken to be 10 days [4] for an adult weighing 70 kg (reference man) with a breathing rate of 7300 m³/y [1].

Adult dose factors (D.F.) for the ingestion and inhalation pathways were obtained from the U.S. NRC report NUREG-0172 [5] by changing the Quality Factor from the 1.7 used there to the value of 1.0 recommended by the ICRP.

\[
\text{Ingestion D. F.} = 6.2 \times 10^{-8} \text{ mrem/y per pCi/y intake} \\
\text{Inhalation D. F.} = 4.5 \times 10^{-4} \text{ mrem/y per pCi/m}^3 \text{ air}
\]

To account for the total-body dose from the passage of gaseous HTO through the skin (transpiration) the above inhalation dose factor was increased by 50% for the dose calculations [6].

Population Doses

Radiation doses were estimated for population groups residing at various distances from a facility:

- Regional population (those persons living within 80 km of the plant)
- Eastern U.S. population
- World population

Equilibrium doses to the first two population groups from atmospheric releases of 1 Ci/y of tritium were estimated from environmental concentrations derived from dispersion data from Hefetter [7]. Concentrations for calculating the doses to the world population were derived from the latitudinal band model [8,9].

Annual Dose to Regional Population

The annual dose from inhalation and transpiration to those persons living within 80 km of the plant was calculated by assuming that the HTO released into the atmosphere was uniformly mixed with the annual average moisture content of the atmosphere near the site (8 ml/m³).

The distribution of the population (6.4 x 10⁶ persons) was taken from 1970 census data for a plant sited at Morris, Illinois. The atmospheric dispersion data was also taken for this site [7,8].

The ingestion pathways were evaluated by assuming that the specific activity of the tritium in all terrestrial foods was equal to that of the
TABLE I. DIETARY ASSUMPTIONS USED IN CALCULATING INTERNAL RADIATION DOSES TO REGIONAL POPULATION FROM FOOD PATHWAYS

<table>
<thead>
<tr>
<th>Food</th>
<th>Consumption (kg/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Leafy vegetables</td>
<td>30</td>
</tr>
<tr>
<td>Other above ground vegetables</td>
<td>23</td>
</tr>
<tr>
<td>Potatoes</td>
<td>65</td>
</tr>
<tr>
<td>Root vegetables</td>
<td>11</td>
</tr>
<tr>
<td>Berries</td>
<td>3.9</td>
</tr>
<tr>
<td>Melons</td>
<td>5.2</td>
</tr>
<tr>
<td>Orchard fruit</td>
<td>33</td>
</tr>
<tr>
<td>Wheat</td>
<td>47</td>
</tr>
<tr>
<td>Other grain</td>
<td>12</td>
</tr>
<tr>
<td>Eggs</td>
<td>20</td>
</tr>
<tr>
<td>Milk</td>
<td>100</td>
</tr>
<tr>
<td>Beef</td>
<td>40</td>
</tr>
<tr>
<td>Pork</td>
<td>40</td>
</tr>
<tr>
<td>Poultry</td>
<td>20</td>
</tr>
<tr>
<td>Drinking Water</td>
<td>440*</td>
</tr>
<tr>
<td>Fish</td>
<td>6</td>
</tr>
<tr>
<td>Invertebrates</td>
<td>0.5</td>
</tr>
</tbody>
</table>

* Half from deep wells.

water vapor in the air. The consumption rates used for average member of the population are listed in Table I. Concentrations of HTO in surface water, drinking water and aquatic foods would be several orders of magnitude lower than those in terrestrial foods. Doses from aquatic pathways would therefore be insignificant for atmospheric releases of tritium. Table II summarizes annual doses to the regional population.

Annual Dose to the Eastern U.S. Population

The dose to the population of the eastern U.S. was calculated on the basis of the HTO released into the atmosphere and later washed out into the surface waters by rain. The inhalation (plus transpiration) dose was estimated for the population of the U.S. living east of the Rocky Mountains using normalized HTO air concentration values derived in Bande et al. The consumption rates of food and water were the same as those used for the Regional Population as given in Table I. Table III summarizes the results of the dose calculations. The dose to people living within 80 km of the plant is not included in the values in this table.
### TABLE II. ANNUAL DOSES TO REGIONAL POPULATION\textsuperscript{a} FROM A RELEASE RATE OF 1 Ci/y OF TRITIUM (HTO) TO THE ATMOSPHERE

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Dose (man-rem)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inhalation and transpiration</td>
<td>$2.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>Drinking water</td>
<td>-- \textsuperscript{b}</td>
</tr>
<tr>
<td>Aquatic foods</td>
<td>--</td>
</tr>
<tr>
<td><strong>Food products:</strong></td>
<td></td>
</tr>
<tr>
<td>Produce</td>
<td>$2.2 \times 10^{-4}$</td>
</tr>
<tr>
<td>Milk</td>
<td>$5.1 \times 10^{-5}$</td>
</tr>
<tr>
<td>Eggs</td>
<td>$4.4 \times 10^{-5}$</td>
</tr>
<tr>
<td>Meat</td>
<td>$3.2 \times 10^{-5}$</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>$5.1 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Those persons living within 80 km of the plant.

\textsuperscript{b} Insignificant contribution to dose.

### TABLE III. ANNUAL DOSES TO EASTERN U.S. POPULATION\textsuperscript{a} FROM A RELEASE RATE OF 1 Ci/y OF TRITIUM (HTO) TO THE ATMOSPHERE

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Dose (man-rem)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inhalation and transpiration</td>
<td>$3.5 \times 10^{-4}$</td>
</tr>
<tr>
<td>Drinking water</td>
<td>-- \textsuperscript{b}</td>
</tr>
<tr>
<td>Aquatic foods</td>
<td>--</td>
</tr>
<tr>
<td><strong>Food products:</strong></td>
<td></td>
</tr>
<tr>
<td>Produce</td>
<td>$8.8 \times 10^{-4}$</td>
</tr>
<tr>
<td>Milk</td>
<td>$3.8 \times 10^{-4}$</td>
</tr>
<tr>
<td>Eggs</td>
<td>$8.8 \times 10^{-5}$</td>
</tr>
<tr>
<td>Meat</td>
<td>$3.8 \times 10^{-5}$</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>$2.1 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Not including regional population.

\textsuperscript{b} Insignificant contribution to dose.
TABLE IV. EQUILIBRIUM HTO CONCENTRATIONS AND POPULATIONS FOR VARIOUS LATITUDINAL BANDS

<table>
<thead>
<tr>
<th>Latitude</th>
<th>HTO Concentration[^8,9] (pCi/m³ H₂O)</th>
<th>Population[^1] (Millions)</th>
</tr>
</thead>
<tbody>
<tr>
<td>90°-70°N</td>
<td>0.10</td>
<td>--</td>
</tr>
<tr>
<td>70-50</td>
<td>0.16</td>
<td>480</td>
</tr>
<tr>
<td>50-30[^b]</td>
<td>0.17</td>
<td>1200</td>
</tr>
<tr>
<td>30-10</td>
<td>0.095</td>
<td>1500</td>
</tr>
<tr>
<td>10°N-10°S</td>
<td>0.045</td>
<td>430</td>
</tr>
<tr>
<td>10-30</td>
<td>0</td>
<td>130</td>
</tr>
<tr>
<td>30-50</td>
<td>0</td>
<td>60</td>
</tr>
</tbody>
</table>

[^a] A uniform surface water depth of 0.5 m is assumed.

[^b] A continuous release rate to the atmosphere of 1 Ci/y HTO is assumed to be located in this latitude band.

Annual Dose to World Population

The annual dose to the world population from a continuous 1 Ci/y release to the atmosphere was estimated using the normalized equilibrium surface water concentrations taken from the latitudinal-band model of Bander et al.[8,9] and populations (total of 3.8 x 10⁹ people) taken from Machta, et al.[10]. These data are listed in Table IV. Again, the tritium concentration of the food and one-half of the water consumed by the people in a latitudinal band was assumed to be the same as that of the surface waters over the land areas of that band. Values of 440 kg/y and 450 kg/y were taken as the average water and food consumption of the world population respectively. Table V summarizes the annual doses calculated for the world population.

Summary of Dose to the Population Groups from Tritium

The annual doses to the three population groups from releases of tritium to the atmosphere are summarized in Table VI.

The total of 0.024 in Table VI may be somewhat conservative since the doses were calculated using different models. Each entry includes, to some degree, those doses immediately above it in the table. On the other hand, the fraction of food and water which were assumed to be contaminated to the equilibrium specific activity of HTO in surface water is subject to question. On balance the final total is probably a reasonable estimate within a factor of 10 of the potential world population dose.
TABLE V. ANNUAL DOSES TO WORLD POPULATION FROM A CONTINUOUS RELEASE RATE OF 1 Ci/y TRITIUM (HTO) TO THE ATMOSPHERE

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Dose (man·rem)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inhalation and transpiration</td>
<td>0.0038</td>
</tr>
<tr>
<td>Drinking water</td>
<td>0.0059</td>
</tr>
<tr>
<td>All foods</td>
<td>0.0120</td>
</tr>
<tr>
<td>TOTAL</td>
<td>0.022</td>
</tr>
</tbody>
</table>

TABLE VI. SUMMARY OF ANNUAL DOSES TO POPULATIONS FROM A CONTINUOUS RELEASE RATE OF 1 Ci/y TRITIUM (HTO) TO THE ATMOSPHERE

<table>
<thead>
<tr>
<th>Population</th>
<th>Dose (man·rem)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Regional</td>
<td>0.0005</td>
</tr>
<tr>
<td>United States</td>
<td>0.002</td>
</tr>
<tr>
<td>World a</td>
<td>0.022</td>
</tr>
<tr>
<td>TOTAL</td>
<td>0.024</td>
</tr>
</tbody>
</table>

a The world dose includes only that U.S. population dose calculated with the latitudinal-band model.

Comparison of World Population Dose with Those from the EPA Model

The model of worldwide dispersion and dosimetry used by the U.S. Environmental Protection Agency[11] assumes that the tritium released from a plant in the continental U.S. is transported eventually to the world's oceans to be mixed uniformly in only that portion in the northern hemisphere. Only the population of the northern hemisphere (80% of the world population) is assumed exposed and their body water is taken to be equal to the specific activity of tritium in the northern hemisphere oceans at equilibrium after a constant release rate. The volume of water available for dilution is taken to be $1.35 \times 10^{16} \text{m}^3$[11], the world population is $3.8 \times 10^9$ people; and the total-body dose factor for a sustained concentration is $100 \text{ rem/y per Ci/m}^3$[11] of tritium in the body. Then the world population dose rate from a release rate of 1 Ci/y of tritium in the form of HTO after equilibrium is just:

$$1 \text{ Ci/y} \times 0.8 \times 3.8 \times 10^9 \text{ people} \times 100 \text{ rem/y} \times 12.3 \text{ y} = 4 \times 10^{-4} \text{ man·rem/y}$$

$$1.35 \times 10^{16} \text{ m}^3 \times \text{Ci/m}^3 \times \ln 2$$
This annual collective dose \((4 \times 10^{-4} \text{ man-rem})\) is exclusive of any localized dose in the region of the nuclear facility. As such, it can be compared with the value of 0.022 man-rem/\(y\) from Table V. On this basis the world population dose calculated in this study is about 50 times that calculated using the EPA method.

**World Population Dose From a Fuels Reprocessing Plant**

From the previous discussion, it can be seen that a continuous release rate of 1 Ci/y of HTO to the atmosphere leads to an annual collective dose to the world population of 0.024 man-rem at equilibrium. (At equilibrium the world inventory of tritium from a 1 Ci/y release rate would be maintained at 17.7 Ci.) In addition, the dose commitment out to all time to a constant world population of \(3.8 \times 10^9\) persons from a curie of HTO released to the atmosphere would also be equal to 0.024 man-rem.

A fuels reprocessing plant releasing \(10^6\) Ci/y of HTO to the atmosphere throughout its 40-year lifetime would contribute a total collective dose commitment to the world population of:

\[40 \times 10^6 \text{ Ci/y} \times 0.024 \text{ man-rem/Ci} = 9.6 \times 10^5 \text{ man-rem}\]

In addition, a continual release rate for 40 years would lead to an annual collective dose during the 40th year of plant operation of:

\[10^6 \text{ Ci/y} \times [1 - \exp(-40/17.7)] \times 0.024 \text{ man-rem/Ci} = 2.2 \times 10^4 \text{ man-rem/y}\]

If the above collective doses are divided by the world population of \(3.8 \times 10^9\) persons, the average dose commitment and the average annual dose would be \(2.5 \times 10^{-4}\) rem and \(5.7 \times 10^{-6}\) rem/\(y\) respectively. These values are insignificant whether compared to existing radiation standards or to a natural background radiation dose rate of approximately 0.1 rem/\(y\).

**SUMMARY**

Dose rates from postulated releases of tritium oxide to the atmosphere were calculated for various population groups. Assuming a release rate of 1 Ci/y of HTO to the atmosphere, the annual collective dose to the population of the world, including the United States and the regional population, was estimated to be 0.024 man-rem.

A release rate of \(10^6\) Ci/y of HTO to the atmosphere for 40 years from a large fuels reprocessing plant would lead to a maximum collective dose to the world population of \(2 \times 10^4\) man-rem in the 40th year, and a collective dose commitment of \(1 \times 10^6\) man-rem during the total time required for the released tritium to decay to insignificance.

**REFERENCES**

DISCUSSION

S. PORTER: For the eastern United States of America, why did you not use the pathway represented by $^3$H in river drinking water? With the grouping and location of reactors in the eastern part of the country, this is the critical pathway in that area. Also, why did you not apply ICRP-23 (Standard Man) data to your human intake figures?

J.K. SOLDAT: The answer to the first question lies in the original assumption — which is I think made clear in the paper — that the release of HTO was from a CTR and hence entirely to the atmosphere. Likewise, in the instance of the hypothetical fuel reprocessing plant there are no radioactive liquid effluents in current designs. Surface water contamination was estimated from rainout for the eastern U.S. population dose calculations, but this contribution to dose was insignificant.

The answer to your second question is quite simple: the calculations were performed before I received my copy of ICRP Publication No.23.
H. BONKA: Why do you integrate the collective dose out to 80 km only (Table II of your paper)? Tritium is transported over a distance of approximately 2000 km before it is washed out.

J.K. SOLDAT: The standard practice of integrating collective dose out to a distance of 80 km apparently developed from a paper prepared by Carl Gamertsfelder of the U.S. Nuclear Regulatory Commission in about 1970. For this paper Gamertsfelder calculated the total-body collective doses from noble gases released at operating U.S. power reactors. He found that addition of the air submersion doses received by persons residing further than 80 km from the point of release did not significantly increase the collective dose. It therefore appeared unnecessary to account for collective doses from noble gases beyond that distance.

T.R. THOMAS: Would you expect the regional dose of $^3$H from a reprocessing plant to be at least a factor of 200 lower if released as HT instead of HTO? Of course the United States or world dose would not be affected by the chemical form. If the regional dose per man as HTO is 10–15 mrem yearly, then a factor of 200 less would bring the $^3$H down to about 0.5 mrem annually. That would appear acceptable in the light of current radiation guidelines.

J.K. SOLDAT: From information presented at this Symposium it does indeed appear that the dose from HT would be 1/200 of that from an equal concentration of HTO. However, as you point out, the collective dose over the world population should be nearly the same for both forms, since HT is converted eventually to HTO in the environment. The doses to a person near the site boundary of the FRP were not given in my paper, though they have been calculated. Assuming a maximum off-site atmospheric dispersion factor of $10^{-7}$ s/m$^3$ and a release rate of $10^4$ Ci HTO/a, the maximum individual dose is 10–15 mrem/a. The hypothetical FRP would service about 40 GW/a of electricity generating capacity each year. Hence the maximum individual dose would be about 0.3 mrem per GW/a, much below the current EPA guideline of 25 mrem per GW/a electrical.

C.E. EASTERYL: May I recall in connection with Mr. Thomas’ question that in 1974 Mr. Jacobs and myself investigated the effect on the global population of releasing HT to the atmosphere.¹ We found that release of HT to the atmosphere would give rise to very nearly the same collective dose (in precise terms slightly less) compared with releases of HTO. In addition, we observed that releases of HTO to the atmosphere result in about ten times less collective dose compared with HTO releases to surface waters.

¹ See Ref. [15], JACOBS, D.G., EASTERYL, C.E., PHILLIPS, J.E., “Influence of the rate of conversion of HT to HTO on projected radiation doses from release of molecular tritium”, these Proceedings, IAEA-SM-232/81.
J.A. GARLAND: Does the model take account of exchange of tritiated water vapour at ocean and land surfaces, and what gives rise to the large difference in the relative contribution of drinking water to the total dose in the regional and world-wide doses?

J.K. SOLDAT: The answer to the first question is yes. Regarding your second question, the answer is probably to be sought in the fact that the doses to the three population groups were calculated using three different atmospheric dispersion and rainout models. Perhaps the surface water concentration in the regional population dose model was slightly underestimated.

G. UZZAN: I should like to mention that as part of a European research programme we are working on a model for evaluating collective doses. The difficulties we are encountering in doing this are the same as you have pointed out, particularly problems connected with long-distance atmospheric transport, and problems associated with the distribution of contaminated materials. Regarding atmospheric transport, can you tell us something about the model you used for your calculations, and regarding dilution in the course of distribution of foodstuffs would you agree with me that the collective dose you have calculated represents a very upper limit of the hazard (since you did not take the above dilution into account)?

J.K. SOLDAT: The model (which is I believe theoretical rather than experimentally based) used for the long-range transport calculations was described in a paper presented in Session II of this Symposium.2

Shipment of tropical fruit to the northern hemisphere can lower the 3H intake of the consumer living in the latter. However, we also have shipment of food in the opposite direction: this would raise the 3H intake of people in the southern hemisphere. Our model simply assumed that the food was all grown in the same latitudinal band as that in which it was consumed. Also, the diets assumed represented only a gross approximation.

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TRITIUM IN THE ENVIRONMENT — NCRP REPORT No. 62

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Abstract

TRITIUM IN THE ENVIRONMENT — NCRP REPORT No. 62.

The NCRP (National Council on Radiation Protection and Measurements) Report No. 62 on tritium is described. Tritium production from various sources, distribution and environmental kinetics, biological behaviour and the dosimetry of tritium are discussed.

* Operated by Battelle Memorial Institute for the U.S. Department of Energy.
Tritium, the heaviest and only radioactive isotope of hydrogen, has been a ubiquitous contaminant produced by nuclear energy programmes. The nuclide is produced copiously by the military nuclear programme and tritium, in the form of water, was distributed world-wide by the nuclear weapons tests that took place in the open atmosphere, particularly during the period 1954 to 1962. The latter source has diminished substantially as a result of the limited nuclear test ban agreement among the major powers, but tritium production by nuclear power reactors is increasing rapidly and will in time become the dominant source.

The world inventory of natural tritium from cosmic ray interactions is estimated to be 70 MCi, corresponding to a production rate of 4 MCi a⁻¹. The tritium inventory from weapons testing reached a maximum of about 3100 MCi in 1963, an amount that will decay to the natural level of 70 MCI approximately by the year 2030.

Tritium is formed in nuclear reactors by ternary fission and by activation of light elements such as boron, which is used for reactivity control in PWRs, and lithium, which is used to control corrosion. Estimates of the production rate by ternary fission range from below 12 to greater than 20 Ci megawatts (thermal) per day (MWt day)⁻¹. Light element activation contributes about 600–800 Ci a⁻¹ to the reactor coolant with an average annual release to the environment per 100 megawatts (electric) (MW(e)) by light-water reactors of 63 Ci a⁻¹ for BWRs and 830 Ci a⁻¹ for PWRs. Barring a resumption of large-scale atmospheric testing, emission of tritium from the nuclear fuel cycle by 1985 will become more important than the residue from weapons testing. Emissions from operating light-water reactors are, and will continue to be, insignificant compared with the releases from fuel reprocessing.

Transfer of tritiated water from the atmosphere to the surface of the earth occurs mainly by precipitation, but also by vapour exchange. The mean residence time of tritiated water vapour in the troposphere ranges from 21–40 days. Deposition of atmospheric tritium is greatest in the latitudinal belt in which the tritium is released. If the atmospheric concentration of tritium is known, the average deposition rate can be estimated from the deposition velocity which has been observed to be in the range of 0.4 to 0.8 cm s⁻¹. The deposition rate is higher over the oceans than over land.

Tritium in groundwater and soil exhibits the diffusion characteristics of water except for insignificant differences in vapour pressure. Absorption in soil is influenced by the state of the soil, soil structure, water content and the amount of organic matter present.

Tritium reaches surface water directly through precipitation, molecular exchange with water in the atmosphere, direct release of tritiated water from power or reprocessing nuclear plants to streams, and indirectly from run-off of groundwater into streams. Tritium in the oceans becomes rapidly distributed in the thin surface layer of relatively warm water, 50 to 100 m deep, known as
the 'mixed layer'. Residence time in the mixed layer is about 22 years at 75 m depth, but this varies with geographic location. In contrast to the oceans, most lakes are mixed vertically each year.

When humans are exposed to tritium as tritiated water by inhalation, ingestion or skin absorption, it is rapidly distributed to intracellular and extracellular water. The kinetics of tritium movement throughout the body follow those of water. A small fraction of the intake becomes organically bound in two separate compartments. The effective half-life of the $^3$H in free water is 9.7 days compared with 30 days and 450 days from the two compartments into which the fraction is bound.

Reported values of the relative biological effectiveness (RBE) of tritium range from less than 1.0 to greater than 2.0. This variation is due, in part, to the fact that the RBE is functionally dependent on the biological endpoint, and on the dose rate, as well as the reference radiation used, i.e. $^6$Co gamma rays or 250 kV X-rays. There are also uncertainties in dose estimation.

Projections of production and release of tritium are tied to economic growth and political decisions that are difficult to foresee. Although the uncertainties of future development of nuclear energy are great, the inventories of tritium produced are certain to increase. In opposition to this increase are probably future improvements in the control of tritium release and containment.

This report assumes that installed world-wide nuclear capacity will reach 120 GW(e) by 1980 and 720 GW(e) by 2000.

The dose to humans residing in the northern hemisphere was calculated and gives the following estimates:

<table>
<thead>
<tr>
<th>Natural tritium (mrad a$^{-1}$)</th>
<th>Fall-out (mrad a$^{-1}$)</th>
<th>Nuclear power (mrad a$^{-1}$)</th>
<th>Weapons production (mrad a$^{-1}$)</th>
<th>Total (mrad a$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1980 0.001</td>
<td>0.005</td>
<td>0.0009</td>
<td>0.002</td>
<td>0.009</td>
</tr>
<tr>
<td>2000 0.001</td>
<td>0.0006</td>
<td>0.01</td>
<td>0.002</td>
<td>0.014</td>
</tr>
</tbody>
</table>

**DISCUSSION**

H.C. ROTHCHILD: Do your projections for nuclear power and tritium production refer exclusively to the United States of America or are they worldwide?
D.G. JACOBS: Perhaps I should reply to this question. The projections for nuclear power generation considered the evaluations made by both the U.S. Energy Research and Development Administration (now the Department of Energy) and the IAEA. World-wide distribution was based on the IAEA projections and the mix of reactor types, and the total nuclear power projections were modified from the USERDA estimates to account for the lower contribution from nuclear power currently projected.
MONITORING OF TRITIUM

Session 9, Part 2, and
Session 10
IMPACT ON THE ENVIRONMENT OF TRITIUM RELEASES FROM THE KARLSRUHE NUCLEAR RESEARCH CENTER

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Abstract

IMPACT ON THE ENVIRONMENT OF TRITIUM RELEASES FROM THE KARLSRUHE NUCLEAR RESEARCH CENTER.

Since 1969 tritium activities of $(1.2 - 2.1) \times 10^3 \text{ Ci/a}$ have been released into the atmosphere from the Karlsruhe Nuclear Research Center. These releases have given rise to contamination which can be detected by means of the liquid scintillation measuring technique (detection limit attained about $0.15 \text{ pCi/ml}$) without enrichment. Estimates based on precipitation measurements have shown that a small portion of $5 \times 10^{-4}$ of the tritium activity released is deposited within an area of 1 km radius around the FR 2 stack. In 1977 the tritium precipitation from fall-out was of the same order of magnitude (213 mCi/km$^2$). The contamination of soil by tritium within the fenced area of the Karlsruhe Nuclear Research Center ranges from 10 to 1 pCi/ml of soil humidity, each value averaged over a depth of 0 to 80 cm. The highest values have been measured in the immediate neighbourhood of the sewage clarification plant of the Nuclear Research Center (ground level source) and of the heavy-water-moderated MZFR reactor (200 MW(th), 100-m high exhaust stack). Simultaneous measurements of the air humidity, precipitations, soil humidity and tissue water of plants show that the tritium concentration in plants follows the variation with time of the tritium contamination in the air humidity and of precipitations. These variations are caused by fluctuating release rates and meteorological conditions. The radiation exposure of persons from tritium remains below 1 mrem/a.

1. INTRODUCTION

Tritium deserves special attention because of its radiological characteristics and its increasing production and application by industry. It is produced by fission and various nuclear reactions in nuclear reactors. Tritium is applied to a great extent in luminous paints, gaseous tritium light devices and in experiments for the development of nuclear fusion. It is released to the environment in the form of tritiated hydrogen gas and gaseous compounds, tritiated water and aerosols of solid compounds. The uptake by inhalation of solid compounds might be of
**FIG. 1.** Tritium releases from the Karlsruhe Nuclear Research Center for the period from 1969 to 1977.

**TABLE I. TRITIUM RELEASES BY THE KARLSRUHE NUCLEAR RESEARCH CENTER IN 1977**

<table>
<thead>
<tr>
<th>Emitter</th>
<th>Height of emission (m)</th>
<th>Tritium release (Ci) 1977</th>
</tr>
</thead>
<tbody>
<tr>
<td>FR 2 (44 MW(th), D₂O-moderated)</td>
<td>99</td>
<td>192</td>
</tr>
<tr>
<td>MZFR (200 MW(th), D₂O-moderated)</td>
<td>99.5</td>
<td>1017</td>
</tr>
<tr>
<td>WAK (reprocessing plant, 40 t/a throughput, 3 X 10⁶ MWd/t)</td>
<td>60</td>
<td>190</td>
</tr>
<tr>
<td>FERAB (incineration facility for solid radioactive wastes)</td>
<td>70</td>
<td>330</td>
</tr>
<tr>
<td>Decontamination plant for liquid wastes</td>
<td>19</td>
<td>35</td>
</tr>
<tr>
<td>Sewage treatment plant (a) evaporation</td>
<td>~2</td>
<td>~3ᵃ</td>
</tr>
<tr>
<td>(b) liquid effluents</td>
<td>~</td>
<td>4000</td>
</tr>
<tr>
<td>Several minor emitters</td>
<td>~10</td>
<td>3</td>
</tr>
</tbody>
</table>

ᵃ Estimate based on an average evaporation rate of 1 mm/d for 800 m of uncovered surface.
FIG. 2. Site plan indicating the main tritium emitters of the Karlsruhe Nuclear Research Center and the sampling locations.

great scientific interest, since in this case the lungs are the critical organ, and it seems to be rather difficult to estimate the lung burden. This pathway of exposure has to be expected from the operation of accelerators and the production of targets for accelerators. Much more important is the release to the environment of tritiated water which enters the water cycle and thus is dispersed world wide. In the vicinity of the point of emission gaseous tritium releases are less important because the radiotoxicity of tritium gas is low.

Since several thousands of curies of tritium are released yearly from the Karlsruhe Nuclear Research Center via the effluent air and water [1], a programme was initiated to investigate thoroughly the consequences of these releases as well as the radio-ecological behaviour of tritium [2]. The experience based on the results of this programme might be useful for future planning of nuclear installations. The prerequisite of the evaluation of the measured results is the knowledge of the existing background contamination from nuclear weapons tests, the main source of today's world tritium inventory. The current sampling programme has therefore been extended to the Upper Rhine Valley region between Mannheim and Kehl (distance about 120 km). Additionally, the sampling programme includes some sampling locations at greater distances.
<table>
<thead>
<tr>
<th>Location of precipitation collectors</th>
<th>1972</th>
<th>1973</th>
<th>1974</th>
</tr>
</thead>
<tbody>
<tr>
<td>Within Karlsruhe Nuclear Research Center</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Roof of building 123</td>
<td>359</td>
<td>538</td>
<td>623</td>
</tr>
<tr>
<td>Measuring hut west</td>
<td>620</td>
<td>661</td>
<td>566</td>
</tr>
<tr>
<td>Measuring hut north-east</td>
<td>612</td>
<td>656</td>
<td>760</td>
</tr>
<tr>
<td>Karlsruhe Reproc. Plant</td>
<td>640</td>
<td>681</td>
<td>425</td>
</tr>
<tr>
<td>Outside Karlsruhe Nuclear Research Center</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Augustenbergr (Grötzingen)</td>
<td>623</td>
<td>619</td>
<td>614</td>
</tr>
<tr>
<td>Bruchsal</td>
<td>429</td>
<td>500</td>
<td>657</td>
</tr>
<tr>
<td>Egerstein</td>
<td>424</td>
<td>590</td>
<td>657</td>
</tr>
<tr>
<td>Teifgesindt waterworks (Leopoldshafen)</td>
<td>-</td>
<td>-</td>
<td>550</td>
</tr>
<tr>
<td>Philippsburg</td>
<td>-</td>
<td>-</td>
<td>584</td>
</tr>
<tr>
<td>Speyer</td>
<td>-</td>
<td>-</td>
<td>453</td>
</tr>
<tr>
<td>Niederstötzing</td>
<td>-</td>
<td>-</td>
<td>456</td>
</tr>
<tr>
<td>Buchmühle</td>
<td>-</td>
<td>-</td>
<td>460</td>
</tr>
</tbody>
</table>

<p>| Mean A precipitation (nci/m²)       | 0.84 | 0.62 | 0.62 |
| Mean A precipitation (nci/m³)       | 537  | 425  | 711  |
| Mean A precipitation (nci/m³)       | 3.06 | 3.06 | 3.06 |
| Mean A precipitation (nci/m³)       | 3.06 | 3.06 | 3.06 |
| Mean A precipitation (nci/m³)       | 3.06 | 3.06 | 3.06 |
| Mean A precipitation (nci/m³)       | 3.06 | 3.06 | 3.06 |</p>
<table>
<thead>
<tr>
<th>Locations of precipitation collectors</th>
<th>1975</th>
<th>1976</th>
<th>1977</th>
</tr>
</thead>
<tbody>
<tr>
<td>M (mm)</td>
<td>$\bar{a}$ (nCi/litr)</td>
<td>$A_F$ (nCi/m²)</td>
<td>M (mm)</td>
</tr>
<tr>
<td>Within Karlsruhe Nuclear Research Center</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Roof of building 123</td>
<td>599</td>
<td>1.87</td>
<td>1118</td>
</tr>
<tr>
<td>Measuring hut west</td>
<td>736</td>
<td>1.18</td>
<td>869</td>
</tr>
<tr>
<td>Measuring hut north-east</td>
<td>733</td>
<td>1.49</td>
<td>1088</td>
</tr>
<tr>
<td>Karlsruhe Reproc. Plant</td>
<td>734</td>
<td>1.23</td>
<td>898</td>
</tr>
<tr>
<td>Outside Karlsruhe Nuclear Research Center</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Augustenber (Grötzingen)</td>
<td>698</td>
<td>0.93</td>
<td>648</td>
</tr>
<tr>
<td>Bruchsal</td>
<td>675</td>
<td>0.86</td>
<td>581</td>
</tr>
<tr>
<td>Eggenstein</td>
<td>647</td>
<td>0.90</td>
<td>579</td>
</tr>
<tr>
<td>Tiefgestade waterworks (Leopoldshafen)</td>
<td>580</td>
<td>0.89</td>
<td>516</td>
</tr>
<tr>
<td>Philippsburg</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Speyer</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Niederstotzingen</td>
<td>467</td>
<td>0.85</td>
<td>395</td>
</tr>
<tr>
<td>Buchmühlhe</td>
<td>507</td>
<td>0.78</td>
<td>396</td>
</tr>
</tbody>
</table>

—: Not measured.
TABLE III. COMPARISON OF ANNUAL MEAN VALUES OF TRITIUM CONCENTRATION EVALUATED FROM THE RHINE AND NECKAR RIVERS AND ABOVE ALL FROM RIGHT TRIBUTARIES TO THE RHINE FOR THE YEARS 1972–77

<table>
<thead>
<tr>
<th>Waters investigated</th>
<th>Average tritium concentration of all sampling locations (nCi/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rhine</td>
<td>0.66</td>
</tr>
<tr>
<td>Neckar</td>
<td>0.37</td>
</tr>
<tr>
<td>Surface waters between Karlsruhe and Kehl (without the Rhine)</td>
<td>0.35</td>
</tr>
<tr>
<td>Surface waters between Mannheim and Karlsruhe (without the Rhine and Neckar)</td>
<td>0.25</td>
</tr>
</tbody>
</table>

The liquid scintillation technique was chosen as the method of measurement for all samples. It has the advantage that a great number of samples can be measured at relatively low expenditure. In addition, the detection limit of about 0.15 nCi/l attained is sufficient since it allows radiation exposures of about 20 μrem/a to be determined.

2. TRITIUM EMISSIONS FROM THE KARLSRUHE NUCLEAR RESEARCH CENTER

Tritium is released to the atmosphere mainly through exhaust stacks and almost exclusively in the form of tritiated water vapour. Other forms of tritium releases are estimated to be insignificant. In the effluent water tritium from all sources at the Nuclear Research Center is discharged via the sewage treatment plant. After passing through a 2.9-km-long sewer, the liquid effluents reach the ‘Altrhein’, a former branch of the river Rhine, and then flow, mixed with surface water, over a distance of 23.6 km to be discharged into the river.

Figure 1 gives a survey of the tritium releases through effluent air and water for the period from 1969 to 1977. Table I is a breakdown of releases from the different emitters of the Nuclear Research Center for the year 1977, and it indicates the heights of the sources above the ground. The locations of the main tritium emitters of the Karlsruhe Nuclear Research Center are shown in Fig.2.
Comprehensive monitoring routine programmes have been carried out to
determine all the tritium releases from the various facilities of the Center. When
direct measurement [3, 4] is impossible, tritiated water vapour is measured by
means of the liquid scintillation measurement technique on continuously condensed
humidity samples of exhaust air [5].

3. TRITIUM IN WATER

In this section results are reported of tritium in ground, drinking and surface
water, and in precipitations. Because of the abundance of data only annual mean
values are given. More details can be found in earlier publications [1, 6, 7].

3.1. Tritium in precipitations

Precipitation samples are of special interest because their tritium concen-
tration together with the amount of precipitation governs the tritium activity fed
into the soil and, consequently, have an impact on all the other media. Table II is
a compilation of values measured in the years 1972–1977. The mean values of
surface load at the four collecting stations located at the Karlsruhe Nuclear
Research Center clearly exceeded the values for the more distant locations,
because of the tritium emissions into the atmosphere from the Nuclear Research
Center. Establishing the difference in surface load values found on the site of and
outside the Nuclear Research Center gives a mean surface load of about 0.3 Ci/km²
yearly from releases. A substantial part of the surface load is caused by fall-out.

If the calculations are based on a mean surface load of 0.3 Ci/km² yearly from
releases, a total annual load of about 1 Ci is obtained for an area of 1 km radius
around the Nuclear Research Center. This means that the portion of about
$5 \times 10^{-6}$ of tritium released to the atmosphere is dispersed by precipitation in this
area. It is not possible to detect a surface load from releases at a greater distance
from the Nuclear Research Center because the variations as a function of time
and location of surface load from fall-out are greater than the small influence
expected to be exerted by the Nuclear Research Center in this more distant area.
The situation would be the same if the measuring accuracy were substantially
improved.

3.2. Tritium in surface waters

In Table III the annual mean values of the rivers Rhine and Neckar and of
some tributaries are compared for the years 1972–77. Values for various
sampling locations on the rivers Rhine and Neckar are listed in Table IV for the
years 1975–77. At most of these locations samples have been taken con-
tinuously by the Institut für Wasser- und Abfallwirtschaft of the Landesanstalt für
Umweltschutz Baden-Württemberg.
### TABLE IV. AVERAGE TRITIUM CONCENTRATIONS FROM THE RHINE AND NECKAR RIVERS FOR 1975—77

#### RHINE

<table>
<thead>
<tr>
<th>km</th>
<th>Sampling locations</th>
<th>$^{3}H$ concentration (nCi/litre)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1975</td>
</tr>
<tr>
<td>29</td>
<td>Ohringen-Stielen</td>
<td>0.63</td>
</tr>
<tr>
<td>91</td>
<td>Reckingen</td>
<td>0.68</td>
</tr>
<tr>
<td>113</td>
<td>Albrück-Dögern</td>
<td>0.66</td>
</tr>
<tr>
<td>155</td>
<td>Wyhlen</td>
<td>0.62</td>
</tr>
<tr>
<td>224</td>
<td>Breisach</td>
<td>0.64</td>
</tr>
<tr>
<td>248</td>
<td>Weisweil</td>
<td>0.65</td>
</tr>
<tr>
<td>294</td>
<td>Kehl*</td>
<td>0.64</td>
</tr>
<tr>
<td>362</td>
<td>Maxau</td>
<td>0.67</td>
</tr>
<tr>
<td>372</td>
<td>Leopoldshafen (ferry)*</td>
<td>0.73</td>
</tr>
<tr>
<td>392.6</td>
<td>Downstream of Rheinschanz isle*</td>
<td>0.99</td>
</tr>
<tr>
<td>400</td>
<td>Spyer*</td>
<td>0.62</td>
</tr>
<tr>
<td>426.2</td>
<td>Mannheim*</td>
<td>0.71</td>
</tr>
<tr>
<td>432</td>
<td>Mannheim*</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>Mean value</td>
<td>0.68</td>
</tr>
</tbody>
</table>

#### NECKAR

<table>
<thead>
<tr>
<th>km</th>
<th>Sampling locations</th>
<th>$^{3}H$ concentration (nCi/litre)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1975</td>
</tr>
<tr>
<td>0</td>
<td>Mannheim*</td>
<td>0.53</td>
</tr>
<tr>
<td>8.1</td>
<td>Feudenheim</td>
<td>0.51</td>
</tr>
<tr>
<td>18.1</td>
<td>Schwabenheim</td>
<td>0.50</td>
</tr>
<tr>
<td>21.0</td>
<td>Heidelberg*</td>
<td>0.58</td>
</tr>
<tr>
<td>31.0</td>
<td>Neckargemünd</td>
<td>0.47</td>
</tr>
<tr>
<td>72.5</td>
<td>Gütenbach</td>
<td>0.48</td>
</tr>
<tr>
<td>93.9</td>
<td>Gundelsheim</td>
<td>0.47</td>
</tr>
<tr>
<td>104.4</td>
<td>Kochendorf</td>
<td>0.49</td>
</tr>
<tr>
<td>118.0</td>
<td>Horkheim</td>
<td>0.49</td>
</tr>
<tr>
<td>125.5</td>
<td>Lauffen</td>
<td>0.47</td>
</tr>
<tr>
<td>143.1</td>
<td>Hossbach</td>
<td>0.50</td>
</tr>
<tr>
<td>165.2</td>
<td>Poppenweiler</td>
<td>0.52</td>
</tr>
<tr>
<td>172.3</td>
<td>Aldingen</td>
<td>0.53</td>
</tr>
<tr>
<td>187.0</td>
<td>Stuttgart-Untertürkheim</td>
<td>0.49</td>
</tr>
<tr>
<td>199.7</td>
<td>Ditzingen</td>
<td>0.54</td>
</tr>
<tr>
<td></td>
<td>Mean value</td>
<td>0.50</td>
</tr>
</tbody>
</table>

* At these sampling locations two samples are taken per month. For all other sampling locations the mean values are derived from concentration values of samples collected continuously over a fortnight.
The differences in the annual mean values shown in Tables III and IV are due to the varying mixing proportions of precipitations and ground water in flowing surface waters with relatively little run-off. Waterworks often process ground water with tritium concentrations lower than those of precipitations. The amounts of waste water so produced and discharged into the surface water are sufficiently high to influence the tritium concentration of the mixture. The tritium concentration in the surface water samples taken between Kehl and Karlsruhe is distinctly higher than that of the samples taken between Karlsruhe and Mannheim. This results from the higher contribution of precipitation water to the flowing surface water and the lower density of population in the Black Forest area where the right tributaries to the river Rhine south of Karlsruhe have their source.

Figure 3 shows the profiles of the mean tritium concentration along the river Rhine between Öhringen and Mannheim for the years 1975 and 1976. It should be mentioned that the tritium concentration in the Rhine river downstream of the Rheinschanz island is remarkably higher than upstream of this island (see also Table IV). This is due to liquid tritium discharges from the Nuclear Research Center, which arrive near this sampling location. Only 8 km downstream of this location average concentrations were found.

3.3. Tritium in ground and drinking waters

In the sampling programme a distinction is made between ground and drinking water samples. Pumped or scooped samples collected from wells at different depths are termed ground-water samples whereas all the samples taken from the tap water system are termed drinking-water samples. The tap water system is frequently supplied by two or more waterworks, even in small communities, so that the drinking water is often composed of a mixture of ground water
### TABLE V. ANNUAL MEAN VALUES OF TRITIUM CONCENTRATION IN GROUND WATER FROM 1973 TO 1977

<table>
<thead>
<tr>
<th>Sampling location</th>
<th>Annual mean values of the tritium concentration of ground water (nCi/ltr)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Vicinity of the Karlsruhe Nuclear Research Center (KfK) and reference locations Baden-Baden</strong></td>
<td></td>
</tr>
<tr>
<td>Southern waterworks in the KfK</td>
<td>0.19</td>
</tr>
<tr>
<td>Leopoldshafen waterworks</td>
<td>0.44</td>
</tr>
<tr>
<td>Linkenheim waterworks</td>
<td>0.33</td>
</tr>
<tr>
<td>Karlsruhe-Hardtwald waterworks</td>
<td>0.37</td>
</tr>
<tr>
<td>Well A (Karlsruhe Reprocessing Plant)</td>
<td>0.32</td>
</tr>
<tr>
<td>Well B (Karlsruhe Reprocessing Plant)</td>
<td>0.48</td>
</tr>
<tr>
<td>Observation well No. 16</td>
<td>0.27</td>
</tr>
<tr>
<td>Observation well No. 20</td>
<td>0.26</td>
</tr>
<tr>
<td>Tiefgestade waterworks</td>
<td>0.50</td>
</tr>
<tr>
<td>'Fettquelle' Baden-Baden</td>
<td>0.12</td>
</tr>
<tr>
<td>'Reitherbrunnen' Baden-Baden</td>
<td>0.17</td>
</tr>
<tr>
<td><strong>Vicinity of Niederwibach Nuclear Power Plant</strong></td>
<td></td>
</tr>
<tr>
<td>Dingolfingen</td>
<td>0.17</td>
</tr>
<tr>
<td>Aumühle</td>
<td>0.17</td>
</tr>
<tr>
<td>Kehl - Wörth</td>
<td>0.15</td>
</tr>
<tr>
<td>Wörth</td>
<td>0.24</td>
</tr>
<tr>
<td>Premises of power plant P I</td>
<td>0.28</td>
</tr>
<tr>
<td>Premises of power plant P II</td>
<td>0.28</td>
</tr>
<tr>
<td><strong>Vicinity of Gundremmingen Nuclear Power Plant</strong></td>
<td></td>
</tr>
<tr>
<td>Well Gulf 3 Gundelfingen</td>
<td>0.66</td>
</tr>
<tr>
<td>Well Lau 3 Lauringen</td>
<td>0.47</td>
</tr>
<tr>
<td>Well Lau 7 Lauringen</td>
<td>0.60</td>
</tr>
<tr>
<td>Gundremmingen waterworks</td>
<td>0.57</td>
</tr>
<tr>
<td>Dillingen waterworks</td>
<td>0.54</td>
</tr>
<tr>
<td>Conveyer Niederstotzingen 1</td>
<td>0.17</td>
</tr>
<tr>
<td>Conveyer Niederstotzingen 4</td>
<td>0.15</td>
</tr>
<tr>
<td>Conveyer Niederstotzingen 6</td>
<td>0.23</td>
</tr>
<tr>
<td>Conveyer Schottlithof 2</td>
<td>0.14</td>
</tr>
<tr>
<td>Conveyer Schottlithof 3</td>
<td>0.19</td>
</tr>
<tr>
<td>Conveyer Schottlithof 5</td>
<td>0.51</td>
</tr>
<tr>
<td>Conveyer Buchmühle</td>
<td>0.35</td>
</tr>
<tr>
<td>Well, premises of power plant</td>
<td>0.53</td>
</tr>
</tbody>
</table>
from different points and depths of pumping, and from spring water, whose tritium concentration may differ quite markedly. The drinking water samples have mainly been taken by the staff of the Safety Department at their places of residence and brought along for measurement.

The tritium concentration of the ground water depends both on the tritium supplied by precipitation and on the tritium supplied by surface water filtered on the river banks when the ground water is raised by waterworks in the direct vicinity of a river. In addition, the tritium concentration of the ground water depends on the depths of sampling and on the rate of vertical migration of precipitations in the soil down to the ground-water-carrying stratum. The migration of the ground water is normally very slow and depends on the composition of the soil. For all these reasons and if detailed knowledge of local conditions is not available, it is generally impossible to establish quantitative relationships between the tritium concentration of ground water, precipitations and possible neighbouring surface waters. It is important above all in such investigations to observe the development with the time of the tritium concentration over extended periods of time.

Most ground-water samples were taken from wells sunk in the vicinity of nuclear facilities. These observation wells were constructed in many cases for the specific purpose of background radioactivity measurements and routine investigations performed under environmental monitoring of nuclear power stations. Only a few ground-water samples investigated under this measuring programme programme were taken from waterworks.

In Tables V and VI the annual mean values of tritium concentration in ground and drinking water for the years 1973–77 have been compiled.

4. TRITIUM IN THE SOIL

Because of the expenditure involved it was not possible to realize a long-term soil monitoring programme. However, a few 'snapshots' of soil contamination show distinctly increased values in the immediate environment of the main tritium emitters. Figure 4 demonstrates the results of a series of measurements carried out in December 1977 [8]. The values given are mean values over a depth of 80 cm. They reveal the influence of the frequency distribution of wind direction which shows maxima for winds from the south-west and north-east. Figure 5 indicates the dependency of tritium concentration on the depth of soil humidity for two locations (see Fig. 4).

In Figure 6 the results are represented of six drillings taken down to the ground-water level. The position of these drillings is also shown in Fig. 2. In general, the tritium concentration does not vary very much with the depth. The exceptional situation of the drilling No. 2 must be explained by an accident. The
TABLE VI. ANNUAL MEAN VALUES OF TRITIUM CONCENTRATION
IN DRINKING WATER FROM 1972 TO 1977

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Marxzell</td>
<td></td>
<td></td>
<td>0.25</td>
<td>0.31</td>
<td>0.27</td>
<td>0.29</td>
</tr>
<tr>
<td>Neuenburg</td>
<td></td>
<td></td>
<td>0.19</td>
<td>0.22</td>
<td>0.17</td>
<td>0.18</td>
</tr>
<tr>
<td>Heidelberg</td>
<td>0.27</td>
<td>0.30</td>
<td>0.14</td>
<td>0.23</td>
<td>0.17</td>
<td>0.20</td>
</tr>
<tr>
<td>Mannheim</td>
<td>0.19</td>
<td>0.17</td>
<td>0.15</td>
<td>0.19</td>
<td>0.10</td>
<td>0.16</td>
</tr>
<tr>
<td>Ludwigshafen</td>
<td>0.25</td>
<td>0.15</td>
<td>0.10</td>
<td>0.10</td>
<td>0.08</td>
<td>0.09</td>
</tr>
<tr>
<td>Speyer</td>
<td>0.20</td>
<td>0.14</td>
<td>0.12</td>
<td>0.16</td>
<td>0.11</td>
<td>0.11</td>
</tr>
<tr>
<td>Kehl</td>
<td>0.64</td>
<td>0.55</td>
<td>0.51</td>
<td>0.66</td>
<td>0.50</td>
<td>0.44</td>
</tr>
<tr>
<td>Baden-Baden</td>
<td>0.28</td>
<td>0.30</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bruchsal</td>
<td>0.40</td>
<td>0.20</td>
<td>0.16</td>
<td>0.16</td>
<td>0.10</td>
<td>0.10</td>
</tr>
<tr>
<td>Eggenstein</td>
<td>0.63</td>
<td>0.24</td>
<td>0.21</td>
<td>0.27</td>
<td>0.18</td>
<td>0.20</td>
</tr>
<tr>
<td>Förch-Niederbühl</td>
<td>0.46</td>
<td>0.48</td>
<td>0.35</td>
<td>0.42</td>
<td>0.33</td>
<td>0.31</td>
</tr>
<tr>
<td>Friedrichstal</td>
<td>0.37</td>
<td>0.37</td>
<td>0.31</td>
<td>0.35</td>
<td>0.24</td>
<td>0.25</td>
</tr>
<tr>
<td>Jöhlingen</td>
<td>0.41</td>
<td>0.13</td>
<td></td>
<td>0.14</td>
<td>0.13</td>
<td>0.19</td>
</tr>
<tr>
<td>Hagenbach</td>
<td>0.29</td>
<td>0.18</td>
<td>0.17</td>
<td>0.19</td>
<td>0.09</td>
<td>0.22</td>
</tr>
<tr>
<td>Karlsruhe</td>
<td>0.38</td>
<td>0.21</td>
<td>0.18</td>
<td>0.27</td>
<td>0.21</td>
<td>0.17</td>
</tr>
<tr>
<td>Durlach</td>
<td>0.56</td>
<td>0.33</td>
<td>0.30</td>
<td>0.34</td>
<td>0.23</td>
<td>0.27</td>
</tr>
<tr>
<td>Kuhardt</td>
<td>0.18</td>
<td>0.14</td>
<td>0.10</td>
<td>0.08</td>
<td>0.08</td>
<td>0.08</td>
</tr>
<tr>
<td>Leimersheim</td>
<td>0.31</td>
<td>0.14</td>
<td>0.09</td>
<td>0.08</td>
<td>0.08</td>
<td>0.08</td>
</tr>
<tr>
<td>Leopoldshafen</td>
<td>0.49</td>
<td>0.43</td>
<td>0.32</td>
<td>0.37</td>
<td>0.33</td>
<td>0.33</td>
</tr>
<tr>
<td>Mörsch</td>
<td>0.42</td>
<td>0.18</td>
<td>0.14</td>
<td>0.25</td>
<td>0.19</td>
<td>0.14</td>
</tr>
<tr>
<td>Neuthard</td>
<td>0.46</td>
<td>0.25</td>
<td>0.15</td>
<td>0.22</td>
<td>0.28</td>
<td>0.15</td>
</tr>
<tr>
<td>Obergrombach</td>
<td>0.38</td>
<td>0.32</td>
<td>0.28</td>
<td>0.37</td>
<td>0.29</td>
<td>0.28</td>
</tr>
<tr>
<td>Oetzrot</td>
<td>0.29</td>
<td>0.25</td>
<td>0.19</td>
<td>0.25</td>
<td>0.17</td>
<td>0.24</td>
</tr>
<tr>
<td>Wendingen</td>
<td>0.29</td>
<td>0.15</td>
<td>0.11</td>
<td>0.19</td>
<td>0.16</td>
<td>0.17</td>
</tr>
<tr>
<td>Wolfartsweier</td>
<td>0.52</td>
<td>0.58</td>
<td>0.41</td>
<td>0.48</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
FIG. 4. Tritium concentration in soil humidity (December 1977) [8].

FIG. 5. Tritium concentration in soil humidity as a function of depth for two locations (see Fig. 4 in [8]).
higher concentrations near the surface suggest the impact of tritium releases into the atmosphere at the Karlsruhe Nuclear Research Center.

Another series of measurements carried out in the first days of June 1978 proves the impact of meteorological conditions on the tritium level. These measurements, made after a short period of abundant rainfall which contributed in three days about 25% of the annual mean value of precipitations and which was followed by hot and dry days, yielded distinct alterations in the tritium concentration in the soil. However, even under these extreme conditions tritium concentrations of some nCi/ltr were found in soil humidity.

5. TRITIUM IN PLANTS

Only limited studies were carried out on the tritium contamination of the free tissue waters of plants. To find the relationship of tritium to the relevant media close to the plants, the tritium concentration in air humidity, precipitations, soil humidity and ground water were investigated simultaneously [2, 8–10]. The
free tissue water of plant samples was extracted by means of azeotropic and vacuum distillations. A comparison of both methods furnished better agreement than expected, considering the number of possible errors.

Figures 7 and 8 contain examples of results of simultaneous measurements of the tritium concentration in pine needles, air humidity, soil humidity and precipitations.

In Table VII the maximum values are summarized of tritium concentrations measured in these programmes. For comparison, the minimum value found at the same location is also reported. The maximum tritium concentration determined in air humidity was 455 ± 13 nCi/ltr. These results show that monitoring of tritium concentration in plants in the vicinity of tritium emitters is necessary, particularly since rapid changes of tritium concentration in plants are possible.

6. MOVEMENT OF TRITIUM IN THE ENVIRONMENT

Figure 9 presents a simplified model of tritium transport in the environment. The diffusion of tritium in the atmosphere can be described with sufficient
FIG. 8. Results for the sampling location 'North-east' of simultaneous measurements of the tritium concentration in pine needles, air humidity and precipitations. For comparison calculated values of tritium concentration in air humidity are also plotted. The time of precipitation is indicated by the width of the columns [8].

TABLE VII. MAXIMUM VALUES OF TRITIUM CONCENTRATION OF FREE TISSUE WATER AND MINIMUM VALUES FOR THE SAME SAMPLING LOCATION

<table>
<thead>
<tr>
<th>Sampled material</th>
<th>Tritium concentration (nCi/litr)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Maximum</td>
<td>Minimum</td>
</tr>
<tr>
<td>Grass</td>
<td>21</td>
<td>1.9</td>
</tr>
<tr>
<td>Foliage (carpinus)</td>
<td>235 ± 6</td>
<td>0.95 ± 0.19</td>
</tr>
<tr>
<td>Foliage (quercus robur)</td>
<td>236 ± 9</td>
<td>1.11 ± 0.21</td>
</tr>
<tr>
<td>Needles (pinus sylvestris)</td>
<td>61.5 ± 1.9</td>
<td>13.1 ± 0.6</td>
</tr>
<tr>
<td>Needles (picea abies)</td>
<td>45.2 ± 1.4</td>
<td>2.5 ± 0.3</td>
</tr>
</tbody>
</table>
FIG. 9. Simplified model of tritium transport in the environment.

accuracy by the familiar dispersion formulae (compare Refs [11, 12]). They are valid for release periods of 1/2 to 1 h and can be verified in the experiment. However, the impact on the environment due to tritium releases is not so much determined by temporary dispersion conditions, but rather by the long-term diffusion factor. The experimental verification of the long-term diffusion factor, based on available tritium measurements, is not possible because the prevailing conditions are not well determined. In the case of the Karlsruhe Nuclear Research Center, in particular, the rates of tritium emission are not constant for the various emitters.

Tritium released via the waste water may lead to a radiation exposure of persons in several ways. For drinking water and respiration air these are:

Tritium seeping into the ground water from leakages in sewage treatment plants and in the sewer system as well as seeping from surface waters.

Evaporation from uncovered water surfaces while contaminating the environment via the air path.

With respect to the influences of temperature, wind speed and the difference between the saturation pressure and pressure prevailing of the water vapour in the air, the determination of the momentary evaporation rate is the most difficult problem.
By contrast, it is hardly possible to estimate with an acceptable accuracy the radiation impact from seeping. Experimental investigations are necessary in each individual case. Generally, leakages occurring in sewer systems are not adequately taken into account. However, if such systems carry tritium-bearing liquid effluents, contaminations become measurable. Tritium, which has reached the ground water, is diluted at a very slow rate only. Periods of months and years must be expected. Also the ground-water flow velocity is low (order of magnitude 1 m/d) so that the diffusion of a possible contamination is very low or the contamination is detected after a relatively long period only [13].

Surface water contaminated with tritium may entail radiation exposure also via the food chain on different exposure paths. The following principal paths have to be considered:

- Inhalation by cattle — milk or meat.
- Fodder and drinking water of cattle — milk or meat.
- Irrigation — vegetables.

The decisive elements with respect to the type of tritium contamination of plants are:

- The influence of the water vapour exchange and the direct influence of precipitation and dew on the surfaces of leaves and needles.

- The contamination of soil humidity by precipitations and the water exchange with the surrounding air.

The measured results do not indicate any direct relationship between the tritium concentrations in ground water and in plants. The influence of tritium in the soil becomes effective through the capillary water at different tritium contamination levels arising from the slow penetration of precipitations into the soil and through capillary water possibly contaminated by tritium in the ground water. The water taken up through the root system at different depths with varying levels of tritium contaminations has an average tritium concentration which results from the different levels of contamination in the soil. In many cases the tritium level in leaves is close to the background level of tritium concentration in air humidity. On the other hand, in accordance with the contamination level of air humidity, there are maximum levels of tritium concentration in the tissue water of plants much higher than the tritium level in soil humidity. The fact that the tritium levels in leaves and needles of trees are close to the tritium levels of air humidity and of the wetting precipitation water demonstrates that water vapour is quickly exchanged between the atmosphere and the leaves and needles respectively. In the case of grass, tritium concentrations were generally identical in the tissue water of plants and in the soil humidity in the range of roots.
The variation of the tritium concentration in tissue water of plants enables the time constants and the half-lives of the process to be determined. The following values are found: for oak and hornbeam leaves $2 \pm 1$ days, for spruce needles $3 \pm 1.5$ and $4 \pm 2$ days, for pine needles $4 \pm 2, 6 \pm 3$ and $9$ days. The time sequence of measurement points and the experimental conditions were not sufficient to make more accurate statements and to record a component having a shorter half-life. However, the time constants and half-lives are expected to be dependent on temperature, air humidity and the phase of the plant, so that the changes with time of tritium concentration in plants cannot be described easily by one constant.

The variety of pathways for the transport of tritium in the environment indicates the need for a long-term monitoring programme which includes the critical pathway. One should be aware that in many countries the food is not consumed at the place of production so that every estimate of tritium exposure may be rather inaccurate.

7. RADIATION EXPOSURE OF PERSONS FROM THE TRITIUM EMISSIONS OF THE KARLSRUHE NUCLEAR RESEARCH CENTER

Assuming that the mean tritium concentration in the human body equals the mean tritium concentration in drinking water and soil humidity, the radiation exposure of the population by tritium can be estimated. One obtains 33 or 100 µrem/a for the population of Leopoldshafen, the village with the maximum exposure in the vicinity of the Karlsruhe Nuclear Research Center. A considerable part of this dose comes from the world-wide fall-out. Within the area of maximum contamination food is not produced and persons normally stay no longer than 50 hours weekly. This means that tritium concentration of few pCi/ml at the maximum should collect in the body and, consequently, the radiation impact remains below 1 mrem/a.

It should be taken into account that because of the rapid changes of tritium concentration in water and food, and the variation in consumption habits, each dose estimate achieves but a limited accuracy.

ACKNOWLEDGEMENT

The measurements reported here were performed within the framework of a co-ordinated programme of research under the sponsorship of the I.A.E.A. (Research Agreement No. 1302/CF).
REFERENCES


DISCUSSION

J. DELMAS: Have you had any large-scale atmospheric point releases, and if so have you made any observations of the $^3$H content in the dry tissue of plants? Did you then observe any significant variation at the time of such accidental release, and was the variation the same irrespective of whether the release took place by day or at night?

L.A. KÖNING: We carried out no measurements of the tritium concentration in the dry tissue of plants, but we also had no large-scale accidental point releases
of tritium to the atmosphere. No differences in the effects as between day and night could be observed.

G.R. NEWBERY (Chairman): Were you able to make measurements, particularly of dry material, at times related to periods of peak discharges, or are your measurements made independently of such periods?

L.A. KÖNIG: Peak discharges did not particularly influence the tritium contamination level of the environment of the Karlsruhe Nuclear Research Center. The variations depend on the duration of exposure and other conditions. Short exposures cause no significant changes.

K.J. VOGT: Commenting on the tritium transport model shown in your Fig.9, I would mention that our own calculations demonstrate that the direct contamination of vegetation by precipitation under certain conditions may be a relevant pathway and should be included in the model.

If environmental HTO measurements of soil water and vegetation have provided evidence that near the sources the deposition of HTO by washout plays the dominant role and exceeds the contribution of dry HTO deposition from the air humidity, then the washout effect near elevated sources must be taken into consideration.

L.A. KÖNIG: It is certainly necessary to do this. In the case of the Karlsruhe Nuclear Research Center, maximum tritium concentrations were found in soil humidity near the main tritium emitters with stack heights up to 100 m. However, it should not be forgotten that we have several emitters, and the tritium concentration in soil humidity at a given location is influenced by the releases from all emitters.

H. BONKA: You referred to a relationship between the tritium concentration in air humidity and in plants. Did you carry out measurements when it was raining? I wonder how fast the tritium concentration in the plant changes under such conditions, and whether equilibrium is re-established after a long rainy period.

L.A. KÖNIG: As stated in my paper, the tritium concentration in leaves and pine needles depends greatly on actual wetting precipitation. Precipitation causes fast variations. The effect of the tritium in air humidity on vegetation depends on the exposure time. This holds good for tall plants. For low plants, such as grass, the tritium concentration in the tissue water corresponds to that in the soil humidity.

K. HUBEL: Figure 1 indicates that over the years there has been a decrease in airborne tritium releases from the Karlsruhe Center and an increase in releases with effluent waters. Is this to be explained in terms of the technical features of the various facilities, or of radiation protection measures, for example?

L.A. KÖNIG: During the early years, tritium was released during incidents involving losses of heavy water at the heavy-water-moderated reactors. The heavy water had tritium concentrations of some Ci/litr. Such incidents are rare now. The increase in releases with the effluent water is attributable mainly to the operation of the reprocessing plant.
MONITORING EXPERIENCES WITH TRITIUM IN JAERI

Y. YOSHIDA, A. YABE, M. KISHIDA
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Abstract

MONITORING EXPERIENCES WITH TRITIUM IN JAERI.

In the Japan Atomic Energy Research Institute, one of the most important sources of internal exposure from tritium has been airborne tritium from the heavy-water-moderated research reactors JRR-2 and JRR-3. Handling of the tritium gas targets of the accelerators also causes internal exposure to tritium. Tritium monitoring techniques developed in JAERI are described, and experiences in operational health physics. The values of personnel doses from tritium determined by urine analysis agreed with those of doses estimated from air monitoring results during repair work in JRR-2. In addition to urine analysis, measurement of tritium concentration in the expired air of workers has also been used to check internal exposure. The experimental investigation showed that the concentrations of tritium in the samples of the expired air agree well with those in the urine samples taken a few hours after inhalation. The individual's biological half-lives were determined through the follow-up measurements. The half-lives obtained for 38 persons ranged from 5 to 16 days, with an average of 9.8 ± 4.1 days.

In the Japan Atomic Energy Research Institute (JAERI), one of the most important sources of internal exposure from tritium has been airborne tritium from the heavy-water-moderated research reactors JRR-2 and JRR-3 [1]. The tritium concentrations in the heavy water in these reactors are both at present nearly 1 mCi/cm³, and the annual release of tritium in gaseous and liquid wastes amounts to several tens of curies and several hundred curies respectively. In addition, handling of tritium gas targets in accelerators also causes internal exposure by tritium. The tritium monitoring techniques that have been developed and experiences in operational health physics in JAERI are here described.

1. MONITORING TECHNIQUES

1.1. Area monitoring

Monitoring techniques of tritium in the working environment in JAERI are summarized in Table I, and the detection limits of these techniques are also shown.
### TABLE I. MONITORING TECHNIQUES OF TRITIUM IN JAERI

<table>
<thead>
<tr>
<th>Contamination measured</th>
<th>Sampling method</th>
<th>Instrument or sampling material</th>
<th>Detection limit (μCi/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air contamination</td>
<td>Direct sampling</td>
<td>Ionization chamber (volume: 1500 cm³) vibrating reed electrometer</td>
<td>$2 \times 10^{-6}$</td>
</tr>
<tr>
<td></td>
<td>Condensation</td>
<td>Ionization chamber type monitor (compensated for gamma) Cold trap, L.S.C.(^a)</td>
<td>$10^{-6} - 10^{-5}$</td>
</tr>
<tr>
<td></td>
<td>Adsorption</td>
<td>Silica gel, L.S.C.(^a)</td>
<td>$10^{-10}$</td>
</tr>
<tr>
<td></td>
<td>Bubbling</td>
<td>Water bubbler, L.S.C.(^a)</td>
<td>$10^{-6}$</td>
</tr>
<tr>
<td>Surface contamination</td>
<td>Smear</td>
<td>Glycerol – ethanol loaded filter paper, gas flow counter</td>
<td>$10^{-4}$ (μCi/cm²)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Filter paper – L.S.C.(^a)</td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) Liquid scintillation counter.

#### 1.1.1. Air contamination

Concentration of tritium in the form of water vapour or hydrogen gas in air is determined by measuring the current of an ionization chamber with which the air is sampled at the place in question. Two types of tritium air monitor, one compensated for gamma rays and one uncompensated, have been used. The former is used mainly as an installed monitor and the latter as a portable one. Sampling air is taken into an ionization chamber through a filter paper to remove the particulate aerosol.

The concentration of tritiated water vapour in air such as HTO or DTO is obtained through the liquid scintillation counting of the activity of tritium in the water sample which is collected using a cold trap or adsorbent such as silica gel. These methods have the advantages of separating tritiated water vapour from other gases such as \(^{41}\)Ar and of very high sensitivity (less than $10^{-10}$ μCi/cm², cf. Table I).

In the cold trap method, a water sample is collected by condensing tritiated water vapour in the sampling air using a freezing mixture such as a dry ice – ethanol mixture. The concentration of tritiated water vapour in air is determined from
the measured activity of tritium in the condensed water, the relative humidity and the density of saturated water vapour which is calculated depending on the temperature of the sampling air. This method is applied mainly for measuring the concentration of tritiated water vapour in the working area. The adsorption method is used mainly for obtaining the average concentration of tritiated water in air for an extended period such as in stack monitoring [2].

1.1.2. Surface contamination

Monitoring of tritium surface contamination has been made through counting smear samples with a 2π gas flow counter or a liquid scintillation counter. Filter papers loaded with 25 wt\% glycerol in ethanol are used for the smear in order to hold the tritium collected in the samples and also to count fairly efficiently.

Sampling efficiency (the ratio of the activity of the smear paper to the total activity of surface contamination) of tritium and the counting efficiency for a 2π gas flow counter are related to the percentage of glycerol in ethanol. The above percentage of glycerol was selected in order to measure tritium surface contamination most effectively, as seen in Fig.1 [3].
<table>
<thead>
<tr>
<th>Frequency of analysis</th>
<th>Routine monitoring</th>
<th>Non-routine monitoring</th>
</tr>
</thead>
<tbody>
<tr>
<td>Subject</td>
<td>Persons working at the area where tritium is handled</td>
<td>Any persons whose doses are estimated to be over 10 mrem by area monitoring</td>
</tr>
<tr>
<td>Sample volume</td>
<td>Every 3 months</td>
<td>At the start, every day, then, every 3 to 4 days</td>
</tr>
<tr>
<td>Time of collecting sample</td>
<td>About 50 ml</td>
<td>About 50 ml</td>
</tr>
<tr>
<td>Interpretation of results</td>
<td>At least 1 h after end of work</td>
<td>Initially about 1.5 h after acute intake</td>
</tr>
<tr>
<td></td>
<td>Above 80 pCi/ml: inquiry for contamination</td>
<td>Above 1 nCi/ml: dose estimation</td>
</tr>
</tbody>
</table>

1.2. Individual monitoring for internal contamination

Urine analysis is made to estimate the internal dose due to tritium whenever the dose of persons is estimated to be above 10 mrem by the measurement of tritium concentration in air in the working area. Urine analysis is also made routinely every three months for persons working in the areas where tritium is handled.

In addition, the measurement of the tritium concentration in the expired air of workers has been carried out to check internal contamination as an auxiliary method in support of urine analysis in JAERI.

1.2.1. Urine analysis

The procedure of urine analysis for tritium in individual monitoring is summarized in Table II [4]. The method of collecting urine is also shown in this table.

For urine monitoring in JAERI, purification of the urine such as decolorization and de-ionization is not made for liquid scintillation counting assuming that the quenching and contribution from radioactive materials other than tritiated water are small. In order to detect 80 nCi per litre of urine, i.e. the checking level of significantly contaminated persons in JAERI for three months sampling, the purification of urine is not required because of advanced liquid scintillation counting techniques, unless large quantities of other radioactive materials exist in the urine.

The toluene-ethanol scintillator solution, a mixture of PPO, POPOP, ethanol and toluene, or the emulsion scintillator solution composed of PPO, detergent
(nonylphenoxypolyethoxyethanol) and toluene is used for monitoring because of low cost and its ready availability.

The samples of untreated urine mixed with these scintillator solutions are counted using a low-background liquid scintillation counter which has a coincidence and an anti-coincidence circuit and is shielded with lead.

1.2.2. Measurement of expired air

Measurement of tritiated water vapour in expired air is more convenient than urine analysis for checking internal contamination due to tritium, because collection of tritiated water from the expired air can be made rapidly using a cold trap, although there is a possibility of cross contaminations when collection is made near the working area. One millilitre of the condensed water, sufficient for measuring by liquid scintillation counting, is collected by about 20 deep breathings [5].

Depending on the circumstances, a column containing 2 g of molecular sieve is used instead of a cold trap. The quantity of water collected by the molecular sieve depends on the temperature of the atmosphere and the total volume of air passing through the column. However, 15 ~ 20 deep breathings are sufficient for collecting a water sample.

For counting, the sieve is mixed with scintillator solution in a vial immediately after collection, and heated at 100°C for 5 min to desorb the tritiated water into the scintillator solution, followed by 1 h cooling. Counting results should be corrected for the quenching caused by the molecular sieve.

2. EXPERIENCES IN MONITORING

2.1. Relation between air monitoring and urine analysis

During the repair of JRR-2 in the middle of the 1960s there was a risk of tritium being taken into the body because a reservoir of the activated heavy water coolant was opened to the air and the tritiated water vapour was dispersed fairly uniformly in the working area. During the repair work the internal doses estimated from the results of air monitoring were compared with those determined by urine analysis.

From the results of air monitoring, the internal doses were evaluated by the formula (1) derived on the basis of continuous intake using the values of the ICRP Committee II.

\[ H = 4 \times 10^5 \times C \times t \] (1)
where \( H \) is the dose commitment to the body tissue (mrem), \( C \) is the average concentration of tritium in the air (\( \mu \text{Ci/cm}^3 \)), and \( t \) is the working time (h).

By urine analysis, the doses in individual persons were determined by the following formula (2):

\[
H = 0.74 \times U \times T
\]

(2)

where \( H \) is the dose commitment (mrem), \( U \) is the initial concentration of tritium in urine (\( \mu \text{Ci/ltr} \)) and \( T \) is the effective (biological) half-life (d) of 12 days. In this case, the value of \( U \) was obtained by plotting urinary concentrations against time, and by extrapolating the plot to the time when a possible intake might have occurred.

It is seen from Fig. 2 that there is a fairly good agreement between the values of doses determined by urine analysis and those of doses estimated from the concentration in air, except at low doses.

2.2. Expired air measurement versus urine analysis

In monitoring by measuring expired air it is necessary to know the tritium concentration in the moisture of expired air relative to that in the body fluids as
a function of time following inhalation because it takes a few hours before uniform mixing of tritiated water is attained in the body fluids, as is well known [6].

Figure 3 shows the change in tritium concentrations in urine and in moisture of expired air as a function of time of sampling following the inhalation. It is seen that the tritium concentration in the moisture of expired air sampled 1 h or less after inhalation is about 10 times higher than that 3–4 h after inhalation; the latter is similar to that in urine samples collected a few days after inhalation.

This tendency to show relatively high values in the samples of expired air moisture taken shortly after inhalation may be due to the initial deposition in the lungs before its absorption into the body fluids. If assimilation of tritium occurred only through the skin, there would be a slow rise in the early period after skin absorption, unlike the case of the inhalation. To check whether an appreciable quantity of tritium is inhaled or not, the sampling of expired air moisture can be made immediately after contamination.

It is evident that the results of the measurement of the expired air moisture sampled 3–4 h after inhalation of tritium can be used in much the same way as those of urine analysis for estimating doses in workers.
2.3. Biological half-life

In the cases where it is possible to follow the change in internal contamination due to tritium with time, follow-up measurement by urine analysis is made to determine the individual values of the biological half-life of tritium in order to evaluate the exact doses.

Figure 4 shows the distribution of the individual biological half-lives among 38 persons whose initial tritium concentrations in urine were over 1 nCi/ml.

In the case of a body burden much above the maximum permissible level, the presence of longer-lasting components in the biological half-life of tritium is reported [7], but in JAERI, only the short component has been observed because of the very low level of exposures.

The biological half-lives for the 38 persons ranged from 5 to 16 days, with an average of $9.8 \pm 4.1$ days (90% confidence level). These values agree with Butler's results of 4–18 (range) and $9.5 \pm 4.1$ days (average) [8]. It is interesting that fairly good agreement was obtained between the Japanese data and data from the west, although there are significant differences in such matters as body build and food habits.
ACKNOWLEDGEMENTS

The authors are indebted to M. Fujita, J. Akaishi and H. Yamamoto for valuable discussions regarding the preparation of this report.

REFERENCES


DISCUSSION

R.V. OSBORNE: In your review of monitoring methods you did not mention how you measure tritium in your liquid effluents. Do you have an on-line method?

A. YABE: At our Institute, most of the tritium discharged to the on-site and off-site environments is monitored at the exits (usually the stacks) of the exhaust air systems in the case of tritium gas, and at the exits of the pipes or tanks in that of tritiated water. The pipes lead to larger channels, which then discharge to the sea, our Institute being located by the Pacific Ocean. The liquid effluents are monitored for tritium at the point of release to the sea.

With the help of measurements of the tritiated water concentration we keep the concentration in liquid effluents to within $3 \times 10^{-3}$ μCi/cm³, i.e. one tenth of the ICRP maximum permissible concentration.

In monitoring our liquid effluents we employ conventional methods of measurement, i.e. the effluents in the individual disposal tanks are periodically sampled before release and are measured with liquid scintillation counters. Specialized techniques such as on-line measurement methods have not yet been introduced.

G.R. NEWBERY (Chairman): Did you observe any correlation between the drinking habits of your 38 subjects and the measured effective half-life of tritium in urine? We have not been able to detect anything of the kind at Amersham.
A. YABE: No, we have observed no such correlation either, though our main concern in monitoring for internal contamination is to detect significant quantities of tritium in the body. Of course, fluid intake is rather dependent upon the ambient temperature, and hence the effective half-life of tritium may vary seasonally. However, I feel that our data from 38 subjects are insufficient to discern such a trend if it exists.
ENVIRONMENTAL ASPECTS OF TRITIUM FROM CANDU STATION RELEASES

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Abstract

ENVIRONMENTAL ASPECTS OF TRITIUM FROM CANDU STATION RELEASES.
The sources, production and releases of tritium in gaseous and liquid effluents from CANDU stations as well as applicable regulatory and derived emission standards are discussed. To assess the environmental impact of these releases, Ontario Hydro has been carrying out an environmental monitoring programme at all nuclear generating sites since 1971. The programme monitors critical and potentially important pathways and consists of monitoring tritium in air, precipitation, milk, drinking water and vegetation. Tritium releases from stations are of the order of 0.1% of the derived release limits leading to doses to individuals in the vicinity of the stations of about 1 mrem/a⁻¹. The environmental impact from an expanded CANDU programme is discussed. The effects on the Great Lakes basin are of particular importance since it is expected that the bulk of Ontario Hydro’s future nuclear sites will be located along the shores of the Great Lakes. The implication of the recently promulgated International Joint Commission water quality objective is examined.

1. INTRODUCTION

Ontario Hydro, the publicly owned electric power utility for the Province of Ontario, currently operates 10 reactors of the CANDU (Canadian Deuterium Uranium) type with a total gross generating capacity of 5567 MWₑ. Another 12 reactors with a total gross capacity of 8724 MWₑ are under construction or in the final design stages. The current and projected stations are listed in Table I. Nuclear power currently accounts for about 20% of the total electric generating capacity of the province and is expected to rise by another 25 000 MWₑ to over 50% of the total by 2000.

2. THE CANDU REACTOR

The CANDU reactor uses natural uranium fuel and heavy water (D₂O) as moderator and coolant. The basic fuel unit is a fuel bundle 50 cm in length and 10 cm in diameter containing about 20 kg of natural uranium dioxide (UO₂). Each fuel bundle is made up of a number of cylindrical pellets of UO₂ sealed in a thin zircalloy sheath.
TABLE I. ONTARIO HYDRO NUCLEAR GENERATING STATIONS

<table>
<thead>
<tr>
<th>Name</th>
<th>Number of units</th>
<th>Maximum continuous rating (MWₜgross)</th>
<th>In service date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operating</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1. Nuclear Power Demonstration (NPD)</td>
<td>1 X 25</td>
<td>25</td>
<td>1962</td>
</tr>
<tr>
<td>2. Douglas Point</td>
<td>1 X 218</td>
<td>218</td>
<td>1968</td>
</tr>
<tr>
<td>Under construction</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Under design</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The reactor itself consists of an array of horizontal zircalloy high pressure tubes running through low pressure tubes in a cylindrical tank (calandria) containing the heavy-water moderator. Each pressure tube contains 12 or 13 fuel bundles depending upon the particular reactor design. A unique feature of the system is that it allows on-power refuelling. Heat is removed by the flow of pressurized heavy water within the pressure tubes, and transferred to ordinary water in boiler heat exchangers to produce steam to run the turbines.

The major source of tritium in a CANDU reactor is neutron activation of the heavy-water moderator and coolant by the reaction $^2H(n,γ)^3H$ with a resultant production rate of about 2400 Ci $\text{MW}_\text{e}^{-1}$ a$^{-1}$.

3. TRITIUM RELEASES AND ENVIRONMENTAL STANDARDS

All nuclear facilities in Canada are subject to the federal Atomic Energy Control Act which is administered by the Atomic Energy Control Board (AECB). Limits on the emission of radionuclides to air and water are based upon the dose limits for members of the general public recommended by the ICRP [1] and required by the AECB [2]. In the case of tritium oxide the applicable limit is 0.5 rem a$^{-1}$ to the whole body. Since it is not practical to measure the radiation dose to individual members of the public, secondary standards or derived release limits (DRL’s) are set by the utility subject to approval by the AECB.
The permissible continuous release rate for an airborne radionuclide is given by the following relationship:

\[ Q = \frac{\text{MPC}_a}{K_a} \text{ Ci s}^{-1} \]  

(1)

\( K_a \), the atmospheric dilution factor, is a function of the distance from the source, the effective height of release, the weather and the averaging time. The weighted mean dilution factors calculated by Bryant [3] are considered to be representative of meteorological conditions in southern Ontario. For airborne tritium exposure the critical pathway is inhalation and absorption through the skin, hence the MPC\(_a\) is applied at the station exclusion boundary (approximately 1 km from station structures) beyond which permanent residence by members of the public is considered to be possible.

The maximum permissible concentration of tritium in any liquid effluent stream is taken to be the MPC\(_w\) for members of the public. There is, in addition, a total annual curie limit on all liquid releases.

Although the legal limit for exposure of a member of the public is 500 mrem a\(^{-1}\), Ontario Hydro has adopted a design and operating target of 1% of the derived release limits, and in most cases the actual releases are less than the 1% target. Table II contains tritium release data from the currently operating nuclear electric generating stations over the period 1971-1977. Average total releases are 730 Ci (MW\(_{e-a}\))\(^{-1}\) for NPD, 190 Ci (MW\(_{e-a}\))\(^{-1}\) for Douglas Point, and 24 Ci (MW\(_{e-a}\))\(^{-1}\) for Pickering. Bruce BGS A is not included as it is not yet fully in service. The NPD and Douglas Point reactors are prototypes and do not reflect current technology. It is anticipated that releases from future plants will be of the same order as those from Pickering.

4. ENVIRONMENTAL MONITORING

Ontario Hydro carries out an environmental monitoring program around all of its nuclear power stations with the objectives of:

1. Evaluating the potential radiation dose to members of the public from releases of radioactive materials in station effluents to demonstrate compliance with regulatory dose limits.

2. Obtaining a body of factual data which can be used to derive correlations between actual releases and environmental radioactivity levels.

3. Detecting any long-term changes or trends in environmental radioactivity levels resulting from the releases.
<table>
<thead>
<tr>
<th>Year</th>
<th>NPD</th>
<th>Douglas Point</th>
<th>Pickering</th>
</tr>
</thead>
<tbody>
<tr>
<td>1971</td>
<td>5.820</td>
<td>24.990 (0.060)</td>
<td>2.670 (0.026)</td>
</tr>
<tr>
<td>1972</td>
<td>3.640</td>
<td>7.280 (0.018)</td>
<td>14.300 (0.014)</td>
</tr>
<tr>
<td>1973</td>
<td>13.100</td>
<td>20.000 (0.048)</td>
<td>40.000 (0.038)</td>
</tr>
<tr>
<td>1974</td>
<td>7.740</td>
<td>5.040 (0.043)</td>
<td>24.990 (0.012)</td>
</tr>
<tr>
<td>1975</td>
<td>12.840</td>
<td>14.840 (0.013)</td>
<td>20.320 (0.020)</td>
</tr>
<tr>
<td>1976</td>
<td>13.290</td>
<td>34.470 (0.083)</td>
<td>24.290 (0.023)</td>
</tr>
<tr>
<td>1977</td>
<td>13.290</td>
<td>11.870 (0.048)</td>
<td>11.870 (0.042)</td>
</tr>
</tbody>
</table>

Gaseous releases (C1) Liquid releases (C1)

Douglas Point Pickering

Numbers in parentheses denote the percentage of the derived emission limit.
TABLE III. AVERAGE AIRBORNE TRITIUM CONCENTRATION AT 1 km FROM ONTARIO HYDRO’S NUCLEAR POWER STATIONS

<table>
<thead>
<tr>
<th>Location</th>
<th>Number of monitoring sites</th>
<th>Average air concentration (% public MPC&lt;sub&gt;9&lt;/sub&gt;)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NPD</td>
<td>4</td>
<td>1974: 0.13, 1975: 0.06, 1976: 0.14, 1977: 0.07</td>
</tr>
<tr>
<td>Pickering NGS ‘A’</td>
<td>4</td>
<td>1975: 0.09, 1976: 0.03, 1977: 0.07, 1978: 0.21</td>
</tr>
<tr>
<td>BNPD</td>
<td>(Douglas Point; Bruce NGS ‘A’)</td>
<td>8</td>
</tr>
</tbody>
</table>

* One public MPC<sub>9</sub> of tritiated water vapour = 3 x 10<sup>-7</sup> Ci m<sup>-3</sup> [4].

Because tritium is the major radionuclide released, the environmental monitoring program is heavily weighted toward measurement of that radionuclide. In particular, measurements are made of tritium in air, precipitation, milk, drinking water and vegetation.

The tritium concentration in air is measured at environmental monitoring sites placed on the boundary of the exclusion area at about 1 km from the station. A reference site is located at a distance of 10-15 km. Airborne water vapour is collected by drawing air through molecular sieve at a flow rate of 50-100 cm<sup>3</sup> min<sup>-1</sup>. Sampling is continuous with an integration period of one month, during which 2-4 m<sup>3</sup> of air are passed through the collector. The absorbed water is removed from the sieve by heating and the tritium content determined by liquid scintillation counting. Tritiated water in milk and vegetation is extracted by freeze drying or by azeotropic distillation in toluene.

5. ENVIRONMENTAL TRITIUM LEVELS AND DOSE ESTIMATES

Annual average tritium in air concentrations at the site boundaries are generally of the order of 0.1% of the MPC<sub>9</sub> (3 x 10<sup>-7</sup> Ci m<sup>-3</sup>) for continuous exposure of members of the public [4]. Background levels are of the order of 0.001% of the MPC<sub>9</sub>. Table III contains data for NPD, Pickering and the Bruce Nuclear Power Development over the period 1974-1977. These levels correspond to whole body doses in the range of 0.1-1 mrem a<sup>-1</sup>.

Long-term atmospheric dilution factors calculated as the quotient of the air concentration (Ci m<sup>-3</sup>) divided by the release rate (Ci s<sup>-1</sup>) are in the range of 10<sup>-7</sup> to 10<sup>-6</sup> s m<sup>-3</sup>, in reasonable agreement with theoretical prediction. Table IV gives representative data from Pickering NGS A taken over a period of five years.
### TABLE IV. LONG-TERM ATMOSPHERIC DILUTION FACTORS AT PICKERING GENERATING STATION (1973–77)

<table>
<thead>
<tr>
<th>Site number&lt;sup&gt;a&lt;/sup&gt;</th>
<th>$K_a$ ($s , m^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$(3.0 \pm 1.1) \times 10^{-7}$</td>
</tr>
<tr>
<td>2</td>
<td>$(4.5 \pm 2.0) \times 10^{-7}$</td>
</tr>
<tr>
<td>3</td>
<td>$(2.0 \pm 1.0) \times 10^{-7}$</td>
</tr>
<tr>
<td>4</td>
<td>$(1.6 \pm 1.2) \times 10^{-7}$</td>
</tr>
<tr>
<td>5</td>
<td>$(4.0 \pm 1.4) \times 10^{-8}$</td>
</tr>
<tr>
<td><strong>Average of sites 1–4</strong></td>
<td>$(2.8 \pm 1.3) \times 10^{-7}$</td>
</tr>
<tr>
<td><strong>Theoretical $K_a$ at 1 km</strong></td>
<td>$9 \times 10^{-7}$</td>
</tr>
</tbody>
</table>

<sup>a</sup> Sites 1–4 lie at a distance of approximately 1 km from the station. Site 5 is at a distance of 13 km. Errors are ±1σ on the mean of a 5-year average.

### TABLE V. DOSE ESTIMATES FROM TRITIUM RELEASES FROM A TYPICAL CANDU STATION

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Annual individual dose (mrem) as a function of distance from the station</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 km</td>
</tr>
<tr>
<td>(1) Inhalation and skin absorption</td>
<td>0.5</td>
</tr>
<tr>
<td>(2) Ingestion of milk&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.1</td>
</tr>
<tr>
<td>(3) Ingestion of home-grown fruits and vegetables</td>
<td>0.1</td>
</tr>
<tr>
<td>(4) Drinking water</td>
<td>0.06</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td><strong>0.8</strong></td>
</tr>
</tbody>
</table>

<sup>a</sup> Milk is from farms at a distance of 10–15 km from station.
Analysis of raw milk from farms around Pickering NGS and Bruce NPP indicate tritium concentrations in the range of 300-5000 pCi l⁻¹. This corresponds to a whole-body dose rate in the range of 0.02-0.4 mrem a⁻¹ to an infant consuming 0.7 litres of milk per day. Studies of the water contained in such food crops as apples, corn, tomatoes, pears, and cabbage grown within a 15-km radius of Pickering NGS show tritium levels ranging from 100 mCi l⁻¹ within the 1 km exclusion area to less than 1 nCi l⁻¹ at 5 km. Dose estimates range from 0.05 mrem a⁻¹ at 1 km to less than 0.01 mrem a⁻¹ at 10 km. These estimates are based on an average daily consumption over a period of 150 days of 380 g of fresh fruit and vegetables with an average water content of 80%.

Drinking water is monitored where a municipal intake could be affected by plant releases. Data from the Pickering water pumping station over several years show an average tritium concentration of 750 pCi/l (0.014% MFCₚ) for an annual average whole body dose of 0.06 mrem to an adult consuming 2200 ml d⁻¹.

Major identified pathways to man of tritium from station releases are 1) inhalation and skin absorption of airborne tritiated water vapour, 2) ingestion of milk, 3) ingestion of fruits and vegetables, and 4) ingestion of water. Dose estimates for an individual resulting from tritium emissions from a typical CANDU station are presented in Table V.

These are maximum estimates, particularly with regard to the milk pathway, as the milk from the monitored farms will be diluted with other milk of a lower tritium content from more distant farms before distribution to the public.

6. IMPACT OF FUTURE EXPANSION ON THE GREAT LAKES

A recommendation from the International Joint Commission (IJC) on radioactivity standards for the Great Lakes has recently been submitted to the governments of the United States and Canada for ratification. The standard is in terms of a dose equivalent to the ICRP Reference Man from a standard annual intake (2200 ml d⁻¹) of Great Lakes water. The recommended objective for general water quality in the Great Lakes outside of the Source Control Area, is that level of radioactivity which results in a whole body dose equivalent not exceeding 0.1 mrem. The Source Control Area is bounded by a radius of 1 km from the point of release. The limiting dose of 1 mrem includes contributions from all sources, including fallout.

At the present time the effective dose equivalent from one year's intake of lake water due to uncontrollable sources such as naturally occurring radionuclides or fallout from weapons testing is 0.019 mrem from tritium, 0.079 mrem from Sr-90, 0.013 mrem from Ra₂₂⁶ and 0.001 mrem from Cs-137, for a total of 0.11 mrem. This means that about 0.9 mrem
is available for other sources including nuclear power. Since it is anticipated that the bulk of Ontario Hydro’s future nuclear sites will be located along the shores of the Great Lakes it is of particular interest to assess their releases in light of the IJC water quality objective.

A chain lake model has been used as shown in Figure 1. Assuming uniform mixing the time variation of concentration in a lake at time \( t \), \( C(t) \) is given by

\[
\frac{dC(t)}{dt} = \frac{R(t)}{V} - \lambda C(t) \tag{2}
\]

where

\( R(t) = \text{Tritium input rate to lake} \ (\text{Ci a}^{-1}) \)

\( V = \text{lake volume} \ (\text{m}^3) \)

\( \lambda = \lambda_r + \lambda_e \)

\( \lambda_r = \text{tritium decay constant} \)

\( = 0.056 \ \text{a}^{-1} \)

\( \lambda_e = \frac{q}{V} = \text{physical removal constant from lake} \) \ (\text{a}^{-1})

\( q = \text{rate of water outflow from lake} \ (\text{m}^3 \ \text{a}^{-1}) \)
the equations have been solved numerically based on two scenarios.

A. A constant tritium release rate of 19 Ci(MW$_e$ - a)$^{-1}$ to air and 5 Ci(MW$_e$ - a)$^{-1}$ to water over the lifetime of the station.

B. A tritium release rate increasing linearly from the values in scenario A at five years of operation to 50 Ci(MW$_e$ - a)$^{-1}$ to air and 80 Ci(MW$_e$ - a)$^{-1}$ to water after 30 years of operation. These latter values correspond roughly to the operating target of 1% of the derived release limits.

The following assumptions are common to both scenarios.

1. Tritium inputs to the lakes are from the Ontario Hydro stations listed in Table I plus one additional station on Lake Ontario in the period 1988-1990.

2. All liquid releases and 50% of the airborne releases (via exchange, rainout and run-off) ultimately enter the lakes.

3. A mean capacity factor of 85% for the nuclear stations.
4. Bomb fallout tritium levels in the lakes are assumed to be 400 pCi l⁻¹ in 1970.

The results are shown in Figure 2 for lakes Huron and Ontario. If the tritium releases from the nuclear generating stations were to increase from their present levels such that the operating targets were reached after 30 years, then the tritium concentration in Lake Ontario could reach 1.5 mCi l⁻¹. This corresponds to an individual whole-body dose of 0.1 mrem a⁻¹.

We therefore believe that Ontario Hydro will meet the IJC ambient water quality objective for the Great Lakes, although continued monitoring of lake tritium levels will be required to verify the models.

7. CONCLUSIONS

Ontario Hydro is committed to the use of nuclear power for a significant fraction of the total electric generating capacity of the province. The current gross nuclear capacity is 5567 MWₑ and this could rise to 30 000 MWₑ by 2000.

Because of the large inventory of heavy water, the major radionuclide released to the environment from a CANDU nuclear electric generating station is tritium arising from neutron activation of deuterium. Current experience has shown that the dose to an individual living continuously at the boundary of a large CANDU station is of the order of 1 mrem a⁻¹ or less due to tritium releases. This is about 1% of the dose from natural background radiation and well within the range of normal fluctuations in the natural background.

Since almost all of the Ontario Hydro nuclear installations are expected to be along the shores of the Great Lakes, special attention must be given to the buildup of contamination in the lakes. Calculations show that given a conservative scenario whereby the stations release tritium at 1% of the derived limits after 30 years of operation (current releases are of the order of 0.1% of the limits) the tritium concentration in Lake Ontario could reach 1.5 mCi l⁻¹ by the year 2000. This would correspond to an annual contribution to the dose from drinking water of 0.1 mrem a⁻¹ which is well within the ambient water quality objective proposed by the International Joint Commission.

REFERENCES


DISCUSSION

R.M. CHATTERJEE: I should like to comment that the objectives you mentioned are still 'proposed objectives'. They were not formulated by the International Joint Commission, but rather by two expert committees, one in the United States of America and the other in Canada, which submitted the draft objectives to the respective governments for ratification. The I.J.C. has not been officially informed of the objectives as yet.

You discussed values of dose equivalents from one year's intake of lake water from uncontrollable sources such as natural radioactivity or weapons fall-out. How were you able to separate the contributions of natural radioactivity and weapons fall-out from that of nuclear reactors?

D.J. GORMAN: The $^{137}\text{Cs}$ is likely to be due to fall-out since no $^{134}\text{Cs}$ is observed; $^{134}\text{Cs}$ is an activation product specific to reactor releases and is not found in weapons fall-out.

Typical $^{137}\text{Cs}/^{90}\text{Sr}$ ratios in liquid effluents are of the order of 50–100:1. Since we see neither $^{137}\text{Cs}$ nor reactor $^{134}\text{Cs}$, we would not expect to see $^{90}\text{Sr}$ either.

The tritium measurements are consistent with general background levels found in the other Great Lakes and in streams feeding Lake Ontario.

H.C. ROTHSHILDS: Have you done any calculations of the collective dose resulting from the Pickering tritium releases?

D.J. GORMAN: Consequent upon 1977 releases from the Pickering station the dose to the population around Lake Ontario from drinking water is about 44 man-rem.

H. BONKA: In your dose calculations, did you base yourself on the average food consumption over a fairly large area, e.g. a 50-km circle as in United Kingdom practice?

D.J. GORMAN: No. There is not a large amount of food grown in the vicinity of the station. The calculation was performed for a critical group of
individuals living at the boundary of the station and growing some vegetables and fruit themselves. Consumption data were taken from ICRP-23.

H.W. PATTERTON: What fraction of the tritium produced is being released?

D.J. GORMAN: The production rate is about 2400 Ci/MW(e) yearly. The release rate is about 24 Ci/MW(e) yearly, so about 1% of the tritium produced is released.

T.R. THOMAS: I have three questions to ask you.
(1) Does the $1 \times 10^7$ Ci/a atmospheric release limit correspond to the 500 mrem/a per person allowed in Canada?
(2) Is this the permissible release limit for a single point source, or for the cumulative sources in Ontario?
(3) Do you expect the release limit for $^3$H to the atmosphere to be lowered in the future?

D.J. GORMAN: I will answer your questions in order.
(1) The $1 \times 10^7$ Ci/a gaseous Derived Release Limit (DRL) corresponds to a dose of 500 mrem/a (from inhalation and absorption through the skin) to an individual continuously resident at the boundary of the site (a distance of 1 km).
(2) The 500 mrem/a is an absolute dose limit to an individual. The DRL is based on the dose to the critical group living at the site boundary from tritium releases by the station. Combined releases accruing from several radionuclides are taken into account, with a further limiting condition that the sum of the actual releases divided by their respective DRLs must not exceed 1.
(3) There are two conditions under which the release limit might change:
   (a) If the dose limit to members of the public of 500 mrem/a were to be altered. This is the responsibility of the Atomic Energy Control Board and I cannot comment on it;
   (b) If an assessment of the environmental behaviour of tritium were to lead to the identification of another pathway to man or a re-evaluation of the significance of an existing pathway. The DRL would then probably be changed in order to reflect this.

Perhaps Mr. Chatterjee would like to comment further on this last question.

R.M. CHATTERJEE: The derived release limits given by Mr. Gorman represent secondary standards. They are the upper limits set by the Atomic Energy Control Board and are based on the dose limits for individuals at the boundary of a station (500 mrem/a). The utilities operate at a much lower level and are encouraged to do so. There is, however, no official move as far as I know to lower these secondary standards as long as the utilities operate at the 'as low as reasonably achievable' levels.

E.L. COMPERE: When you speak of tritium production in terms of curies per megawatt/year, do you mean MW(th) or MW(e) and is the year the 365-day year or the operating year?

D.J. GORMAN: The MW referred to are gross MW(e) and the year is an operating year.
TRITIUM MONITORING AT THE SANDIA TRITIUM RESEARCH LABORATORY*

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Abstract

TRITIUM MONITORING AT THE SANDIA TRITIUM RESEARCH LABORATORY.

Sandia Laboratories at Livermore, California, is at present beginning operation of a Tritium Research Laboratory (TRL). The laboratory incorporates containment and clean-up facilities such that any unscheduled tritium release is captured rather than vented to the atmosphere. A sophisticated tritium monitoring system is in use at the TRL to protect operating personnel and the environment, as well as ensure the safe and effective operation of the TRL decontamination systems. Each monitoring system has, in addition to a local display, a display in a centralized control room which, when coupled with the TRL control computer, automatically provides an immediate assessment of the status of the entire facility. The computer controls a complex alarm array and integrates and records all operational and unscheduled tritium releases.

THE TRITIUM RESEARCH LABORATORY

Sandia Laboratories, Livermore, is at present beginning operation of a Tritium Research Laboratory (TRL) designed for experimental work with isotopes of hydrogen and their compounds. A diverse range of experiments is possible involving gram amounts of tritium. The design basis of the facility incorporates the U.S. Department of Energy recommendation of limiting release to the environment to 'as low as reasonably achievable', which is implemented through containment and recovery. This is a major departure from the established practice of using high-velocity air hoods exhausted through stacks to the environment in order to protect operating personnel.

The TRL (Fig. 1) houses offices, laboratories, and containment and decontamination systems. The facility consists essentially of a central corridor with adjacent laboratories (Figure 2). Personnel protection is assured by a pressure-gradient-controlled once-through ventilation system which exhausts to a 30-m

* Work sponsored by the U.S. Department of Energy.
FIG. 2. TRL floor plan.
FIG. 3. TRL containment concept.

stack. The TRL containment concept is illustrated in Fig. 3. To contain any unintentional release as well as operational leakage (due to diffusion and venting), all experiments are conducted in sealed glove boxes.

The glove boxes are constructed of welded stainless steel and are equipped with 34 glove ports, 14 viewing windows and an air lock pass through (Fig. 4). Normally, the glove box pressure is maintained by the box pressure control system at $-0.25$ to $-1.0$ kPa with respect to the room with dry nitrogen or argon. A cooling system removes heat generated by experiments. Both tritium concentration and humidity control are maintained by connection with the decontamination system.

Decontamination of the glove box atmosphere is accomplished by two centralized systems [1], the Gas Purification System (GPS) and the Vacuum Effluent Recovery System (VERs). The systems were designed to be capable of reducing tritium concentrations to the low parts-per-billion level. Performance tests conducted on the systems show tritium removal with a concentration
FIG. 4. Sealed glove box.

FIG. 5. GPS schematic.
reduction factor (ratio of inlet to exhaust concentration) much in excess of 1000 per pass at an inlet concentration of 1 ppm (2.6 Ci/m³ for T₂) or less for both tritium and tritiated methane [2, 3].

The GPS (Fig. 5) protects against either an accidental release or a slow buildup of background tritium concentrations. The 340-m³/h flow-rate system consists of a central manifold connected to each of the laboratory glove boxes, a catalytic reactor to oxidize the tritium, two molecular sieve driers in series to collect the tritiated water, a blower to circulate the glove box atmosphere through the system, and a control and diagnostics system. The normal recirculation mode moves glove box gases through the GPS and back to the box until the contamination is reduced to an acceptable level.

The VERS (Fig. 6) decontaminates exhaust gases from the laboratory vacuum system before venting to the stack. The 17-m³/h flow-rate system consists of a laboratory vacuum manifold and two holding tanks to collect the effluent, a catalytic reactor to oxidize the tritium, the molecular sieve driers to collect the tritiated water, the necessary pumps to evacuate the laboratory manifold and transfer the effluent through the system, and a control and diagnostic system.

THE TRL MONITORING SYSTEM

The TRL monitoring system is designed to protect operating personnel and the environment, as well as to ensure the safe and effective operation of the decontamination systems. Tritium air concentrations are monitored in the room air, glove boxes, stack, GPS and the VERS with the use of Betatec tritium monitors. (All liquid wastes from the facility, collected in one of two 2500-gallon hold tanks, are monitored by standard liquid scintillation techniques before
FIG. 7. TRL monitoring system schematic.

FIG. 8. Dual 2-litre and 20-cm³ ion chambers.
FIG. 9. A meter module.

FIG. 10. A remote display module.
release to the sanitary waste system.) Each tritium monitor is interfaced with a central control computer, which continuously scans tritium concentrations, assesses the hazards based on the monitor readings, and automatically takes the appropriate action according to the hazard involved. Monitoring capability ranges from $\mu$Ci/m$^3$ to kCi/m$^3$ levels.

**Room air monitoring**

The ambient air in each room of the TRL is constantly monitored for tritium with a range of 1 $\mu$Ci/m$^3$ to 20 Ci/m$^3$. Each room air monitoring system (Fig. 7) consists of remotely selectable multiple inlet lines, a metal bellows air mover, flow gauge, dual 2-litre ionization chambers (Fig. 8) with vibrating reed electrometer, and several digital displays. The five-decade autoranging digital displays are located in the rooms themselves (Fig. 9), in the central corridor at the doorways to each room (Fig. 10) and in the control room (Fig. 11). The meter module [4] provides direct digital readout of the tritium concentration and independent, fully adjustable low- and high-level audible/visual alarms. Console displays are similar to the meter module, but in addition are the interface between the tritium monitor and the TRL control computer, and provide the four sample mani-
FIG. 12. Schematic of the TRL stack tritium monitoring system.

FIG. 13. The stack effluent sample array.
fold valve controls and a fifth control for a backup tritium monitor located on the room-air calibration manifold.

A unique feature of the room-air monitoring system is the room-air sample manifold. This manifold allows localized monitoring in each room. Each manifold consists of a remotely selectable, four-way inlet system, and is fitted with an in-line particulate filter and normally open solenoid valve. The drop locations are selected on the basis of most likely release of tritium to the room, for example, at the working face of a glove box. In the normal mode of operation, all the valves are open such that the average room-air concentration is measured. Should a particular area of the room require monitoring, the valves can be selectively closed from the appropriate display module in the control room.

Glove box monitors

The tritium concentration of each glove box is continuously monitored with dual 20-cm³ ion chambers with a sensitivity of 1 mCi/m³ to a maximum of 20 kCi/m³. This range was selected to monitor glove box tritium loads of the order of 10 g. The glove box monitor provides a control function in that when a glove box tritium concentration exceeds a predetermined level, the box is connected to the GPS for decontamination. Decontamination of a box precludes tritium build-up in the room from permeation through gloves. A five-decade display is located in each glove box control compartment (attached to the box), as well as in the control room. The control room console display also controls a backup glove box monitor located on the glove box calibration line.

Stack effluent monitoring system

There are no scheduled releases of tritium to the 30-m stack. However, some low-level tritium is released in the laboratory areas and glove boxes, largely because of maintenance procedures. Tritium is released either as elemental gas or in chemical combinations such as tritiated water or methane. Ultimate operating-personnel protection is assured by the pressure-gradient-controlled once-through ventilation system exhausting to the stack.

Air exhausted from the facility is collected in a central duct and is drawn through an air equalizing and straightening section, past the stack tritium monitoring sampling array, through the exhaust fans, and up the stack (Figure 12). The sampling array features an expanded aluminium honeycomb straightening section (Figure 13) which reduces variances in the velocity profile of the air flow leaving the section. All air rotation (turbulence) is eliminated and uniform laminar air flow is delivered to the sensor sections. The air flow is thus fully processed, and directionalized air flow is delivered to the sensor sections for accurate measurement.
The sensor sections function on the principle of a Pitot tube. The system includes separate static and total pressure sensing and averaging manifolds, and the stack tritium monitor sampling array.

The stack monitor consists of overlapping low-level and high-level monitors, and is capable of monitoring the real time concentration in the stack between 1 $\mu$Ci/m$^3$ and 2 kCi/m$^3$. In addition to the multipoint sampling array in the ventilation exhaust duct, the system consists of a dual 2-litre and a single 2-litre ion chamber with electrometer, the pumping station, and meter modules for both monitors located in the control room (Fig. 14). Each module interfaces directly with the control computer. Each module output also drives a totalizer.
The totalizer integrates the total amount of tritium released to the stack during a preset integration interval. An accumulated readout is provided which is the total tritium released to the stack since the totalizer was last reset to zero. At the end of each integration interval, the reading and time of day is printed. The totalizer is provided with a low- and a high-level alarm, which are activated if the tritium totals exceed their respective preset levels during the integration interval. The stack concentration is also integrated by the control computer, which samples and stores the stack tritium concentration about every ten seconds.

Under normal operating conditions, HT, HTO and tritiated hydrocarbons (e.g. from the vacuum pump exhaust) can be expected in the stack gases. A bubbler system is being installed and tested to distinguish between these species. Preliminary data indicate a sensitivity for HTO at the nCi/m³ level.

FIG.15. GPS control panel.
Initial testing

Although the individual components of the stack monitoring system had been tested before assembly, the complete system had not been tested with tritium. Therefore general tests were performed involving releases of curie amounts of tritium to the ventilation system [5]. Known amounts of tritium were released under continuous and instantaneous release conditions. Both the real time stack concentration and the totalizer readings agreed with the calculated values within 5%, assuring reliability of the stack monitoring system.
Decontamination systems monitors

The GPS, through which the glove box atmospheres are circulated, has inlet, interval and exhaust tritium monitors with sensitivities from 1 $\mu$Ci/m$^3$ to 20 kCi/m$^3$, depending on the desired level of each. For example, the inlet monitor is identical with a glove box monitor, with a range from 1 $\mu$Ci/m$^3$ to 20 kCi/m$^3$. This monitor also drives a totalizer which records the total amount of tritium processed by the GPS.

The VERS, which collects the effluent from all the vacuum systems in the TRL, has internal and exhaust monitors with sensitivities from 10 $\mu$Ci/m$^3$ to 200 Ci/m$^3$. Display modules for the VERS, as well as the GPS, are located at the entrance to the systems' equipment room and in the control room at the appropriate control station (Figs 15 and 16).
AUTOMATED TRITIUM MONITORING

The TRL control computer is a Digital Equipment Corporation PDP 11/40 computer (Fig. 17). The tritium concentration is automatically read and recorded for each tritium monitor location. It scans containment and decontamination equipment control systems for any malfunctions. The scan rate is of the order of one scan every ten seconds. It also generates control functions. For example, when a high glove box tritium level exists, the computer selects the proper operating parameters for the GPS and connects the box to the GPS until the tritium level has been sufficiently reduced.

As previously mentioned, all TRL tritium monitors are equipped with distinguishable low- and high-level audible alarms with selectable set points. Should a tritium level exceed a specified threshold for a monitor, in addition to local alarms, an alarm function is generated by the computer. This computer-generated alarm is independent of meter module alarm, and is typically set some 10 to 20% lower in order to alert control room personnel to the impending alarm condition.

Whenever the computer detects an alarm condition, it activates the guard alarm interface. A large number of alarm functions are multiplexed into the interface unit. An alarm may be generated by the computer or any of several TRL systems such as the GPS, VERS, any tritium-monitor, ventilation system, compressed air, chilled water or the electrical power distribution system. The interface unit generates one of four alarm functions based on the severity of the malfunction. In order of increasing importance, these are: operational, hazard, danger and emergency. An audible alarm is activated and the appropriate area of malfunction visually indicated. During non-operational hours, the alarm is relayed to Security Headquarters indicating which of the four alarm functions has been activated so that the appropriate personnel can be notified. The emergency alarm is relayed to Security Headquarters at all times.

REFERENCES


DISCUSSION

G.R. NEWBERY: Could you explain your reasons for dropping high-flow containment in favour of depression containment with glove boxes? It is our experience at Amersham that the latter are much less efficient in operational use, i.e. they lengthen the time taken by the operators and increase the risk of occupational exposure.

T.K. DEVLIN: This fully contained concept was used in response to the U.S. Department of Energy (DOE) requirement that exposures and releases be limited to 'as low as reasonably achievable' (ALARA). I agree that more time and expense may be involved in performing a given function in a glove box rather than a high velocity air hood. However, this is the basic design philosophy, and it does stress the limitation of worker exposure, which is of prime importance.

G.R. NEWBERY: How far have you considered containment of the tritium at source, i.e. by local containment and suitable absorbers?

T.K. DEVLIN: All experiments are conducted with high reliability apparatus. Certified hydrogen-compatible materials and construction practices are required. With larger quantities of tritium secondary containment is employed. The glove boxes are typically the tertiary containment vessel.

G.R. NEWBERY: Did you consider high-flow containment using water scrubbers to remove tritium (HTO)?

T.K. DEVLIN: The glove box gas purification system operates at 340 m³/h. Using a once-through full-time system would require much more extensive equipment and the cost would be prohibitive.

G.R. NEWBERY: Would you please describe your HT/HTO monitor?

T.K. DEVLIN: The HT/HTO monitor samples the stack gas via a series of 3 glycol bubblers to remove HTO, via a heated precious-metal catalyst (operating at 500°C) to oxidize the HT, and via another series of three glycol bubblers. The bubbler liquid is assayed with liquid scintillator techniques.

P.J.C. DINNER: Can you indicate the approximate cost of the monitoring system you described, and the number of monitoring points in the work area served?

T.K. DEVLIN: There are 30 tritium monitors in the facility, each costing about $3000. The decontamination systems cost about $0.5 million. The tritium monitors are disposed approximately as follows:

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Room air</td>
<td>12</td>
</tr>
<tr>
<td>Stack</td>
<td>2</td>
</tr>
<tr>
<td>Glove boxes</td>
<td>12</td>
</tr>
<tr>
<td>Decontamination system</td>
<td>4</td>
</tr>
</tbody>
</table>
P.J.C. DINNER: Secondly, could you say whether you have done any cost-benefit analysis for your tritium emissions control systems?

T.K. DEVLIN: The facility — which has cost $4–6 million in all so far — represents an attempt at full compliance with the D.O.E.'s 'ALARA' principle, and as such is a research effort rather than a production facility. Whether it significantly reduces exposure to workers or the environment will be evaluated after several years of operation.

P.F. SAUERMANN: What is the average tritium inventory in your laboratory?

T.K. DEVLIN: It was designed to use 100 g of tritium. During the initial start-up and testing phases, gram quantities are being used.

I. HANDYSIDE: You removed tritium from gaseous waste and so you presumably now have a solid waste material to deal with. Could you tell me what happens to it?

T.K. DEVLIN: As a matter of fact it's water. The HT is oxidized to HTO and collected on molecular sieve dryers. The dryers are regenerated as needed (2–4 times per year) and the tritiated water is collected in small containers of absorber or molecular sieve. If amounts are small (less than 1 g), the containers are sent for land burial. Large amounts will be recovered.

I. HANDYSIDE: So your intention, therefore, is to recycle the tritium rather than to dispose of it?

T.K. DEVLIN: Present plans are to ship the containers with the HTO absorbed on molecular sieve to another D.O.E. facility for recovery and re-use. So far the amounts originating from the Tritium Research Laboratory have not been large enough for this approach to be feasible.

E.L. COMPARE: Could you tell us something more about the permeability of window and glove materials to tritium and tritiated water?

T.K. DEVLIN: In general, tritium permeation is dependent on many variables such as temperature, moisture content, pressure differences, aging, etc., and as such is the subject of an intensive study. The permeation of tritium (primarily HTO) through glove materials and elastomer window seals is a major problem with sealed glove boxes containing high concentrations of tritium. For this reason, concentrations in the glove boxes are kept as low as practicable (of the order of 1 ppm), and the glove ports are covered when not in use and evacuated to the vacuum effluent recovery system to remove permeated tritium vapour.

E. SBERLO: Can you say something about monitoring contaminated surfaces outside the glove boxes?

T.K. DEVLIN: Contaminated surfaces are 'swiped' with dry filter paper circles and counted in a liquid scintillator. Every effort has been made to keep contamination as low as possible. The facility health physicist determines the action required for each contaminated surface, but general guidelines are as follows:
< 1000 dis/min per 100 cm²: unlimited release
< 10 k dis/min per 100 cm²: onsite release
10 k-1M dis/min per 100 cm²: controlled use — classified as 'contaminated'
> 1M dis/min per 100 cm²: classified as 'highly contaminated'

Measurable off-gassing may occur at removable surface contamination levels > 10 k/100 cm². In this case the rate is monitored with a portable monitor and is treated as highly contaminated. No direct-reading surface monitoring is planned at present.

E. WILLIAMSON: In a plant obviously designed to release negligible quantities of tritium to the stack, you quote for your stack monitoring system an efficiency such that the measured values of stack concentration are within 5% of those calculated.

Will you please say a little more about the tests conducted on the system?

T.K. DEVLIN: Known amounts of tritium were released to the stack in two modes — continuous and instantaneous. 1 to 5 Ci of tritium were released at a time. Knowing the amount released or the release rate and stack flow we could calculate the expected concentrations. This was compared with actual readings at the release point and in the stack. The testing is described in greater detail in Ref. [5] of the paper.
TRANSPORT THROUGH A TRENCH COVER OF GASEOUS TRITIATED COMPOUNDS FROM BURIED RADIOACTIVE WASTES*

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Abstract

TRANSPORT THROUGH A TRENCH COVER OF GASEOUS TRITIATED COMPOUNDS FROM BURIED RADIOACTIVE WASTES.

The wastes in commercial radioactive-waste land burial sites produce anaerobic decomposition products such as CH₄, CO, CO₂, NH₃ and H₂S which permit migration of ³H and ¹⁴C to the atmosphere. In the low-level radioactive-waste trenches at West Valley, New York, the most abundant radioactive gas was ³HCH₃ (as high as 2 nCi/cm² in some samples). A new empirical method has been developed to determine the bulk permeability of the trench cover and to estimate rates of gas production and gas release. The pressure differential between trench gas and atmosphere is measured with a sensitive, electrically operated transducer. At West Valley the trench covers were found to be highly permeable to pressure-induced flow of gases and air. Interconnected fractures apparently penetrate through the trench cap and determine the rate of flow of ³HCH₃ and other radioactive gases from the trenches. The pressure of gas in the trench has been reproduced with an empirical model, GASFLO, using pressure differentials measured in the field and parameters chosen to obtain the best fit to the experimentally observed pressures. The amount of ³HCH₃ released to the environment was estimated at approximately 1 to 8 Ci annually from various trenches.

INTRODUCTION

Low-level radioactive wastes are buried at the Western New York Nuclear Services Center, West Valley, New York, in 12 trenches approximately 180-m long, 10-m wide, and 6-m deep in a silty clay bed. Each trench is covered with 2 to 5 m of the excavated till. The water level in each trench is monitored by a sump pipe passing through the cover and waste to the bottom of the trench. In addition, as part of an ongoing study to determine radionuclide mobility in the trenches [1],

* Partly funded by USEPA, Contract 68-01-354.
well points were driven through the cover into the wastes at various depths within several of the trenches.

Trench-gas samples collected through the well points contain copious quantities of gaseous radioactive compounds, primarily $^3\text{HCH}_3$, $^3\text{HCO}_2$, $^3\text{HH}_2$ and other tritiated hydrocarbons, apparently formed by anaerobic decomposition of the buried wastes. Other gaseous radionuclides in the trenches include $^{85}\text{Kr}$, $^{222}\text{Rn}$, $^{14}\text{CO}_2$, $^{14}\text{CH}_4$ and $^{14}\text{C}$-hydrocarbons. These early Radiological Sciences Laboratory (RSL) field studies provided the first evidence that radioactive gases are produced in the burial trenches, and that $^3\text{HCH}_3$ and other radioactive gases are escaping to the environment [1].

Experiments are being conducted to determine the bulk permeability of the trench cover, and to estimate the amount of gas produced in the trenches. These experiments provided data for the calibration of a generic model, GASFLO, which calculates the flow of gas through the trench cover. The model also provides for incidental calculations of soil permeability, void volume and CH$_4$ production rates. This incidental information is expected to help define the mechanisms which lead to breakdown of the trench cover, permitting water to intrude into the trenches [2].

EXPERIMENTS

The first field experiment, conducted during 24–28 Oct. 1977, was designed to prove the concept that the trenchwide permeability of the trench cover could be evaluated. Water was being pumped out of burial trenches 3, 4 and 5 by the site operator in order to reduce the level of water standing in the trenches. All openings in each trench were closed to the atmosphere, and the gas pressure in each trench was monitored with an electrically operated pressure-differential transducer inserted into one of the well points. The associated measuring devices included a generator, constant-voltage transformer, power supply and digital multimeter. The transducers are specified by the manufacturer to have a pressure range of 7.5 mbar with an accuracy of 0.25% and repeatability of ±0.02% of the range; the temperature effect is ≈ 0.009%/°C. Atmospheric pressure was independently monitored with a barograph.

Additional field measurements were made for trenches 3, 5, 8 and 11 in February 1978, when the trench caps were frozen and covered by several centimetres of snow. All external vents, such as sump pipes and well points, were again sealed to the atmosphere. Instead of pumping water out of the trenches, gas was pumped out (or air pumped in) through a well point or sump pipe. The pressure differential between the atmosphere and a trench was monitored at a different well point. The pumping rate was measured with a gas flow meter.

A third set of field measurements was made on trenches 5, 8, 9, 10, 11 and 12 in April 1978 to identify possible seasonal variations of the bulk permeability.
A brief series of measurements were performed during July 1978 on trenches 3 and 5 just before compaction of the trench covers and the addition of another 1 to 2 m of covering material. Trenches 8, 11 and 12 were also tested.

Trench-gas samples were also collected for mass-spectrometric and radioc- hemical analyses to obtain the molecular and radionuclide concentrations used to estimate release rates. The analytical methods have been described previously [3, 4].

MATHEMATICAL MODEL

The behaviour of gas in a trench is influenced by changing atmospheric pressure and by pumping of water or gas out of the trench. If all external vents, such as sump pipes or well points, are sealed, the removal of a known volume of water (or gas) provides a way to measure indirectly the sum of the air flowing into the trench and gas production in the trench.

The cap permeability coefficient, $K_t$, can be defined by the Darcy-flow equation [5]

$$Q = -\frac{K}{\mu} \frac{A}{L} \Delta p = -K_t \Delta p = -K_t (p - p_a)$$  \hspace{1cm} (1)

where

- $Q$ = volume flow rate (cm$^3$/s)
- $\Delta p$ = pressure differential between trench gas and atmosphere (mbar)
- $K$ = permeability
- $\mu$ = viscosity coefficient
- $A$ = effective cross-sectional area
- $L$ = effective thickness of the cover
- $p$ = trench-gas pressure
- $p_a$ = atmospheric pressure

The parameter $K_t$ is determined rather than $K$. The pressure differential, $\Delta p$, is monitored at a well point different from that used for pumping.

Equation (1), which is similar in form to that for steady water flow, applies only when $\Delta p$ is small compared with atmospheric pressure, thus limiting the effects of gas compressibility.

When the pump is on and the atmospheric pressure is constant, the observed trench-gas pressure decreases. As the atmospheric pressure becomes greater than the trench-gas pressure, atmospheric air is drawn into the trench through the fractures in the cap. The trench-gas pressure continues to drop until the rate at
which water (or gas) is being removed is equal to the rate at which gas is supplied by the influx of air and by gas being produced anaerobically in the trench. At equilibrium, the rate at which gas enters the trench, $Q$ in Eq.(1), is equal to the net rate of pumping and gas production:

$$Q = -(Q_p + Q_V) = -K_1(\Delta p)_1$$

(2)

where

- $Q_p$ = volumetric gas production rate
- $Q_V$ = volumetric gas pumping rate
- $(\Delta p)_1$ = observed pressure differential at equilibrium

Numerical values for $Q_V$ are entered for calculation as positive values when air is pumped into the trench-gas reservoir and as negative values when gas is pumped out of the reservoir. Thus $Q$ also becomes negative for net outward flow of gas and positive for inward flow of air. To account for gas production from decay of wastes, $Q_p$ is always positive, but flow direction is defined by the negative sign in Eq.(2).

When the pump is turned off at constant atmospheric pressure, a new equilibrium pressure is established, and the rate at which the gas leaves the trench is equal to the rate of gas production:

$$Q = -Q_p = -K_1(\Delta p)_2$$

(3)

If equilibrium can be established, $Q_p$ and $K_1$ can thus be solved simultaneously from Eqs (2) and (3).

Equilibrium may not be reached during the period of observation, however, due to variations in barometric pressure or from inability of RSL staff to control pumping (as in the October 1977 experiments described in Figs 1 and 2). Therefore a mathematical model has been developed to compute trench-gas behaviour and to determine total flow rates under non-equilibrium conditions. The model is chosen to represent a system consisting of two permeable reservoirs — the atmosphere and the void volume in the trench — separated by a trench cover through which the gas flows linearly through a network of fractures. As trench gas is pumped out, the following expression will satisfy the mass balance in the trench:

$$m_p + m_V + m = \frac{\partial}{\partial t} (V_B)$$

(4)
where

\[ m_p = \text{mass production rate, } \rho Q_p \text{ (g/s)} \]
\[ m_Y = \text{mass pumping rate, } \rho Q_Y \]
\[ m = \rho Q, \text{ with } Q \text{ defined in Eq. (1)} \]
\[ t = \text{time} \]
\[ V = \text{void volume of the trench} \]
\[ \rho = \text{density of gas in trench} \]

We also use the ideal gas law to describe the trench-gas pressure:

\[ p = \rho RT \quad (5) \]

where

\[ R = \text{the gas constant} \]
\[ T = \text{temperature (°K), assumed constant} \]

Equations (1), (4) and (5) may now be combined (with appropriate differentiation) to obtain an equation in the form

\[ \frac{V}{p} \frac{dp}{dt} = -K_1(p - p_a) + Q_Y + Q_p \quad (6) \]

When trench water is being removed by pumping, the term \( Q_Y \) in Eq. (6) is replaced by \( -(\Delta V/\Delta t) \).

Equation (6) can be solved by any numerical method using values obtained from field measurements and parameters chosen to obtain a best fit to the observed behaviour. The Runge-Kutta numerical method, with time interval \( \Delta t = 6 \text{ s} \), was used for the calculations described here. Values for \( p_a \) and \( Q_Y \) are interpolated at 1-min intervals from the experimental barograph and flow-meter measurements. Once \( K_1 \), \( Q_p \) and \( V \) are calibrated and the initial trench-gas pressure is measured, the natural pumping action of the atmosphere is reproduced by the model using the interpolated values for \( p_a \) as input variables.

RESULTS OF EXPERIMENTS

Experiments in October 1977

The pressure differentials observed in October 1977 between the atmosphere and trenches 3 and 5 have been converted to pressure levels, which are plotted with the corresponding barographic readings in Figs 1 and 2.
FIG. 1. Pressure changes from trench 3 and atmosphere on 24 Oct. 1977 (---, atmosphere; ---, trench gas; --, GASFLO calculation).

FIG. 2. Pressure changes from trench 5 and atmosphere on 26 Oct. 1977 (---, atmosphere; ---, trench gas; --, GASFLO calculation).
Well point 3–1B is 60 m south of the sump pipe from which trench water was removed at 820 cm$^3$/s. During the first 37-min period when water was pumped out (beginning at 11.08 h), the pressure within the trench dropped from 0.15 mbar above atmospheric pressure to 0.50 mbar less than atmospheric. When the sump was turned off (at 11.45 h), the pressure in the trench gradually increased, and a positive differential of 0.20 mbar developed from the combined effects of decreasing barometric pressure and gas production within the trench. Subsequent pumping led to near-equilibrium conditions (at approximately 15.40 h, before the pump was shut off) with a resulting pressure differential of ~0.58 mbar.

In trench 5 water was pumped at the same rate from a sump pipe 60 m north of well point 5–2C and 120 m north of well point 5–3D, the two points used for pressure measurements. Similar pressure differentials were observed at the two well points, 60 m apart, demonstrating the high degree of permeability of the wastes inside trench 5. The trench-gas pressure behaved much like that in trench 3, but the trench cover exhibited a greater bulk permeability. The pressure differential at equilibrium was ~0.40 mbar, i.e. a smaller pressure gradient was needed to drive an equal amount of air into trench 5 than into trench 3.

In trench 4 there was no measurable pressure drop in well point 4-1B, 130 m from the sump pipe which was being pumped. An impermeable barrier apparently divides the reservoirs. Further evaluation would require installation of more well points into the trench.

**Experiment in February 1978**

The February 1978 experiment required pumping gas (rather than water) out of the trenches or pumping air into them. In trench 5 gas was removed from well point 5-3D at 802 cm$^3$/s. The pressure differential at equilibrium at well point 5-2C was 0.86 mbar (Fig.3), more than double that measured in October 1977. When the pump was turned off at 11.32 h, the pressure in the trench tended to equilibrate with the atmosphere, even though the trench cap was frozen and covered by several centimetres of snow. Later (at 15.31 h) air was pumped into well point 5-3D at 717 cm$^3$/s. While atmospheric pressure was increasing by 0.62 mbar, the pressure in the trench increased from approximately 0.55 mbar below atmospheric pressure to 0.20 mbar above it during the 51 min of pumping.

In trench 11 the pressure at well point 11-2A was approximately 0.62 mbar above the atmosphere when monitoring began at 13.40 h. (Fig.4). This high positive pressure is likely to have resulted from the relatively impermeable cap and the declining barometric pressure. Although the pressure in the trench decreased, the atmospheric pressure dropped more rapidly, and the pressure differential increased somewhat. At 14.17 h air was pumped into the trench at 720 cm$^3$/s through a sump pipe 50 m south of the well point. The pressure in the trench immediately increased, apparently because the volume of air pumped in exceeded
FIG. 3. Pressure changes from trench 5 and atmosphere on 7 Feb. 1978 (——, atmosphere; ···, trench gas; ---, GASFLO calculation).

the volume of gas vented through the trench cover. The rate of increase gradually slowed, but the pressure differential had reached 1.57 mbar at the time the pump was turned off (15.47 h).

A transducer was then inserted into the sump pipe. Identical pressure variations were observed within the well point and the sump pipe, demonstrating the high degree of permeability of the waste buried in trench 11. A sealed pipe with a cross-sectional area of 1.27 cm² mounted over a sanitary seal in the sump pipe was opened twice (at 16.36 and 16.52 h) to test its effect on the gas flow rate (Fig.4). Each time the pipe was opened, the flow of gas out of the trench was triple the rate occurring when the pipe was closed. Though resistance to flow in an open pipe is much less than that in fractures, the comparison shows that the effective cross-sectional area of the fracture network is small (apparently less than 1.27 cm² over a trench area of approximately 1750 m²).

Pressure measurements were also obtained for trench 8. Trench-gas behaviour for trench 8 was similar to that for trench 11, though the pressure differential at equilibrium was only about 0.9 mbar.

In trench 3 gas pressure was monitored at well point 3-1B. Pumping on the sump pipe did not produce a pressure differential at the well point, indicating a barrier to gas flow between the sump pipe and the well point. Pressure differentials created by changing atmospheric pressure were measured. The permeability of the cap on trench 3 was substantially less than in October 1977 (Table I). Whether or not the low permeability is an artifact of the change in experimental method remains a matter of conjecture at this time, however.

Experiment in April 1978

The influence of seasonal variations on bulk permeability in trenches 5, 8 and 11 was observed during April 1978. Trenches 9, 10, 12 and 14 were also tested. Pumping failed in trench 14 because the water level had risen above the perforations in the bottom of the pipe.

In trench 5 gas pressure was monitored at well point 5-2C while well point 5-3D was pumped at 665 cm³/s (Fig.5). Gas was pumped out beginning at 09.25 h; then the pump was turned off at 10.12 h to determine the rate of pressure recovery while atmospheric pressure remained constant. Air was pumped in at 774 cm³/s beginning at 11.02 h, and trench-gas pressure rose rapidly from 0.6 mbar below to 0.4 mbar above atmospheric pressure. When the pump was turned off at 11.41 h, the trench-gas pressure decreased at a very slow rate, indicating that the bulk permeability of the trench cap had substantially changed from that measured in February. After the 1.6-cm-diameter observation pipe was opened at 12.18 h, the pressure differential dropped to zero in about 11 min.

Data obtained for trenches 8–12 showed gas behaviour similar to that in trench 5 in response to pumping and changing atmospheric pressure, though the permeability varied from trench to trench.
TABLE I. VALUES OF CAP PERMEABILITY COEFFICIENT ($K_i$), VOLUMETRIC GAS PRODUCTION RATE ($Q_p$) AND VOID VOLUME OF THE TRENCH ($V$) OBTAINED IN FOUR EXPERIMENTS

<table>
<thead>
<tr>
<th>Trench</th>
<th>$Q_p$ (cm$^3$/s)</th>
<th>$V$ (cm$^3 \times 10^3$)</th>
<th>$K_i$ (cm$^3$/s per mbar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>47</td>
<td>1.9</td>
<td>1130$^a$</td>
</tr>
<tr>
<td>5</td>
<td>57</td>
<td>2.0</td>
<td>1960$^a$</td>
</tr>
<tr>
<td>8</td>
<td>15</td>
<td>1.6</td>
<td>1040</td>
</tr>
<tr>
<td>9</td>
<td>7</td>
<td>2.0</td>
<td>480</td>
</tr>
<tr>
<td>10</td>
<td>3</td>
<td>0.9</td>
<td>550</td>
</tr>
<tr>
<td>11</td>
<td>2</td>
<td>2.2</td>
<td>410</td>
</tr>
<tr>
<td>12</td>
<td>2</td>
<td>1.6</td>
<td>990</td>
</tr>
</tbody>
</table>

$^a$ Calculated from pressure differentials produced by pumping water from trenches.
$^b$ Calculated from pressure differentials produced only from atmospheric pressure changes; gas pumping was not possible.

FIG. 5. Pressure changes from trench 5 and atmosphere on 7 Apr. 1978 (—, atmosphere; •••, trench gas; ———, GASFLO calculation).
Experiment in July 1978

A brief series of measurements were performed during July 1978 in trenches 3 and 5 just before compaction of the trench covers. Ten other well points in trench 5 had been cut off below grade and filled with bentonite, then covered with excavated till. The bulk permeability of the cover on trench 5 was so great that a pressure differential of 0.1 mbar in well point 5-2C could only be observed when pumping on well point 5-3D while atmospheric pressure was constant. The extraordinary change in bulk permeability of the trench cap is mainly attributed to very dry weather conditions which had existed for more than 6 weeks.

The permeability of the cap on trench 3 was also dramatically changed compared with that in February. Pumping gas from, or air into, the sump pipe was again attempted, but without any more success than in February. Pressure differentials were therefore again measured from the natural pumping action created by changing atmospheric pressure. A sharp increase in permeability, similar to that for trench 5, was observed for trenches 8, 11 and 12, as well.

CALCULATIONS USING GASFLO MODEL

A Darcy-flow model, GASFLO, was developed to calculate bulk trench-cap permeability, void volume and gas production rate by fitting calculated pressure differentials to those observed.

The calculated gas pressure for trench 3 is compared in Fig.1 with values measured in October 1977. The best fit was obtained using \( V = 1.9 \times 10^6 \text{ cm}^3 \), \( K_i = 1130 \text{ cm}^3/\text{s per mbar} \) and \( Q_p = 47 \text{ cm}^3/\text{s} \). The pressure observed in the trench continued to decrease for a few minutes whenever pumping ceased, an indication of some impedance to gas flow in the void volume of the trench. The calculated curve responded immediately because the model assumes infinite permeability within the trench.

A similar comparison for trench 5 is shown in Fig.2. The best fit of experimental and calculated pressure was obtained when \( K_i = 1960 \text{ cm}^3/\text{s per mbar} \), \( Q_p = 57 \text{ cm}^3/\text{s} \), and \( V = 2.0 \times 10^6 \text{ cm}^3 \). The trench-gas pressure calculated with the model closely reproduced the observed values.

The void volume estimated by this model is the total volume formed by interconnected voids. Completely enclosed voids of any size are therefore excluded from the computation. The void volume might be altered for a given trench if the measurements are made at widely separated times. The best-fit value for void volume in trench 5 agrees well with the value of \( 2.1 \times 10^6 \text{ cm}^3 \) obtained by comparing water levels in the sump to the volume of water pumped from the trench since 1975. Unfortunately, the only other trench to overflow, trench 4, could not be tested by this method. Though this is the only model parameter

which can be tested experimentally and the data are very limited, we feel the excellent agreement lends some degree of confidence to the model.

The consistency of the model performance, regardless of whether air is pumped in or gas is pumped out, is illustrated in Fig. 3. The calculated pressure in trench 5 in February 1978 was obtained by using the same values for $V$ and $Q_p$ as in October 1977, but lowering $K_1$ to 880 cm$^3$/s per mbar. This value of $K_1$, about half of that obtained in October 1977, was used both for trench gas flowing out of the trench and for air flowing into it.

For trench 11 in February 1978 (Fig. 5) the best fit was obtained by using $V = 2.2 \times 10^6$, $K_1 = 410$ cm$^3$/s per mbar, and $Q_p = 2$ cm$^3$/s. When the sump was opened to the atmosphere, the rate at which air flowed out of the trench increased, according to model calculations, from 350 to 960 cm$^3$/s at $\Delta p \approx 0.73$ mbar and from 210 to 560 cm$^3$/s at $\Delta p \approx 0.44$ mbar.

The pressure differentials observed in trench 5 during April 1978 were reproduced with the GASFLO model by keeping $V$ and $Q_p$ constant, while adjusting $K_1$ to 280 cm$^3$/s per mbar (Fig. 5). The low value of $K_1$ may be caused by infiltration of the fractures with precipitation and with water from melting snow.

Similar calculations were made for trenches 3, 8, 9, 10, 11 and 12 (Table I). The values of $K_1$ for any particular trench decrease sequentially from October through April, as long as $Q_p$ and $V$ are kept constant.
TABLE II. EFFECT OF $K_1$ VALUES ON ESTIMATES OF TOTAL VOLUME OF GAS RELEASED IN SIX HOURS OF NATURAL PUMPING

<table>
<thead>
<tr>
<th>$K_1$ (cm$^3$/s per mbar)</th>
<th>Volume (cm$^3 \times 10^6$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2.0 \times 10^9$</td>
<td>0.06</td>
</tr>
<tr>
<td>$10^1$</td>
<td>0.57</td>
</tr>
<tr>
<td>$10^2$</td>
<td>3.65</td>
</tr>
<tr>
<td>$10^3$</td>
<td>6.62</td>
</tr>
<tr>
<td>$10^4$</td>
<td>8.86</td>
</tr>
</tbody>
</table>

The pressure differential of 0.1 mbar observed in trench 5 during July 1978 produced a $K_1$ value of 12000 cm$^3$/s per mbar, whereas those for trench 3 produced a $K_1$ value of 1400 cm$^3$/s per mbar (Table I). Calculations for trenches 8, 11 and 12 also produced exceptionally large $K_1$ values of 11000, 11800 and 11000 cm$^3$/s per mbar respectively.

ESTIMATES OF GASEOUS $^3$HCH$_3$ ESCAPING TO THE ATMOSPHERE

One method used to estimate the amount of $^3$HCH$_3$ released to the environment is illustrated in Fig.6. Barograph readings are plotted with the calculated gas pressure in trench 5 using the best-fit values for $V$, $Q_p$ and $K_1$ obtained in February. The flow rates generated from Eq.(1) using the trench-gas and barometric pressures result in an estimated volume of $6 \times 10^6$ cm$^3$ of gas released during 6 h of natural (atmospheric) pumping. Assuming that the atmosphere pumps gas out of the trench during half of the year, approximately $5 \times 10^9$ cm$^3$ of gas would be released annually. Concentrations of gaseous $^3$HCH$_3$ measured in samples collected from well point 5-2B have averaged approximately 0.8 nCi/cm$^3$, so that each year approximately 4 Ci of $^3$HCH$_3$ are released to the environment from trench 5.

Estimation of the volume of gas released to the environment depends on the values obtained for $K_1$, which can vary seasonally with climatic changes. The effect on total gas volume released of variations in $K_1$ for 6 h of atmospheric pumping is shown in Table II. The total volume of gas released is not linearly related to $K_1$ because $\Delta p$ is also a function of $K_1$. For example, it only triples when $K_1$ increases 100-fold from 200 to 20000 cm$^3$/s per mbar, values which are representative for all trenches. Thus the releases of $^3$HCH$_3$ projected from total gas flow can be considered reasonably independent of $K_1$. 
TABLE III. ESTIMATED ANNUAL RELEASES OF $^3$HCH$_3$ FROM BURIAL TRENCHES AT THE WESTERN NEW YORK NUCLEAR SERVICES CENTER

<table>
<thead>
<tr>
<th>Trench</th>
<th>Annual gas outflow (cm$^3 \times 10^6$)</th>
<th>$^3$HCH$_3$ concentration (nCi/cm$^3$)</th>
<th>$^3$HCH$_3$ release rate (Ci/a)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Trench gas</td>
<td>CH$_4$ fraction</td>
</tr>
<tr>
<td>3</td>
<td>4.2</td>
<td>0.3</td>
<td>1</td>
</tr>
<tr>
<td>5</td>
<td>4.4</td>
<td>0.8</td>
<td>3</td>
</tr>
<tr>
<td>8</td>
<td>3.3</td>
<td>0.8</td>
<td>10</td>
</tr>
<tr>
<td>9</td>
<td>3.7</td>
<td>0.3</td>
<td>20</td>
</tr>
<tr>
<td>10</td>
<td>1.9</td>
<td>1.2</td>
<td>90</td>
</tr>
<tr>
<td>11</td>
<td>3.2</td>
<td>0.7</td>
<td>120</td>
</tr>
<tr>
<td>12</td>
<td>3.0</td>
<td>0.2</td>
<td>30</td>
</tr>
</tbody>
</table>

$^a$ Using total gas flow.

$^b$ Annual CH$_4$ release rates were calculated from Q$_p$ values in Table I.

An alternate method for estimating $^3$HCH$_3$ release to the environment assumes that the calculated incidental production rate, 57 cm$^3$/s, is predominantly methane and is constant for the entire year. The volume of CH$_4$ released annually would then be approximately $2 \times 10^8$ cm$^3$. This method yields an estimate of 6 Ci of $^3$HCH$_3$ released annually from trench 5.

Estimates of $^3$HCH$_3$ released from trenches 3, 5 and 8–12 are provided in Table III by two methods: A, using total gas flow, and B, using the CH$_4$ production rates from Table I. Taking into consideration errors in averaging $^3$HCH$_3$ and CH$_4$ concentrations for a limited number of samples from each trench, broadly spaced in time and location, the release rates are considered accurate to within a factor of 2 or 3. Annual release rates calculated for trenches 9–12 by method B may have been overestimated, because values of Q$_p$ obtained for these trenches are small, and do not markedly affect the GASFLO calculations.

DISCUSSION

The trench covers are highly permeable to pressure-induced flow of gas and air. It appears that a network of fractures play a dominant role in the rate of flow of gases from the trenches. Though permeability can be measured as described above, the sizes of the fracture openings and the fracture porosities are not directly
observable, and may not be correlated with permeability. The permeability of the trench covers appears to be dramatically affected by climatic conditions.

The assumption of Darcy flow appears satisfactory for calculating trench-gas flow through the soil cover of low-level radioactive-waste burial trenches.

The GASFLO model appears to provide reasonable estimates of tritium release rates for burial trenches which have a relatively high degree of permeability, as where the trench cover consists of redistributed soil which had been excavated during construction. Since the covers for all commercial radioactive-waste burial sites in the United States of America are prepared from excavated material, we presume this model can be generally applied.

ACKNOWLEDGEMENTS

The authors express appreciation to those who contributed to the success of the field measurements, particularly C. Kunz, D. Prudic, S. Molello, W. Mahoney, T. Miller, C. Rem, M. Trier and C. Schwenker.

REFERENCES


DISCUSSION

P.J.C. DINNER: Would you please indicate the total quantity of tritium present in each trench, so that we can see the fractional release rate your values represent?

A.H. LU: The total amount of tritium now present in each of the trenches studied can be estimated from burial and pumping records only if we assume that
wastes recorded as 'Mixed and Miscellaneous' contained no tritium. Thus the approximate quantities of tritium in each trench as of 1978 would be: trench 3, 3000 Ci; trench 5, 8000 Ci; trench 8, 10000 Ci; trench 9, 12000 Ci; trench 10, 22000 Ci; trench 11, 27000 Ci; and trench 12, 5000 Ci. Since the 'Mixed and Miscellaneous' wastes represent up to 40% of the total waste in some trenches, the quantities of tritium actually buried might be greater than the estimates above, perhaps by 50% or more.
FIFTEEN YEARS OF EXPERIENCE IN HANDLING TRITIUM PROBLEMS IN CONNECTION WITH LOW-ENERGY PARTICLE ACCELERATORS

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Institute of Chemistry,
Nuclear Research Center Jülich, Jülich,
Federal Republic of Germany

Abstract

FIFTEEN YEARS OF EXPERIENCE IN HANDLING TRITIUM PROBLEMS IN CONNECTION WITH LOW-ENERGY PARTICLE ACCELERATORS.

The leakage of tritium to the environment in connection with transport, storage and installation of high-activity tritium targets was investigated. The release and distribution of tritium in the environment from target changing and during normal operation of the accelerator were also studied, as well as tritium hazards arising from the contamination of shipping containers, certain sections of the accelerator system, cooling water and pump oil. New procedures suitable for the conditioning of tritium such as those found in pump oils, scintillator solutions, cooling water, absorption cartridges and used targets, are discussed.

INTRODUCTION

Low-energy particle accelerators (neutron generators) are widely used for producing fast neutrons by the °H(d,n) °He-reaction. The activity of tritium targets for a neutron output of 10¹¹ s⁻¹ amounts to about 100 Ci, and, since this activity is partly released to the environment, this tritium source offers a favourable opportunity of studying the distribution and behaviour of tritium after its release to the environment.

On the other hand, each step of the tritium target cycle (Fig. 1) presents specific risks, mainly arising from tritium leaking into the environment and contamination in the working area. In particular, the loss of tritium from high-activity targets during transport and storage should be kept in mind, since leak rates of shipping containers and storage cases may be considerably high. The main radiation protection problems arising from the operation of neutron generators are tritium release and contamination upon changing the target,
FIG. 1. Tritium target cycle.

FIG. 2. Layout of neutron generator and auxiliary equipment.
contamination of cooling water and pump oil, and, last but not least, the handling and disposal of tritium-containing wastes.

In the Nuclear Research Center at Jülich, different types of neutron generators with open target systems or sealed tubes are in use, but most information has been obtained from operating the Dynagen 300 generator manufactured by Radiation Dynamics [1, 2]. In Fig. 2 the layout of this accelerator [1] and the auxiliary equipment is shown. The maximum ion beam current is about 5 μA and the maximum neutron output about $2 \times 10^{11}$ s$^{-1}$. The rotating tritium target (Fig. 3) is cooled by water in a closed primary cooling circuit and an open secondary cooling circuit, as shown in Fig. 2. The vacuum is produced and maintained by a diffusion pump and a mechanical pump, decontamination of the exhaust being achieved by means of an absorption trap consisting of a heated tube with CuO and molecular sieves [3].

The rotating target, manufactured by Nukem, is shown in Fig. 3 [4]. Its usual activity was about 80 Ci, almost half of which is lost by outgassing, dislodging and sputtering in the course of the tritium target cycle.

How to manage the hazards arising from this situation is described below.
FIG. 4. Shipping container for high-activity tritium targets.

FIG. 5. Tritium release from shipping container and target case. Target activity: 80 Cl.
TABLE I. TRITIUM RELEASE FROM SHIPPING CONTAINER AND TARGET CASE

*Target activity: 80 Ci*

<table>
<thead>
<tr>
<th></th>
<th>µCi/d</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
<td>2</td>
<td>6</td>
</tr>
<tr>
<td>HTO</td>
<td>17.3</td>
<td>1.0 \times 10^4</td>
<td>86%</td>
</tr>
<tr>
<td>HT, T₂</td>
<td>17.3</td>
<td>1.7 \times 10^3</td>
<td>14%</td>
</tr>
<tr>
<td>Total</td>
<td>17.3</td>
<td>1.2 \times 10^4</td>
<td></td>
</tr>
</tbody>
</table>

*FIG. 6. Shipping container for tritium target of high activity.*

RELEASE OF TRITIUM TO THE ENVIRONMENT

Release from shipping container

In Fig.4 the shipping container normally used for transporting tritium targets is shown. It is made of sheet-iron and sealed-off with rubber packing. The target case is supported by styropor parts and sealed by a rubber gasket.

The tritium leakage from this assembly was measured in the closed box shown as number 1 in Fig.5. In the steady state the release is 1.2 \times 10^{-2} \text{ µCi/min. Opening the container (number 2) results in an increase of leakage to 8.3 µCi/min and, finally, opening the target case (number 3) is followed by a release of 38 µCi/min. The results are summarized in Table 1. By application of the freezing-out method [5] we were able to show that about 85% of the released tritium was HTO.*

These results led to the development of an improved container (Fig.6). This is made of aluminium and sealed with indium packing. By means of the valve connectors, tritium can be removed before opening the container. In addition, the target case is filled with argon instead of air to prevent the generation of HTO.
FIG. 7. Release of tritium caused by target change. Target activity: 80 Ci.

FIG. 8. Tritium emission through two target periods.
Release of tritium upon target changing

Installation of a new target, which must be carried out two or three times yearly, requires opening of the vacuum system followed by the release of tritium to the environment. The amount of released tritium depends on the operation conditions before the target change. If the neutron generator has been out of operation for some time and the hand valve between target area and pumps closed, tritium release of up to 1 Ci may result, as shown in Fig.7, whereas only a few millicuries are released if the pumps have been recently in operation.
TABLE II. CORRELATION BETWEEN EMISSION AND ADMISSION AFTER RELEASE OF 1 Ci TRITIUM GAS UNDER DIFFERENT WEATHER CONDITIONS

<table>
<thead>
<tr>
<th>Experiment No.</th>
<th>Emission: maximum concentration (µCi/cm³)</th>
<th>Admission: maximum concentration (µCi/cm³)</th>
<th>Incorporation at admission point (µCi)</th>
<th>Distance (m)</th>
<th>Difference in altitude (m)</th>
<th>Temperature (°C)</th>
<th>Relative humidity (%)</th>
<th>Wind velocity (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$2.1 \times 10^8$</td>
<td>$3.4 \times 10^{-6}$</td>
<td>0.54</td>
<td>25</td>
<td>15</td>
<td>19</td>
<td>56</td>
<td>1.9</td>
</tr>
<tr>
<td>2a</td>
<td>$3.1 \times 10^{-3}$</td>
<td>$2.7 \times 10^{-5}$</td>
<td>6.2</td>
<td>25</td>
<td>6</td>
<td>16.8</td>
<td>48</td>
<td>3.0</td>
</tr>
<tr>
<td>2b</td>
<td>$3.1 \times 10^{-3}$</td>
<td>$7.5 \times 10^{-7}$</td>
<td>0.45</td>
<td>50</td>
<td>6</td>
<td>16.8</td>
<td>48</td>
<td>3.0</td>
</tr>
<tr>
<td>3a</td>
<td>$2.6 \times 10^{-3}$</td>
<td>$1.2 \times 10^{-6}$</td>
<td>0.69</td>
<td>25</td>
<td>6</td>
<td>11</td>
<td>55</td>
<td>7.0</td>
</tr>
<tr>
<td>3b</td>
<td>$2.6 \times 10^{-3}$</td>
<td>$1.2 \times 10^{-7}$</td>
<td>0.027</td>
<td>50</td>
<td>6</td>
<td>11</td>
<td>55</td>
<td>7.0</td>
</tr>
<tr>
<td>4</td>
<td>$2.9 \times 10^{-3}$</td>
<td>$7.6 \times 10^{-5}$</td>
<td>3.5</td>
<td>5</td>
<td>6</td>
<td>18</td>
<td>52</td>
<td>2.1</td>
</tr>
</tbody>
</table>
Release of tritium upon normal operation of the accelerator

In Fig. 8 tritium releases for a period of three quarters of a year are shown. On the average, about 9 Ci were released monthly. In this period we experienced the validity of the rule of thumb which states that half the activity of a fresh target is released to the environment during its life-time or buried in ion getter pumps or absorption traps.

In order to reduce drastically our tritium releases to the environment, we installed some years ago the absorption trap mentioned earlier. This measure resulted in a reduction in tritium release by a factor of $10^6$ under normal operating conditions, as shown in Fig. 9.

DISTRIBUTION OF TRITIUM IN THE ENVIRONMENT

Releases of curie amounts of tritium to the environment have the advantage that the correlation between the emission and admission in the proximity of buildings can be studied in order to evaluate the possible exposure of people in this area. Only a few such experiments have been performed so far showing the expected dependence on geometrical and weather conditions. They are summarized in Table II.

In addition, the distribution of the different tritium species as a function of operating time was studied [3, 5, 6]. Because of the different radiotoxicity of tritium gas and tritiated water the elucidation of this problem is of some importance. As stated earlier, the releases of fresh tritium targets consist mainly of tritiated water. This is not the case with tritium from the target installed in the vacuum system of the accelerator. At the beginning of the life-time of a target about 80% $T_2$ and 20% HT are found, as shown by radiogas-chromatic studies [3, 7]. With increasing operating time the percentage of DT rises, whereas the percentages of the other components decrease, as shown in Fig. 10.

The rate of transformation of HT and $T_2$ to HTO was studied by us some years ago [5]. We found out that this rate was about 0.3% daily. From this it follows that only a small fraction of tritium gas leaking from nuclear installations into the working environment is converted to tritiated water. Consequently, the higher radiation protection standards for tritium gas may be applied for setting limits for controlled areas. On the other hand, the tritium gas activity released into the general environment will be totally converted. Therefore, the radiation protection standards for tritiated water must be applied in this case.
FIG.10. Distribution of tritium species as function of operating time.

FIG.11. Results of smear tests taken inside a shipping container for high-activity tritium targets.

CONTAMINATION PROBLEMS

Contamination of shipping container and packing material

As shown in Fig.4 and Table 1, the predominant tritium species inside the target case and shipping container is HTO. Consequently, intense contamination of the inner parts of these vessels is to be expected. In Fig.11 the results of smear tests, ranging from 16 μCi to 55 mCi, are recorded. Sometimes a target certificate was included by the target manufacturer in the shipping container. The activity of the certificate was evaluated at three times the limit (~300 μCi) [8].
Contamination upon target changing

Opening the vacuum system to change the target results in air and surface contaminations. It can be concluded from Table 1 and Fig.10 that the extent of these contaminations depends on the operating conditions of target and accelerator shortly before target change.

Figure 12 shows the results of wipe tests taken on areas of about 100 cm² shortly after the installation of a new 80-Ci target (remaining activity of the used target: ∼40 Ci) [8].
Considerable contamination was observed since the operating conditions of container and accelerator had been such that high HTO-concentrations were in these vessels.

Taking into account our experiences, we changed the target-change procedure in so far as the shipping container is now vented through the absorption trap shown in Fig.2 before it is opened in the glove box shown in Fig.13. By these means we were able to prevent air and surface contaminations and to reduce drastically tritium effluence to the environment, as shown in Fig.7. Furthermore, tritium incorporation by workers could be reduced by a factor of 100.

**Contamination of cooling water**

As shown in Fig.2, the cooling system consists of a closed primary cycle and an open secondary cycle. It is apparent from Fig.14 that the primary cycle is increasingly contaminated as a function of operating time, the rise of contamination being stepwise. The steep slope of the steps (about 40 μCi/ltr) originates from contaminations of the back-side of the target disc and the flat slope is due to diffusion of tritium through the copper disc and, possibly, the sealing (see Fig.3). The rise in tritium concentration from diffusion is about 0.05 μCi/ltr per hour of operation. Taking into account the volume of 12000 ltr of the primary circuit, about 600 μCi of tritium are added by diffusion to this circuit hourly (about
TABLE III. SOLIDIFICATION OF TRITIUM-CONTAINING LIQUID WASTE

<table>
<thead>
<tr>
<th>Waste</th>
<th>Volume ratio gypsum/pumice (gp)</th>
<th>Volume ratio gypsum/waste (mix)</th>
<th>Volume ratio mix/water</th>
<th>Solidification time (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pump oil</td>
<td>1 : 1</td>
<td>3 : 1</td>
<td>No water addition</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>3 : 1</td>
<td></td>
<td>+ 10% H₂O</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>3 : 1</td>
<td></td>
<td>+ 20% H₂O</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>3 : 1</td>
<td></td>
<td>+ 50% H₂O</td>
<td>2</td>
</tr>
<tr>
<td>Liquid scintillator</td>
<td>1 : 1</td>
<td>3 : 1</td>
<td>No water addition</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>1 : 1</td>
<td>4 : 1</td>
<td>No water addition</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>1 : 1</td>
<td>2 : 1</td>
<td>No water addition</td>
<td>2</td>
</tr>
<tr>
<td>Cooling water</td>
<td>1 : 1</td>
<td>2 : 1</td>
<td>No water addition</td>
<td>0.1</td>
</tr>
</tbody>
</table>

30 mCi per target period of about 50 hours), whereas every target change contributes about 480 mCi. Thus, the cooling water contains about 5 Ci of tritium after consumption of ten 80-Ci targets.

WASTE HANDLING

Retention of tritium from the environment by measures as described above results in the production of different kinds of wastes. Firstly, high-activity waste
arises from oxidizing tritium gas in the copper oxide absorption trap; secondly, tritium-containing medium- and low-activity liquid wastes arise from the cooling circuit, vacuum and scintillation measuring equipment. Disposal of these wastes requires them to be conditioned, i.e. conversion into solid form.

In Fig. 15 the conditioning of tritium-loaded absorption cartridges is shown [3]. The molecular sieve tube contains about 100 g of molecular sieve, and its water-retaining capacity is about 20 g. Up to now, we have loaded the tubes with about 100 Ci tritium only, but the maximum capacity is about 20000 Ci. The tubes are, as shown in Fig. 15, embedded in gypsum or synthetic resin enclosed in an aluminium tube and closed by a metal-sealed flange.

This arrangement is not only suitable for final disposal but also appropriate for retrievable storage since HTO can be recovered by heating the molecular sieve tube. Tritium-containing liquid wastes often pose many problems although only small amounts of them are produced by operation of neutron generators.

Solidification of pump oils and scintillator solutions in our laboratory is performed by means of a method developed by us some years ago [9, 10]. This uses the solidification properties of gypsum and the absorbing capacity of pumice. Some ways of making use of this simple and inexpensive method are given in Table III.

Used tritium targets are returned to the manufacturer for reloading but, in principle, they could be prepared for final storage in a similar way to the absorption cartridges.

CONCLUSION

Neutron generators can be intensive tritium sources within the environment. If no measures are taken to prevent these tritium releases, about half the activity of tritium targets employed for neutron production is distributed in the environment. By using closed cycles and different retaining techniques we succeeded in reducing our yearly tritium releases from more than 100 Ci to a few millicuries. As a result we obtained tritium wastes conditioned according to the requirements for final storage.

Unfortunately, these wastes accumulate in our intermediate storage facility because the former salt mine, Asse II, which serves as the final disposal store for low- and medium-radioactive wastes in the Federal Republic of Germany, only accepts up to ten millicuries tritium in 200-ltr barrels.

REFERENCES

DISCUSSION

T.K. DEVLIN: Is the gypsum packing material or absorption medium solidified or does it remain a slurry?

P.F. SAUERMANN: As shown in Table III of the paper, used pump oils and liquid scintillator solutions are solidified without addition of further water, since these wastes already contain small amounts of water. With increasing addition of water the solidification time shortens whereas the compressive strength of the gypsum/pumice solids rises. But the value of this solidification method depends not only on these properties but also on the leaching behaviour of solidified samples. This was investigated by us in 1977. As was expected, the leaching behaviour of samples based on gypsum is far from good, the reason being that gypsum is not absolutely insoluble in water. Indeed, the leaching rates we obtained of about $10^{-2}$ cm·d⁻¹ are, on the average, of the same order of magnitude as the maximum solubility of gypsum in water.

The leaching behaviour may be improved substantially by suitable admixtures or coatings. Thus, paraffin coating of our samples resulted in a >10⁴-fold decrease in the activity release. The leaching rates are then of the order of $10^{-5}$ cm·d⁻¹ or less. Reported leaching rates for the bitumen method are in the region $10^{-4}$ to $10^{-5}$ and for the cement method $10^{-2}$ to $10^{-4}$, for uncoated samples (polymer impregnated concrete: $10^{-2}$ to $10^{-4}$).
Taking into account the other criteria for the applicability of a solidification method, such as cost, simplicity of handling, trouble from heating, gas evaporation, content of acids or organic components, it is obvious that the gypsum procedure presents surprisingly numerous advantages compared with other methods, and these advantages largely offset the poor leaching properties.

I. HANDYSIDE: Why have you gone to this trouble to solidify tritium wastes when you appear to have no disposal route for them?

P.F. SAUERMANN: You have touched upon a difficulty here. We will have to store these wastes for a long time, because the final storage facility for radioactive wastes, which is planned as part of the nuclear fuel reprocessing centre at Gorleben, Niedersachsen (FRG), will not be in use before the end of the century. Until then the tritium-containing wastes have to be held in our intermediate waste storage facility, but we are allowed to do this only if they are conditioned so as to meet the requirements for long-term storage. These wastes have consequently to be solidified.

The molecular sieve cartridges loaded with HTO originating from oxidized tritium gas can be conditioned in such a way that tritium can be recovered later if it is needed as fuel for thermonuclear reactors.

A.P. HULL: At Brookhaven National Laboratory, we have encountered a similar situation with a Van de Graaff accelerator which utilizes tritium targets. Most of our releases of tritium occur after the system has been opened up, i.e. when it is being pumped down to vacuum conditions again. Providing for decontamination by tritium retention at the much higher flow rates obtaining during pump-downs than during normal operation appears quite expensive. Is this a problem in your experience?

P.F. SAUERMANN: As shown in Fig. 7 of the paper, in our case the tritium release during target change could be reduced by a factor of 100 by changing the operating conditions of the accelerator before opening the vacuum system. The same procedure may also work with your Van de Graaff. The cupric-oxide-tritium scrubber used by us only operates at slow flow rates of about 50 ml/min.
<table>
<thead>
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<th>CHAIRMAN</th>
<th>COUNTRY</th>
</tr>
</thead>
<tbody>
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<td>1</td>
<td>D.G. JACOBS</td>
<td>United States of America</td>
</tr>
<tr>
<td>2</td>
<td>T. SHIOKAWA</td>
<td>Japan</td>
</tr>
<tr>
<td></td>
<td>W. FELDT</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>3</td>
<td>G. MASTINU</td>
<td>Italy</td>
</tr>
<tr>
<td></td>
<td>R.V. OSBORNE</td>
<td>Canada</td>
</tr>
<tr>
<td>4</td>
<td>R. KIRCHMANN</td>
<td>Belgium</td>
</tr>
<tr>
<td>5</td>
<td>A. YUTHAMANOP</td>
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<td>Netherlands</td>
</tr>
<tr>
<td>9</td>
<td>N.B. JUAN</td>
<td>Philippines</td>
</tr>
<tr>
<td></td>
<td>G.R. NEWBERY</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>10</td>
<td>M. MENDELSOHN</td>
<td>United States of America</td>
</tr>
</tbody>
</table>
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</tr>
<tr>
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<tr>
<td>Name</td>
<td>Address</td>
</tr>
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<td>-------------------------------------------------------------------------</td>
</tr>
<tr>
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</tr>
<tr>
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</tr>
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<td>Porter-Gertz Consultants, 76 Rittenhouse Place, Ardmore, PA 19003</td>
</tr>
<tr>
<td>Name</td>
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<tr>
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<tr>
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</tr>
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</tr>
<tr>
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The following conversion table is provided for the convenience of readers and to encourage the use of SI units.

**FACTORS FOR CONVERTING SOME OF THE MORE COMMON UNITS TO INTERNATIONAL SYSTEM OF UNITS (SI) EQUIVALENTS**

**NOTES:**
1. SI base units are the metre (m), kilogram (kg), second (s), amper (A), kelvin (K), candela (cd), and mole (mol).
2. ▶ indicates SI derived units and those accepted for use with SI.
3. ▶ indicates additional units accepted for use with SI for a limited time.
4. ▶ indicates conversion factors given exactly; other factors are given rounded, mostly to 4 significant figures. 
5. ▶ indicates a definition of an SI derived unit. [ ] in column 2-4 enclose factors given for the sake of completeness.

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<th>Column 1</th>
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<td>Multiply data given in:</td>
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<td>to obtain data in:</td>
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</tr>
<tr>
<td><strong>Radiation units</strong></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>◀ becquerel</td>
<td>1 Bq</td>
<td>(has dimensions of s⁻¹)</td>
<td></td>
</tr>
<tr>
<td>disintegrations per second (= dis/s)</td>
<td>1 s⁻¹</td>
<td>= 1.00 × 10⁰ Bq</td>
<td>◀</td>
</tr>
<tr>
<td>◀ curie</td>
<td>1 Ci</td>
<td>= 3.70 × 10¹⁰ Bq</td>
<td>◀</td>
</tr>
<tr>
<td>◀ roentgen</td>
<td>1 R</td>
<td>= 2.58 × 10⁻⁴ C/kg</td>
<td>◀</td>
</tr>
<tr>
<td>◀ gray</td>
<td>1 Gy</td>
<td>= 1.00 × 10⁰ J/kg</td>
<td>◀</td>
</tr>
<tr>
<td>◀ rad</td>
<td>1 rad</td>
<td>= 1.00 × 10⁻² Gy</td>
<td>◀</td>
</tr>
<tr>
<td>sievert (radiation protection only)</td>
<td>1 Sv</td>
<td>= 1.00 × 10⁶ J/kg</td>
<td>◀</td>
</tr>
<tr>
<td>rem (radiation protection only)</td>
<td>1 rem</td>
<td>= 1.00 × 10⁻⁴ J/kg</td>
<td>◀</td>
</tr>
<tr>
<td><strong>Mass</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>◀ unified atomic mass unit (u) of the mass of ¹²C</td>
<td>1 u</td>
<td>= 1.660 538 782 × 10⁻²⁷ kg, approx.</td>
<td></td>
</tr>
<tr>
<td>◀ tonne (= metric ton)</td>
<td>1 t</td>
<td>= 1.00 × 10³ kg</td>
<td>◀</td>
</tr>
<tr>
<td>pound mass (avoirdupois)</td>
<td>1 lbm</td>
<td>= 4.536 × 10⁻¹ kg</td>
<td>◀</td>
</tr>
<tr>
<td>ounce mass (avoirdupois)</td>
<td>1 ozn</td>
<td>= 2.835 × 10⁻² g</td>
<td>◀</td>
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<tr>
<td>ton (long) (= 2240 lbm)</td>
<td>1 ton</td>
<td>= 1.016 × 10⁰ kg</td>
<td>◀</td>
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<tr>
<td>ton (short) (= 2000 lbm)</td>
<td>1 short ton</td>
<td>= 9.072 × 10⁻² kg</td>
<td>◀</td>
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<tr>
<td><strong>Length</strong></td>
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</tr>
<tr>
<td>statute mile</td>
<td>1 mile</td>
<td>= 1.609 × 10⁰ km</td>
<td>◀</td>
</tr>
<tr>
<td>nautical mile (international)</td>
<td>1 n mile</td>
<td>= 1.852 × 10⁰ km</td>
<td>◀</td>
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<tr>
<td>yard</td>
<td>1 yd</td>
<td>= 9.144 × 10⁻¹ m</td>
<td>◀</td>
</tr>
<tr>
<td>foot</td>
<td>1 ft</td>
<td>= 3.048 × 10⁻¹ m</td>
<td>◀</td>
</tr>
<tr>
<td>inch</td>
<td>1 in</td>
<td>= 2.54 × 10⁻² mm</td>
<td>◀</td>
</tr>
<tr>
<td>mile (= 1.609 km)</td>
<td>1 mil</td>
<td>= 2.54 × 10⁻³ mm</td>
<td>◀</td>
</tr>
<tr>
<td><strong>Area</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>◀ hectare</td>
<td>1 ha</td>
<td>= 1.00 × 10⁴ m²</td>
<td>◀</td>
</tr>
<tr>
<td>◀ barn (effective cross-section, nuclear physics)</td>
<td>1 b</td>
<td>= 1.00 × 10⁻²⁸ m²</td>
<td>◀</td>
</tr>
<tr>
<td>square mile, (statute mile)²</td>
<td>1 mile²</td>
<td>= 2.590 × 10⁶ km²</td>
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</tr>
<tr>
<td>acre</td>
<td>1 acre</td>
<td>= 4.047 × 10³ m²</td>
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</tr>
<tr>
<td>square yard</td>
<td>1 yd²</td>
<td>= 8.361 × 10⁻⁴ m²</td>
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</tr>
<tr>
<td>square foot</td>
<td>1 ft²</td>
<td>= 9.290 × 10⁻² m²</td>
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<tr>
<td>square inch</td>
<td>1 in²</td>
<td>= 6.452 × 10⁻⁴ mm²</td>
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<tr>
<td><strong>Volume</strong></td>
<td></td>
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</tr>
<tr>
<td>◀ litre</td>
<td>1 l or 1 ltr</td>
<td>= 1.00 × 10⁻³ m³</td>
<td>◀</td>
</tr>
<tr>
<td>cubic yard</td>
<td>1 yd³</td>
<td>= 7.645 × 10⁻¹ m³</td>
<td>◀</td>
</tr>
<tr>
<td>cubic foot</td>
<td>1 ft³</td>
<td>= 2.832 × 10⁻² m³</td>
<td>◀</td>
</tr>
<tr>
<td>cubic inch</td>
<td>1 in³</td>
<td>= 1.639 × 10⁻⁴ mm³</td>
<td>◀</td>
</tr>
<tr>
<td>gallon (imperial)</td>
<td>1 gal (UK)</td>
<td>= 4.546 × 10⁻¹ m³</td>
<td>◀</td>
</tr>
<tr>
<td>gallon (US liquid)</td>
<td>1 gal (US)</td>
<td>= 3.785 × 10⁻¹ m³</td>
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<tr>
<td><strong>Velocity, acceleration</strong></td>
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<tr>
<td>foot per second (= fps)</td>
<td>1 ft/s</td>
<td>= 3.048 × 10⁻¹ m/s</td>
<td>◀</td>
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<tr>
<td>foot per minute</td>
<td>1 ft/min</td>
<td>= 0.08 × 10⁻³ m/s</td>
<td>◀</td>
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<tr>
<td>mile per hour (= mph)</td>
<td>1 mile/h</td>
<td>= 2.237 × 10⁻¹ m/s</td>
<td>◀</td>
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<tr>
<td>◀ knot (international)</td>
<td>1 knot</td>
<td>= 1.852 × 10⁰ km/h</td>
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<tr>
<td>◀ free fall, standard, g</td>
<td>1 g</td>
<td>= 9.807 × 10⁻² m/s²</td>
<td>◀</td>
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<tr>
<td>foot per second squared</td>
<td>1 ft/s²</td>
<td>= 3.048 × 10⁻³ m/s²</td>
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</table>

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<tr>
<td>Multiply data given in</td>
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<tr>
<td>Density, volumetric rate</td>
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<td></td>
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</tr>
<tr>
<td>pound mass per cubic inch</td>
<td>1 lbm/in³ = 2.786 × 10⁶ kg/m³</td>
<td></td>
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<tr>
<td>pound mass per cubic foot</td>
<td>1 lbm/ft³ = 1.602 × 10³ kg/m³</td>
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<tr>
<td>cubic feet per second</td>
<td>1 ft³/s = 2.832 × 10⁻³ m³/s</td>
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<tr>
<td>cubic feet per minute</td>
<td>1 ft³/min = 4.719 × 10⁻⁴ m³/s</td>
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<tr>
<td>Force</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>newton</td>
<td>1 N = 1.00 × 10⁶ N</td>
<td>m kg·s⁻²</td>
<td>#</td>
</tr>
<tr>
<td>dyne</td>
<td>1 dyn = 1.00 × 10⁻¹ N</td>
<td>#</td>
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<tr>
<td>kilogram force (≈ kilogram (kgf))</td>
<td>1 kgf = 9.807 × 10⁰ N</td>
<td></td>
<td></td>
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<tr>
<td>poundal</td>
<td>1 pdl = 1.383 × 10⁻¹ N</td>
<td></td>
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</tr>
<tr>
<td>pound force (avoirdupois)</td>
<td>1 lb = 4.448 × 10⁰ N</td>
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<td></td>
</tr>
<tr>
<td>ounce force (avoirdupois)</td>
<td>1 ozf = 2.780 × 10⁻¹ N</td>
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<tr>
<td>Pressure, stress</td>
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</tr>
<tr>
<td>pascal</td>
<td>1 Pa</td>
<td>= 1.00 × 10⁶ N/m²</td>
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</tr>
<tr>
<td>atmosphere (atm)</td>
<td>1 atm = 1.013 25 × 10⁵ Pa</td>
<td>#</td>
<td></td>
</tr>
<tr>
<td>bar</td>
<td>1 bar</td>
<td>= 1.00 × 10⁵ Pa</td>
<td>#</td>
</tr>
<tr>
<td>centimetres of mercury (0°C)</td>
<td>1 cmHg</td>
<td>= 1.333 × 10⁻³ Pa</td>
<td>#</td>
</tr>
<tr>
<td>dyne per square centimetre</td>
<td>1 dyn/cm² = 1.00 × 10⁻⁰ Pa</td>
<td>#</td>
<td></td>
</tr>
<tr>
<td>feet of water (4°C)</td>
<td>1 ftH₂O</td>
<td>= 2.989 × 10⁵ Pa</td>
<td></td>
</tr>
<tr>
<td>inches of mercury (0°C)</td>
<td>1 inHg</td>
<td>= 3.386 × 10⁰ Pa</td>
<td>#</td>
</tr>
<tr>
<td>inches of water (4°C)</td>
<td>1 inH₂O</td>
<td>= 2.491 × 10⁰ Pa</td>
<td></td>
</tr>
<tr>
<td>kilogram force per square centimetre</td>
<td>1 kg/cm² = 9.807 × 10⁰ Pa</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pound force per square foot</td>
<td>1 lbf/ft² = 4.788 × 10ⁱ Pa</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pound force per square inch (≈ psi)</td>
<td>1 lbf/in² = 6.895 × 10⁴ Pa</td>
<td></td>
<td></td>
</tr>
<tr>
<td>torr (0°C) (≈ mmHg)</td>
<td>1 torr</td>
<td>= 1.333 × 10⁻¹ Pa</td>
<td></td>
</tr>
<tr>
<td>Energy, work, quantity of heat</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>joule (≈ W·s)</td>
<td>1 J</td>
<td>= 1.00 × 10⁶ N·m</td>
<td>#</td>
</tr>
<tr>
<td>electronvolt</td>
<td>1 eV</td>
<td>= 1.602 19 × 10⁻¹⁹ J, approx.</td>
<td></td>
</tr>
<tr>
<td>British thermal unit (International Table)</td>
<td>1 Btu</td>
<td>= 1.055 × 10⁰ J</td>
<td></td>
</tr>
<tr>
<td>calorie (thermochemical)</td>
<td>1 cal</td>
<td>= 4.184 × 10⁰ J</td>
<td>#</td>
</tr>
<tr>
<td>calorie (International Table)</td>
<td>1 cal₁</td>
<td>= 4.187 × 10⁰ J</td>
<td>#</td>
</tr>
<tr>
<td>erg</td>
<td>1 erg</td>
<td>= 1.00 × 10⁻⁵ J</td>
<td>#</td>
</tr>
<tr>
<td>foot-pound force</td>
<td>1 ftlb</td>
<td>= 1.356 × 10⁰ J</td>
<td></td>
</tr>
<tr>
<td>kilowatt-hour</td>
<td>1 kW·h</td>
<td>= 3.60 × 10⁶ J</td>
<td>#</td>
</tr>
<tr>
<td>kiloton explosive yield (PNE) (≈ 10¹⁵ g·c)</td>
<td>1 kt yield</td>
<td>= 4.2 × 10⁶ J</td>
<td>#</td>
</tr>
<tr>
<td>Power, radiant flux</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>watt</td>
<td>1 W</td>
<td>= 1.00 × 10⁶ J/s</td>
<td>#</td>
</tr>
<tr>
<td>British thermal unit (International Table) per second</td>
<td>1 Btu/s</td>
<td>= 1.055 × 10⁰ W</td>
<td></td>
</tr>
<tr>
<td>calorie (International Table) per second</td>
<td>1 cal/s</td>
<td>= 4.187 × 10⁰ W</td>
<td></td>
</tr>
<tr>
<td>foot-pound force/second</td>
<td>1 ftlb/s</td>
<td>= 1.356 × 10⁰ W</td>
<td></td>
</tr>
<tr>
<td>horsepower (electric)</td>
<td>1 hp</td>
<td>= 7.46 × 10⁰ W</td>
<td>#</td>
</tr>
<tr>
<td>horsepower (metric) (≈ ps)</td>
<td>1 ps</td>
<td>= 7.355 × 10⁰ W</td>
<td></td>
</tr>
<tr>
<td>horsepower (550 ft·lb/s)</td>
<td>1 hp</td>
<td>= 7.457 × 10⁰ W</td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>temperature in degrees Celsius, °C</td>
<td>T = T₀ + ΔT</td>
<td>gives</td>
<td>t (in degrees Celsius)</td>
</tr>
<tr>
<td>where T is the thermodynamic temperature in kelvin</td>
<td></td>
<td></td>
<td>#</td>
</tr>
<tr>
<td>and T₀ is defined as 273.15 K</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>degree Fahrenheit</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>degree Rankine</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>degree temperature difference</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 Btu/in²·h·°F (International Table Btu)</td>
<td>1 Btu/(ft²·h·°F) (International Table Btu)</td>
<td>1 cal/ï¿œ (International Table Btu)</td>
<td>5.192 × 10⁻⁵ W·m⁻¹·K⁻¹</td>
</tr>
</tbody>
</table>

a atm abs, ata: atmospheres absolute; atm (g), ata: atmospheres gauge.  
b lbf/in² (g) (≈ psi): gauge pressure;  
lbf/in² abs (≈ psia): absolute pressure.  
c The abbreviation for temperature difference, deg (≈ degK = degC), is no longer acceptable as an SI unit.
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