

5 TRITIUM IN THE ATMOSPHERE

5.1 CHARACTERISTICS OF TRITIUM

The radioactive isotope of hydrogen of mass 3 (^3H , or *tritium*) has a half-life of about 12 years and thus a lifetime commensurate with many hydrological processes (more details are given in Volume I). By and large, ^3H is introduced into the hydrological cycle in the atmosphere, where it is produced naturally by the interaction of cosmic radiation with atmospheric components. The major reaction involved is that of thermal neutrons with nitrogen



High-energy spallation reactions and direct accretion of ^3H from the solar wind are believed to be of less importance (Nir et al., 1966).

As from the early 1950s anthropogenic sources, especially from nuclear tests in the atmosphere, overshadowed the natural production for more than a decade, as will be discussed.

In groundwater-hydrology and oceanography ^3H concentrations are generally given as TU (Tritium Unit), equivalent to a concentration of 10^{-18} . Other disciplines may use the specific radioactivity in Bq (Bequerel) or mBq, related to TU by:

$$1\text{ TU} \equiv 0.118 \text{ Bq/L of water} (\equiv 3.19 \text{ pCi/L})$$

or: $1 \text{ Bq/L} \equiv 8.47 \text{ TU}$

The most up-to-date value for the half-life is 12.32 year (Lucas and Unterweger, 2000).

Fig.5.1 shows the ^3H concentration in monthly precipitation at Vienna (data from the GNIP network). Data from the Ottawa station show that the ^3H increase started already during the 1950ies.

From known-age wine samples Begemann (1959) and, independently, Roether (1967) estimated that the natural ^3H content of rain before the nuclear test series in the 1950s began was about 5 TU in central Europe. The average natural production rate was estimated to be about $0.20 \text{ }^3\text{H atoms/cm}^2\text{sec}$. The production of ^3H takes place preferentially in the upper troposphere and lower stratosphere. It is introduced into the hydrologic cycle following oxidation to tritiated waters ($^3\text{H}^1\text{HO}$), seasonally leaking down into the troposphere mainly through the tropopause discontinuity at mid-latitudes.

At the peak ^3H concentration during spring 1963 the ^3H content of precipitation at the northern hemisphere was about 5000 TU (Fig.5.1). Considering the rough estimate of the amount of water in the troposphere of about 2×10^{16} kg (≈ 4 g/cm²), the total ^3H inventory in the northern troposphere was then in the order of 10 kg or roughly 3 kmole. This is to be compared with an estimated amount of simultaneously present ^{14}C of about 50 kmole (Chapter 6). The cross section for the nuclear reaction, however, is more than a few hundred times smaller than that for ^{14}C production (Libby, 1965). The conclusion then must be that the bomb-derived ^3H largely originated from a direct release of ^3H from the bombs, rather than by the nuclear reaction by neutrons released during the explosions. Mason et al. (1982) established that the direct ^3H injection in the atmosphere by fusion bombs is about 1.5 kg per megaton of explosive force.

The pattern shown in Fig.5.1 is repeated at most northern hemisphere stations, albeit with slightly varying amplitudes and phase shifts. The notable feature of this curve is a yearly cycle of maximum concentrations in spring and summer and a winter minimum, with typical concentration ratios of 2.5 to 6 between maximum and minimum values. The annual cycle is superimposed upon the long-term changes which have ranged over three orders of magnitude since 1952.

There is a marked latitude dependence: concentrations are highest north of the 30th parallel, with values lower by a factor of 5 or so at low-latitude and tropical stations. In the southern hemisphere (represented by Pretoria) the yearly cycle is displaced with the season by half a year, and the mean ^3H levels in atmospheric waters are lower than at comparable north-latitude stations. This is a reflection of the predominant northern location of weapon testing sites and the slow inter-hemispheric transport of tracers. Consequently, in the southern hemisphere the increase has been a factor of 10 to 100 smaller (Fig.5.1), because of the equatorial barrier in the global air mass circulation and the fact that the annual ^3H injected during spring from the stratosphere into the troposphere is removed from the latter very efficiently within weeks.

5.2 GEOPHYSICAL ASPECTS

Tritium concentrations and inventories, like that of other atmospheric components of stratospheric origin, are dominated by the timing, location and intensity of exchange of tropospheric and stratospheric air masses, as well as, of course, the ^3H concentration in the stratosphere at the time that such an exchange takes place. Exchange occurs predominantly during late winter and in spring (the so-called spring leak of the tropopause) in the region of baroclinic zones and tropopause discontinuities of the mid-latitudes (Newell, 1963).

The changing ^3H inventory of the stratosphere of recent years reflects the massive injections by weapon tests in 1954, 1955, 1958 and again during 1961-1962, mostly in the northern hemisphere, reaching high altitudes in the stratosphere.

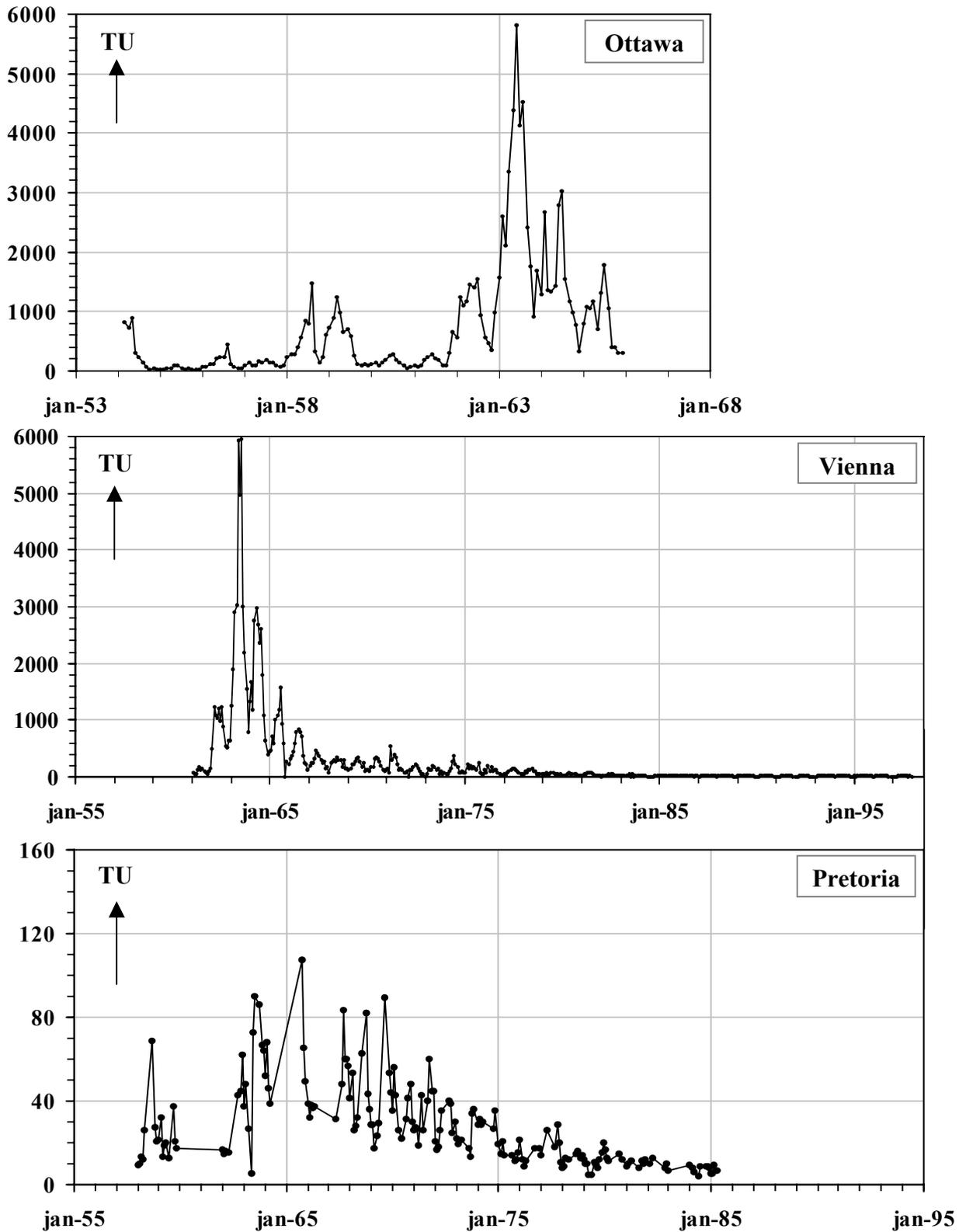


Fig.5.1 ^3H in monthly precipitation samples of stations representative for the northern (Ottawa, Vienna) and the southern hemisphere (Pretoria) (data from the GNIP network) (data valid for the year of sample collection).

At any time, the inventory decreases by 5.5% per year through radioactive decay and some of the ^3H leaks into the troposphere from where it is lost into the ocean or groundwater, both of which can be considered a sink for the stratospheric ^3H . Estimated residence times of tritiated water vapour in the lower stratosphere are of the order of a few years. Inter-hemispheric mixing in the stratosphere seems to occur on a similar time scale.

The residence time of water in the lower troposphere, on the other hand, is of the order of 5 to 20 days. This is a short period relative to large-scale north-to-south mixing in the troposphere, but within the time scale of horizontal atmospheric motions. As a result ^3H is deposited onto the surface of the earth within the latitude band of its penetration from the stratosphere or more precisely of its distribution on top of the so-called moist layer, which extends to 500mbar approximately (Eriksson, 1966).

From measurements on atmospheric vapour by Ehhalt (1971) it appears that above the 2-km level the ^3H content is uniform over both land and sea, along each latitude band. Over the continents during summer, there is an increase in ^3H amount in the lowest 2 km, apparently due to re-evaporation of part of the winter and spring precipitation. In contrast, ^3H levels are low over the sea throughout the year, as a result of the uptake of ^3H by molecular exchange into the oceans. Moisture evaporated from the ocean is consequently low in ^3H content due to the long residence time of water in the ocean. The delay in the appearance of the annual ^3H peak in precipitation relative to the time of its injection (June vs. the late winter months) is attributed by Ehhalt to this re-evaporation of moisture from the continents, which provides an additional source of ^3H to the atmosphere during summer.

^3H build-up over the continent comes about as a result of the cutting off of the supply of low activity oceanic vapour, while influx from aloft continues. The inland gradient could be expected to be highest during late winter, spring and summer when the downward flux is at its peak value. This effect is, however, somewhat balanced by the lower content of vapour in the winter atmosphere.

As stated, re-evaporation of moisture from the continent during summer acts to extend the spring maximum of ^3H concentrations into the summer, but does not affect the inland build-up of ^3H content (expressed in ^3H units) except when there is a hold-up (delay) of water, so that the re-evaporated moisture has a noticeably different ^3H age than the atmospheric moisture. Ages of a few years are quite typical for soil moisture or for waters in sizeable inland lakes. During periods of rising ^3H levels, such as the decade of 1952-1963, the continental water reservoirs were relatively low in ^3H content and their evaporation reduced the continental gradient. During years with declining atmospheric ^3H levels, the continental reservoirs may retain the memory of high ^3H levels of the past and reverse the normal ^3H flux, contributing ^3H to the atmosphere. In this case the inland ^3H gradient is increased.

Build-up of ^3H concentrations over continents is quite gradual. For example, TU levels double over Central Europe over a distance of 1000 km. The interaction over the ocean, on the other hand, becomes effective over very short distances, especially when the continental air is very

unsaturated relative to ocean surface waters. An extreme case is found in the Mediterranean Sea, where the intense sea-air interaction is mirrored in the extreme drop in ^3H activity, to 10% of the continental value. The inland gradient on the Eastern Mediterranean shore also is more abrupt than usual, the TU doubling length being about 100 km (Gat and Carmi, 1970) due to the limited extent and intra-continental position of the Mediterranean Sea.

Precipitation is the main mechanism for removing ^3H from the atmosphere over the continent. Moreover it is the vehicle for the downward transport of ^3H within the troposphere. As a result of the rapid exchange of isotopes between the rain droplets and ambient vapour, the falling rain drops contribute ^3H to the lower troposphere during the period where strong vertical ^3H gradients exist. Indeed, Ehhalt (1971) has noticed an additional source of ^3H in the lower atmosphere, at a height just below the freezing level. Only the frozen phases, i.e. hail and snow, are not subject to this exchange and can carry the high ^3H levels all the way to the ground.

On a global average, however, the molecular exchange of water between the air and the ocean needs to be added to loss of ^3H by precipitation. This exchange appears to increase the rate of ^3H loss by a factor of up to 1.9 (Lipps and Helmer, 1992).

The atmospheric moisture system has been treated as a box model by Bolin (1958) and Eriksson (1967). In these models ^3H is added from aloft (F_T); loss occurs through rainout and exchange with the ^3H deficient surface waters over the ocean while over land the loss is to the groundwater systems or through runoff. The soil, which returns most of the precipitated ^3H through evapotranspiration acts as a buffer which maintains high continental ^3H levels.

Fig.5.2A shows the ^3H budget of the atmosphere in a box model representation. a , a' and b are the water content in continental and marine atmosphere and in the soil column, respectively. S_a , $S_{a'}$, S_b , $S_0 = ^3\text{H}$ content of these reservoirs and of the oceanic surface layers, respectively (in TU); S_b^* , the ^3H content of recharging waters, may differ from S_b because of hold-up of water in the soil column. $F_T = ^3\text{H}$ influx from aloft (in moles/s). P_c , P_m , E , E_T , R_s , R_G and $X =$ the precipitation amounts (continental and marine), evaporation and evapotranspiration fluxes, surface and groundwater runoff and the molecular exchange flux between ocean and atmosphere, respectively.

Fig.5.2B is a schematic picture of deposition of ^3H and concentration of ^3H in precipitation in a westerly zonal air current (Eriksson, 1967). The length of the arrows is proportional to the rate of vertical transports. The dashed line indicates the smoothing in the concentrations which will take place due to longitudinal eddy mixing.

During recent years, the atmospheric reservoir has been practically exhausted of the ^3H introduced by the nuclear tests so that the atmospheric ^3H levels have almost returned to the pre-1952 levels, except for some local anthropogenic releases of ^3H from the nuclear industry and other uses of tritiated materials.

In the next sections we will discuss some points of special hydrological interest in the ^3H curves.

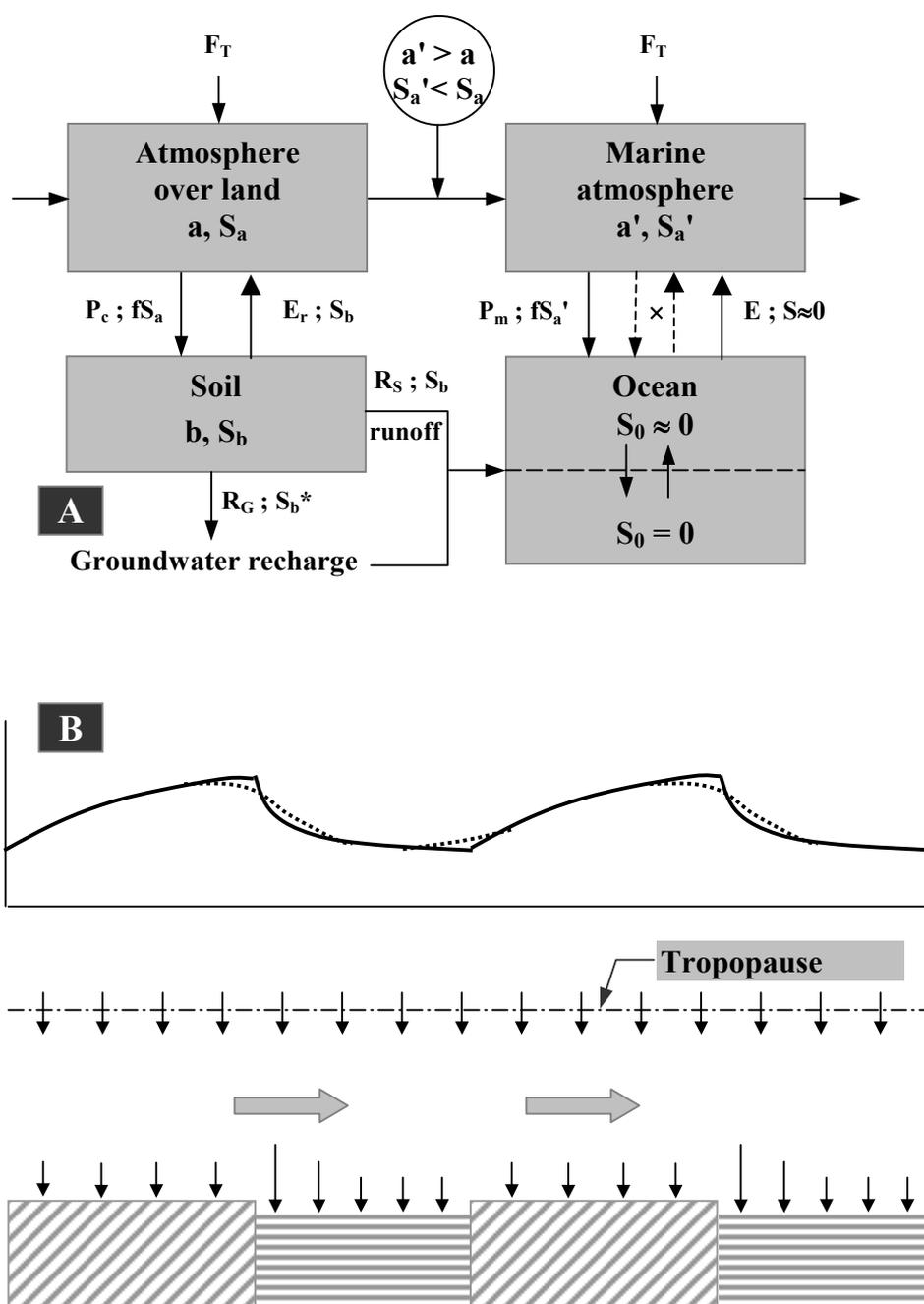


Fig.5.2 A. ^3H budget in the atmosphere in box model representation (after Bolin, 1958 and Begeman, 1960); a , a' and b are the water content in continental, marine and soil layer; s values refer to the respective ^3H contents; F_T is the ^3H influx from aloft.

B. schematic presentation of the concentration of ^3H and its deposition on land or sea in a westerly air current (Eriksson, 1967).

5.3 HYDROLOGICAL ASPECTS

For the hydrologist it is of interest to be able to judge ^3H concentrations in the underground, in order to have an first impression of the hydrological situation, and of the necessity and specific conditions of sample collection. Since the ^3H content of precipitation has decreased almost to their natural level, part of the applications exploited during the 1960ies and 1970ies are no longer possible. Nevertheless, excess ^3H is still present in the ground and in surface waters. Therefore, knowledge of possible variations is still relevant.

In the next sections we will give a broad survey of ^3H variations in precipitation.

5.3.1 LONG-TERM RECOVERY OF NATURAL ^3H LEVELS

There are two reasons why ^3H in precipitation has returned quickly to its natural level, at least in comparison with ^{14}C in atmospheric CO_2 :

- 1) ^3H has a relatively short half-life (12.32 years; Lucas and Unterweger, 2000). This means that by decay alone the peak concentration from 1963 would become reduced by a factor of $2^{10} \approx 1000$ in a period of about 120 years (= 10 half-lives) provided no further nuclear tests in the atmosphere are made. In the period of three half-lives since 1963 the peak concentration has been reduced by a factor of 8. Fig.5.3 shows the data for Vienna and Pretoria from Fig.5.1 when corrected for radioactive decay.
- 2) The atmospheric water circulation through ocean-air exchange is very vigorous. The water in the troposphere is replaced about every 10 days. Therefore, bomb ^3H is rapidly transported to the ocean, even though ^{14}C is not (Sect.6.4). On the other hand, most of the ^3H produced by fusion bombs entered the stratosphere, from which it has only gradually been leaking back to the troposphere, so that ^3H stayed in the atmosphere for a longer time.

Because of the short turn-over time of atmospheric water and not much ^3H is left in the stratosphere, the tropospheric ^3H concentration has decreased to the relatively stable original level. It is only since the seventies, following a long period of limited nuclear test activity, that the northern and southern hemisphere ^3H concentrations are becoming comparable.

5.3.2 SEASONAL VARIATIONS IN ^3H

The seasonality of the stratosphere-to-troposphere transport results in the marked seasonal cycle in the ^3H content of precipitation (Fig.5.1), opposite in phase between the northern and southern hemisphere.

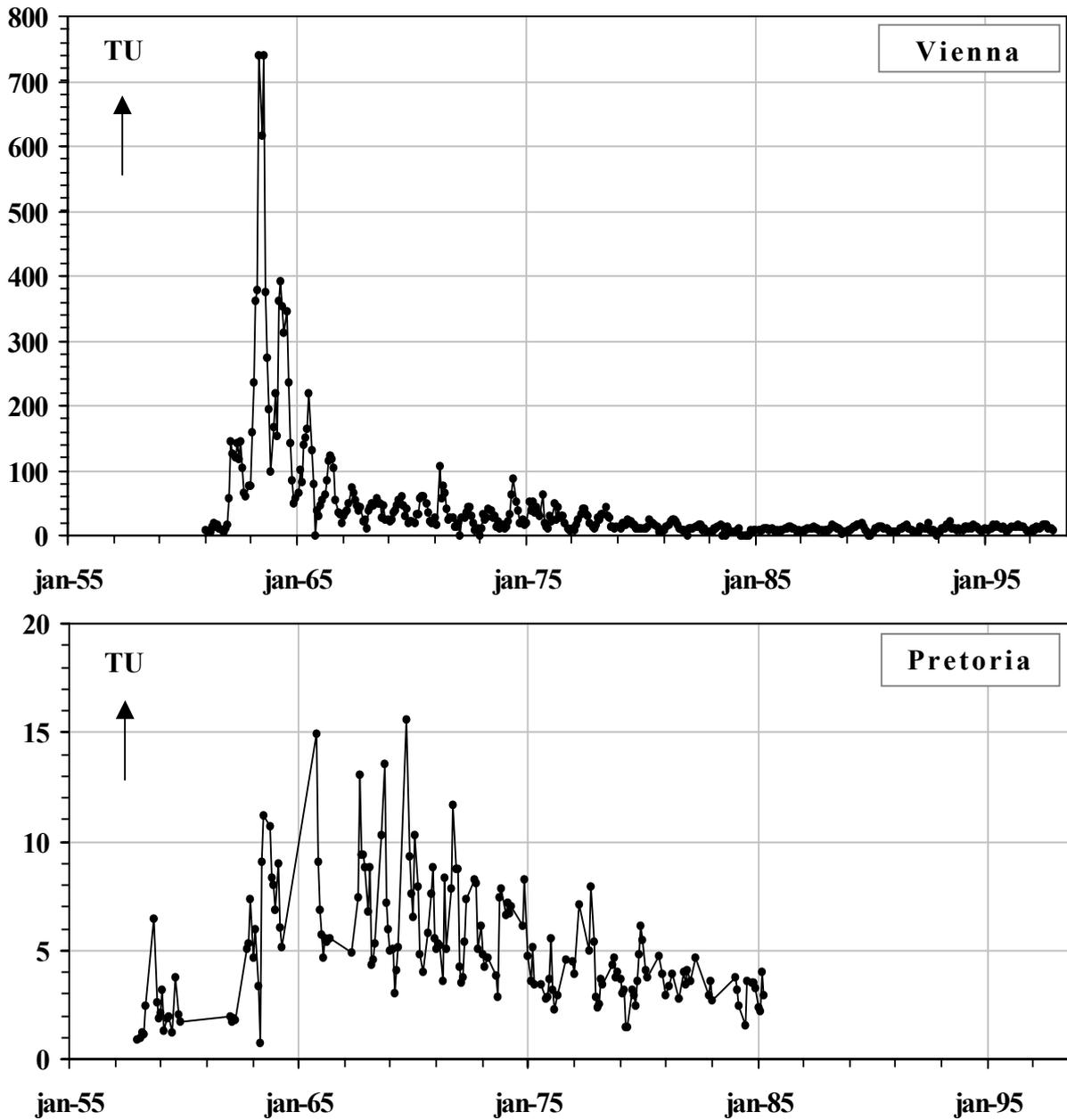


Fig.5.3 ^3H in precipitation at Vienna (representing the northern hemisphere) and Pretoria (representing the southern hemisphere), corrected for radioactive decay during the period between the moment of sampling and the year 2000 (original data from Fig.5.1).

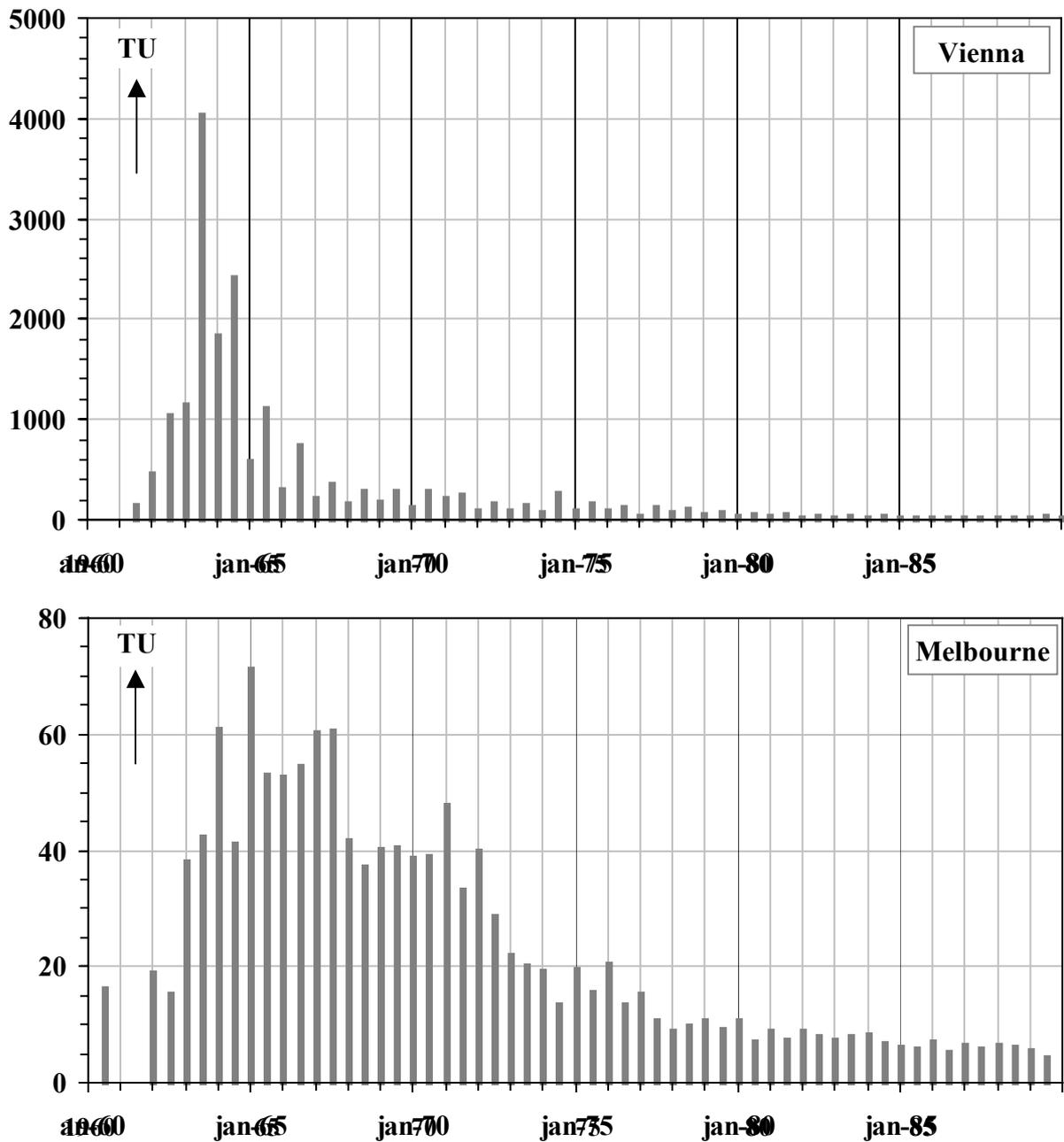


Fig.5.4 ^3H values for "winter" (October-March) and "summer" (April-September, between scale divisions) precipitation in Vienna, representing the northern hemisphere and similar values for Melbourne at the southern hemisphere. The seasonal cycle for the latter series is less pronounced (weighted averages from the GNIP network). In the southern hemisphere the ^3H (and ^{14}C) maxima have a phase shift of half a year, because the leak between stratosphere and troposphere occurs in early spring. Fig.5.5 shows an example of the absence of high "summer" values in the unsaturated zone in a dune area in NW Europe, meaning that here the "summer" rain did not reach the groundwater table (see also: Volume IV, Sect.5.1.4.2) (data valid for the year of sample collection).

Fig.5.4 contains two graphs of separate "winter" and "summer" ^3H patterns, one containing the weighted means for the months October-March, taken as representative for winter, April-September for the summer months at the northern hemisphere, whereas similar data of Melbourne represent the southern hemisphere. This phenomenon is caused by temporary mixing between the stratosphere and troposphere at high latitudes in early spring, so that ^3H , originally injected into the stratosphere by nuclear explosions, can return to the troposphere.

For hydrologists this is a very important aspect of the temporary elevated ^3H levels about 30 years ago. The reason is that the infiltration of precipitation is not a phenomenon distributed evenly over the year. Generally groundwater recharge occurs after heavy rainfall and without significant (evapo)transpiration of the vegetation. In moderate climates infiltration is therefore limited to the "winter-period" with relatively low ^3H values (see later Fig.5.5). This point is illustrated in Fig.5.5 where a ^3H profile in a sandy dune area with presumably vertical infiltration is compared with ^3H in winter precipitation (October-March). Because for this region no ^3H data are known for the early 1960ies, we have used for comparison the data from Vienna and Ottawa which are comparable for the later overlapping period. A degree of dispersion has been allowed for equivalent to a running average of the ^3H data of 5 consecutive years. Obviously the summer precipitation does not significantly infiltrate.

5.3.3 GEOGRAPHICAL VARIATIONS IN ^3H

In Figs.5.1 and 5.4 we have compared the ^3H content of precipitation at different stations. As we have seen, the most striking point is that the very high ^3H levels are confined to the northern hemisphere: in the southern hemisphere, the ^3H content increased by a factor of hardly more than 10 above the pre-bomb levels. The variations at stations in the N. hemisphere show generally the same pattern. However, the ^3H concentrations themselves may be quite different from one station to another.

By an effect similar to the continental effect and the (small) seasonal effect for ^{18}O and ^2H , low ^3H values are found near the ocean. A large fraction of the local water vapour, and thus of the precipitation, consists of oceanic vapour which is low in ^3H . The highest values apply to samples from the North Atlantic Ocean (Östlund and Fine, 1979; Weiss et al., 1979).

5.3.4 SMALL-SCALE ^3H VARIATIONS

Local variations are likely to be small, because the ^3H content in rain is not influenced by temperature variations (as are ^{18}O and ^2H). Although also ^3H is fractionated during evaporation and condensation processes -at twice the extent as ^2H (Bolin, 1958)-, the variations involved are in the order of 16% (twice 80‰), equivalent to just one year of radioactive decay. Therefore, they can not be clearly distinguished in hydrological realities and are thus neglected.

Under normal conditions we would not expect significant variations in the ^3H content of the vapour within one air mass.

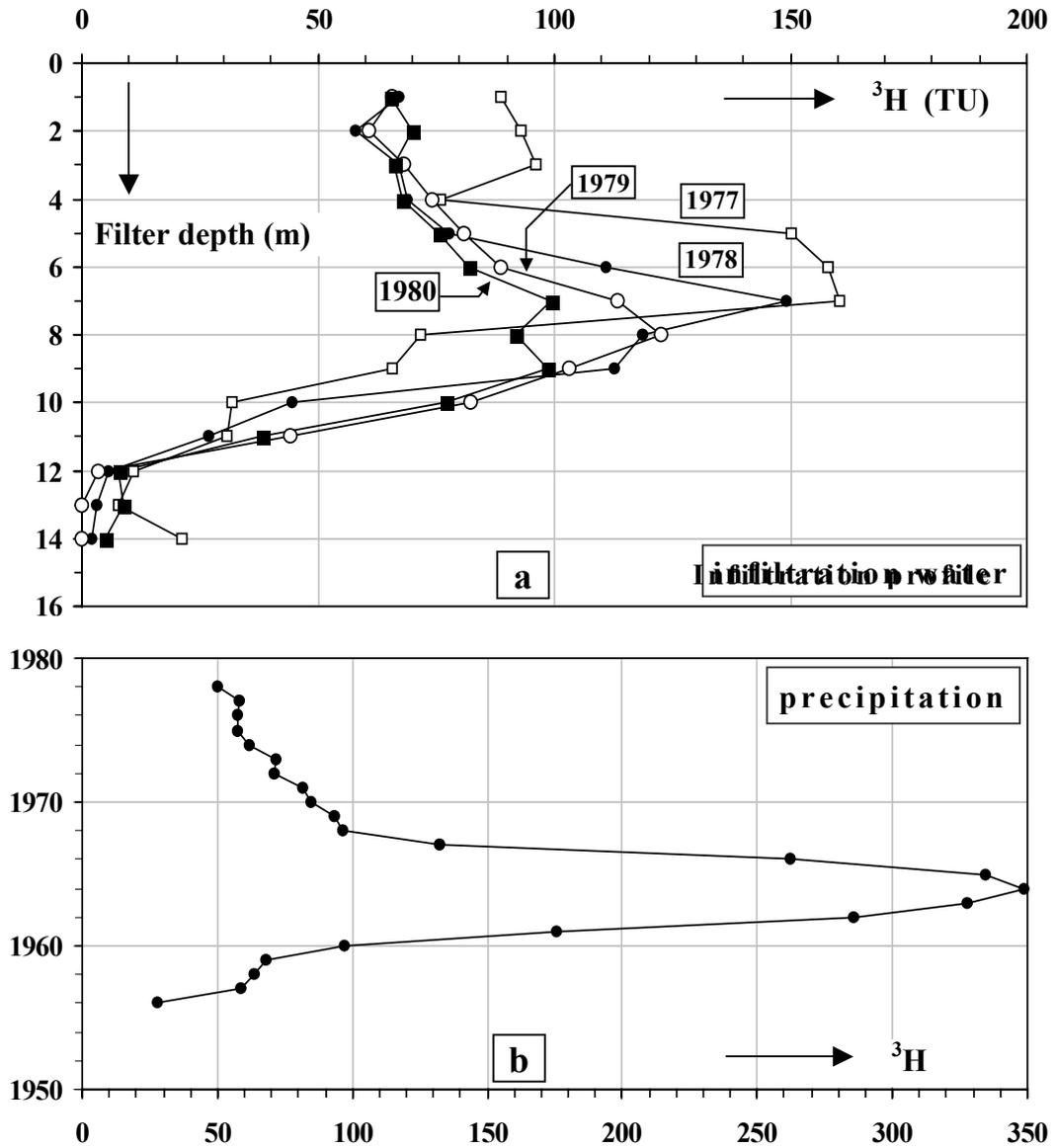


Fig.5.5 a) ^3H profile of infiltrated water in a dune area in NW Europe (Monster, the Netherlands). The high ^3H level of 1963 did not reach the groundwater (all data corrected for radioactive decay till 1980);
 b) ^3H in precipitation at Vienna/Ottawa, reasonably representative for precipitation in the study area. The values refer to winter precipitation (October-March); a dispersion has been introduced equivalent to 5 years moving average (all data valid for 1980).

5.3.4.1 SMALL-SCALE SPATIAL ^3H VARIATIONS

From a series of ^3H data of monthly precipitation from 15 stations within the small country of the Netherlands (about 30 000 km²) we concluded that between the stations there are no significant regular differences in seasonal effect. Existing differences over this region are irregular and small. Averaged over the year, however, a small continental effect is apparent.

Another example over a longer period is presented by four stations within 50 km around Vienna. The seasonal variations are parallel and the yearly averages are comparable. Discrepancies between the stations do not seem to be systematic.

5.3.4.2 SMALL-SCALE TEMPORAL ^3H VARIATIONS

Fast fluctuations in ^3H can be expected, if in a short period different air masses contribute to the precipitation at a site. Precipitation collected during a severe convective storm showed no significant differences within a period of 30 minutes (Groeneveld, 1977), although significant variations in ^{18}O and ^2H were noted. A fast change in ^3H might occur during the passage of another air mass, for instance correlated with a cold front. Large differences in ^3H content have been observed within a hurricane, resulting from complicated meteorological conditions (Östlund, 1967)