Underground radioactivity measurements *in HADES and Europe*

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IRMM - Institute for Reference Materials and Measurements

*Geel - Belgium*

http://irmm.jrc.ec.europa.eu/

http://www.jrc.ec.europa.eu/
NEW TECHNOLOGIES

• New technological developments drive science and society forward

• Developments in low-level radioactivity measurements open opportunities for studying more processes in nature (atmosphere/ocean water/food chain…)

Background Comparison – Gamma-ray spectrometry

International Symposium on Isotopes in Hydrology, 27 March - 1 April 2011, Monaco

Energy (keV)

A: “Normal”
B: “Low-level”
C: Felsenkeller
D: HADES
E: Gran Sasso

511 keV
1460 keV
2614 keV

Ge(n,n'γ)
Pb(n,n'γ)

Normalised count rate (keV⁻¹ d⁻¹ kg⁻¹ Ge⁻¹)

0 250 500 750 1250 1500 1750 2000 2250 2500

0.0001 0.001 0.01 0.1 1 10 100 1000 10000
Primary cosmic ray: $10^3 \text{ m}^{-2} \text{s}^{-1}$

9 % $\alpha$

90 % p

1 % heavier nuclei (up to Fe)

Extremely high energies from outer space, GeV range from sun

Atmosphere

Earth

225 m underground ($\text{m}^{-2}\text{s}^{-1}$)

<table>
<thead>
<tr>
<th>$\pi^\pm$</th>
<th>p</th>
<th>$\mu^\pm$</th>
<th>$\nu_\mu$</th>
<th>$n_{\text{therm}}$</th>
<th>$n_{\text{fast}}$</th>
<th>$\nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>2</td>
<td>40</td>
<td>60</td>
<td>200</td>
<td>500</td>
<td>$10^{11}$</td>
</tr>
</tbody>
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<td>&lt;2</td>
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<td>$10^{11}$</td>
</tr>
</tbody>
</table>
\[ A = \frac{C}{t_m P_{\gamma} \varepsilon} \]

- \( A \) = Activity (Bq)
- \( C \) = Net peak counts
- \( t_m \) = Measurement time (s)
- \( P_{\gamma} \) = Gamma-ray emission probability
- \( \varepsilon \) = detection efficiency
Activity calculations with correction factors

\[ A = C_{TOT} - C_{Peak} - C_{Continuum} \times \frac{\epsilon_{MC}}{\epsilon_{REF}} \times \frac{\epsilon_{Sample}}{\epsilon_{REF}} \times e^{\lambda t_d} \times \frac{\lambda}{(1-e^{-\lambda t_m})} \times K_1 K_2 K_3 \]

- \( K_1 \) = summing correction
- \( K_2 \) = Branching correction
- \( K_3 \) = Equilibrium correction

\( t_d \) = decay time (to a reference date)
\( t_m \) = measurement live time

Combined activities from several gamma-rays to activity for one radionuclide

Ex.: \(^{226}\)Ra from \(^{214}\)Bi and \(^{214}\)Pb

Combined activities from several daughters to one parent

Reference sample similar (sum corrected?)

Correction factor from e.g. MonteCarlo code

Equilibrium?
**Minimum Detectable Activity**

\[ MDA \propto \frac{\sqrt{CR_{Bkg}}}{\sqrt{t_m}} \cdot \frac{1}{\varepsilon} \]

*MDA* = Minimum Detectable Activity (Bq)

*CR* sub *Bkg* = Background Count Rate (s^-1)

*tm* = Measurement time (s)

*\varepsilon* = detection efficiency
Improving MDA

$$MDA \propto \sqrt{\frac{CR_{Bkg}}{t_m}} \cdot \frac{1}{\varepsilon}$$

\(\varepsilon\): Increasing detector size will also increase background.

\(\varepsilon\): Increasing sample size may also increase background.

\(t_m\): “only” scales with square root.

It is worth while spending efforts to reduce background in order to obtain better MDAs.
Background

Note that for low-background systems peak count-rates may decrease more than continuum levels

<table>
<thead>
<tr>
<th></th>
<th>Peak count rate / d$^{-1}$ per kg Ge</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>295 keV</td>
</tr>
<tr>
<td>&quot;Normal&quot;</td>
<td>2953</td>
</tr>
<tr>
<td>Low-background</td>
<td>0.75</td>
</tr>
<tr>
<td>ultra-low-background</td>
<td>0.25</td>
</tr>
</tbody>
</table>
**Detection limits**

### Interference free detection limits for a 7-day measurement

<table>
<thead>
<tr>
<th></th>
<th>Air filter mBq</th>
<th>Water on 2L Marinelli mBq/L</th>
<th>Water on filter* Ba Co-prec. mBq/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{140}$Ba</td>
<td>0.2</td>
<td>6</td>
<td>-</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>0.1</td>
<td>2</td>
<td>-</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>0.2</td>
<td>4</td>
<td>0.5</td>
</tr>
</tbody>
</table>

*Depends on amount of water used. Here 2 L.*
How long measurement time can you afford?

1 mBq ~ decay per hour
1 μBq ~ decay per week

⇒ To carry out big projects and measurement of numerous samples, networking is essential

Collaboration of European Low-level underground LAboRatories
Collaboration of European Low-level underground LAboRatories

Mission: To promote higher quality and sensitivity in ultra low-level radioactivity measurements for the improvement of crisis management, environment, health and consumer protection standards of Europe.
• Modane - France (-2200 m)
• Gran Sasso - Italy (-1700 m)
• Asse/PTB - Germany (-415 m)
• HADES – EU/Belgium (-225 m)
• Unirea, Romania (-208 m)
• University of Iceland (-165 m)
• Baradello Hill, Italy (-100 m)
• Ferrière (LEGOS)-France (-80 m)
• Felsenkeller - Germany (-50 m)
• CAVE – Monaco (-15 m)
• MPI-Heidelberg - Germany (-10 m)
+ associated partners
  e.g. Solotvina salt mine (Ukraine)
Unirea salt mine, Romania

Experiments for radiation background measurement

- high resolution spectrometry systems (Canberra, Ortec)
- TLDs
- Eberline FH 40G
- epithermal neutron activation analyses of salt and salt impurities
- passive Radon detectors
HADES = High Activity Disposal Experimental Site
– Operated by EURIDICE* and located at SCK•CEN in Mol

*European Underground Research Infrastructure for Disposal of nuclear waste In Clay Environment

HADES

Overburden:
~ 175 m sand
~ 50 m clay

Location of IRMM’s ULGS setup

First shaft
Constructed 1999

Second shaft
Constructed 1999

Connecting gallery
Constructed 2003

Test drift
67 m

PRACLAY gallery

223 m

84 m

39 m
Going underground...
The Sandwich Spectrometer

Increased solid angle

Pb shield = radiopure lead, 4 cm, 2.5 Bq/kg
+14.5 cm lead, 20 Bq/kg

Cu lining = radiopure copper, 3.5 cm

Detector mass ~ 1.9 kg each
Low-level measurements – a growing field!

Isotopic fingerprints
- Hiroshima
- Neutrinos / $\beta\beta$

JET
- Tokai-mura
- Safeguards

Ref. measurements
- Decay data
- Radiation Protection

HADES 1999-2002 (FP5)
- Small samples
- High temporal resolution
- Benchmarking
- Fast measurements
- Radiopurity for detector construction

- $^{210}$Pb in lung cells 3%
- $^{210}$Pb and Th in human bones 15%
- $^{26}$Al intercomparison 3%
- Neutron data 3%
- Reference materials 4%
- $^{60}$Co in steel from Hiroshima 3%
- $^{60}$Co in German steel 1%
- Maintenance and new installations 7%
- Various 7%
- $^{137}$Xe in GaAs 2%
- BOREXINO 5%
- Nuclear safeguards 3%
- Decay data 3%
- Neutrinos 3%
- JCO accident 11%
- Bkg+calibration 19%
- Radioprotection 14%
- Ref.measurements 4%
- Neutrinos / $\beta\beta$ 7%
- Maintenance and new installations 7%
- 60Co in German steel 1%
- $^{210}$Pb in lung cells 3%
- 60Co in steel from Hiroshima 3%
- Various 7%
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- Radiopurity for detector construction
At present: Decay energy of $^{115}$In(9/2+) $\rightarrow$ $^{115}$Sn(3/2+) $1.7\pm4.0$ keV ($\Delta m = 499$ keV)


⇒ Not for sure if it is energetically possible or not
Lowest decay energy known to man

Half-life: $4.1 \times 10^{20}$ years

Decay energy: 155 eV
Low-level measurements – a growing field!

Isotopic fingerprints

Hiroshima

Neutrinos / $\beta\beta$

JET

Tokai-mura

Ref. measurements

Decay data

Radiation Protection

HADES 1999-2002 (FP5)

Small samples

High temporal resolution

Benchmarking

Fast measurements

Radiopurity for detector construction
Gamma-ray spectrum of one of the spoons measured in HADES (red) and background above ground (black) and in HADES (blue).
Thermal neutrons fluence values estimated from measurement of $^{60}$Co and $^{51}$Cr in stainless steel spoons (10$^6$ cm$^{-2}$)
Why Gamma-ray Spectrometry?

• Easy sample preparation
  • Non destructive
  • Low running cost

Why Ultra Low-level Gamma-ray Spectrometry?

In addition to above:
  • Low detection limits
    (improvement: 10-300 times)
    • More robust
    • Potentially faster
  • Potential to achieve higher temporal resolution
    • Potential to sample small volumes

⇒ More interesting applications are feasible
“Reference” for other methods

• No sample preparation!
  Direct measurement; Non-Destructive

• There is a need to check methods
  that require sample preparation

Radiochemical methods  Mass spectrometry methods

Some examples

$^{40}\text{K}$ in water (ICP-MS),  $^{26}\text{Al}$ in meteorites (AMS)

Zn in GaAs (ERS and GDMS),  $^{60}\text{Co}$ (radiochemistry)

Checking of neutron detectors
International collaborations

To compare radioactivity measured in different laboratories one need to ascertain correct measurements

- reliable and comparable measurement results are based on their traceability to measurement units
- participation in Proficiency Testing schemes to confirm

IRMM is organising Proficiency Testing for nominated European laboratories monitoring radioactivity in the environment

- $^{137}\text{Cs}$ in air filters (2007)
- $^{137}\text{Cs}$, $^{40}\text{K}$, $^{90}\text{Sr}$ in milk powder (2008)
- $^{226}\text{Ra}$, $^{228}\text{Ra}$, $^{234}\text{U}$, $^{238}\text{U}$ in mineral waters (2010)
- run at present: radionuclides in soil, among them several NORM
Euratom comparisons

IRMM approach to these comparisons

• EU member states nominate monitoring laboratories to participate in European comparisons (Treaty obligation)

• IRMM provides comparison samples, carrying reference values traceable to SI and SIR

• example: in anticipation of a new European directive on drinking water quality, IRMM organised a water comparison to see where monitoring laboratories stand.

Metrology approach to being correct and accepted by other laboratories

• allows realistic estimate of accuracy under routine conditions

• reliable monitoring results are necessary to assess the exposure of the population as a whole (done by DG ENER of the European Commission)
Key comparisons and traceability

Key Comparisons

IRMM

NMIs world-wide

Nat. Calibr. Service

CIPM/BIPM

K-C-Ref-Value
input ⇒ SIR
≈ 60 Rad.Nucl.

IRMM Ref-Value

Monitoring labs

REM intercomparison

Hospitals

Industry
Comparison results mineral water

Lab means* compared with reference value $A_{ref} \pm U_{ref}$

$^{226}$Ra - W1

Laboratory Activity concentration / mBq.L$^{-1}$

* 41 submitted results
Comparison results mineral water

Lab means* compared with reference value

$^{238}\text{U} - \text{W1}$

Activity concentration / mBq.L$^{-1}$

32 labs $< \pm 20\%$

4 labs $>2\times$

40 submitted results
Not always necessary for obtaining the lowest detection limits

Much better control of background components than above ground ⇒ more robust measurements ⇒ Important for better QC of reference samples.
...a growing field of science, engineering and metrology
Thank you for your attention!
EXTRA SLIDES