Possibilities and limitations in low level gamma ray spectrometry for solid NORM samples

Mikael Hult

www.jrc.ec.europa.eu

Serving society
Stimulating innovation
Supporting legislation

Dec. 4, 2012, EAN NORM, Dresden
The EU Institutions

- Court of Auditors
- Court of Justice
- European Parliament
- The Council of Ministers
- Committee of the Regions
- Economic and Social Committee
- The European Commission (the ‘College’ of Commissioners)

Directorates General: the “Commission services”

JRC Institutes:
- IHCP
- IPSC
- IPTS
- IRMM
The **JRC** is a Directorate-General (DG) of the European Commission

Founded under EURATOM treaty 1957

The mission of the **JRC** is to provide customer-driven scientific and technical support for the conception, development, implementation and monitoring of **EU policies**. As a service of the European Commission, the JRC functions as a **reference centre of science and technology for the Union**. Close to the policy-making process, it serves the common interest of the Member States, while being independent of special interests, whether private or national.

**IRMM =**

Institute for Reference Materials and Measurements

**Mission:** To promote a common and reliable European measurement system in support of EU policies
5 km

Formerly CBNM – Central Bureau for Nuclear Measurements

- Umicore
- Belgonuclear
- FBFC
- IRMM
- SCK•CEN + VITO (+ HADES)
- BR1 (700 kW) + BR2 (10 MW)
- Belgoprocess
- Canberra Semiconductor
Dec. 4, 2012, EAN NORM, Dresden

Method development and standardisations (SIR, KC,..)

Decay Data for stakeholders (IAEA, NEA,..)

Standardisation via ICRM and IAEA

Comparability and quality of NMI:s and field laboratories

RADMET AL: Uwe Wätjen

Stefaan Pommé
Primary Standardisation

Uwe Wätjen
Intercomparisons

Mikael Hult
Low-level Radioactivity

Ref. Mat., LSC, GS, Rad.Chem.

Standardisation and development via CELLAR
IRMM worked/works a lot within ICRM (International Committee for Radionuclide Metrology)

Conferences every 2 years:
...
2005: Oxford
2007: Cape Town
2009: Bratislava
2011: Tsukuba
2013: Antwerp
2015: _____

ICRM Low-level Working Group organises Low-level conferences every 4 years
...
1999: Mol
2003: Vienna
2008: Braunschweig
2012: Jeju
2016:_______
19th International Conference on Radionuclide Metrology and its Applications

First Announcement & Call for Papers

ICRM 2013
17 - 21 June 2013
Antwerp, Belgium

Organized by:
The International Committee for Radionuclide Metrology (ICRM)
European Commission - Joint Research Centre
Institute for Reference Materials and Measurements (EC-JRC-IRMM)

Proceedings published in a special issue of Applied Radiation and Isotopes

Key Projects 2012

**EMRP – MetroFission**
Metrology for new generation nuclear power plants
Sept 2010 – Sept 2013

**EMRP – MetroRWM**
Metrology for Radioactive Waste Management

**EMRP – MetroMetal**
Ionizing Radiation Metrology for Metallurgical Industry
Dec 2011 – Dec 2014

**EMIT**
Europe and Metrology in Turkey (DG ELARG)
Funded by DG ELARG (AA)

**IAEA-CRP**
Benchmarking Calibration for Low-level Gamma Spectrometric Measurements of Environmental Samples
2009 – 2013

**PT support REM**
Radioactive Environmental Monitoring
On direct request from DG ENER (MoU JRC-DG ENER)
Since 2003 and will probably run as long as the EURATOM treaty is valid

**Work for ITRAP +10**
Illicit Trafficking Radiation detection Assessment Programme
Direct support to DG HOME (AA)

**Fukushima support**
Ultra Low-level Radioactivity Measurements of Pacific Sea Water (DG MARE)
<table>
<thead>
<tr>
<th>Year</th>
<th>Sample</th>
<th>Radioisotopes</th>
</tr>
</thead>
<tbody>
<tr>
<td>2003</td>
<td>Air filter</td>
<td>$^{137}$Cs</td>
</tr>
<tr>
<td>2005</td>
<td>Milk powder</td>
<td>$^{134/137}$Cs, $^{40}$K, $^{90}$Sr</td>
</tr>
<tr>
<td>2008</td>
<td>Water</td>
<td>$^{238/234}$U, $^{226/228}$Ra</td>
</tr>
<tr>
<td>2010</td>
<td>Soil</td>
<td>$^{40}$K, $^{137}$Cs, $^{212/214}$Bi, $^{212/214}$Pb, $^{226}$Ra, $^{230/232}$Th, $^{234/235/238}$U, $^{238/239/240}$Pu, $^{90}$Sr</td>
</tr>
<tr>
<td>2011</td>
<td>Bilberry</td>
<td>$^{90}$Sr, $^{137}$Cs, $^{40}$K</td>
</tr>
<tr>
<td>2012</td>
<td>Water</td>
<td>Gross alpha/beta activity</td>
</tr>
<tr>
<td>2013/2014</td>
<td>New Air filter</td>
<td>$^{137}$Cs+...?</td>
</tr>
</tbody>
</table>
International comparisons for field laboratories

Organizing comparisons for laboratories monitoring environmental radioactivity in the member states and neighbouring countries of the EU

→ see example of results:

Evaluation completed: $^{137}\text{Cs}, ^{40}\text{K}, ^{90}\text{Sr}$ in bilberry powder; 88 labs, comparison report being drafted, completion by end 2012

Comparison in execution: gross alpha/beta activity in water
70 results

Relative deviations:
• 89% of results within 20% from reference

$E_n$ numbers:
• 72% compatible (50 labs)
• 11% warning signal
• 17% action signal

Inappropriate calibration/Reference material???
38 results
Relative deviations:
• 26% of results within 20% from reference value

\( E_n \) numbers:
• 42% compatible
• 16% warning signal
• 42% action signal
From recent ILC: radioactivity in mineral water

- In anticipation of new European requirements for monitoring radioactivity in drinking water (COM(2012)147final), IRMM benchmarked labs determining low concentrations of natural radioactivity in mineral waters.
- 14% of all radium results are off by a factor of two or more.
- The comparison clearly demonstrates that a number of monitoring labs need to improve their analysis procedures for radium in order to correctly identify drinking water sources for which remedial action is necessary.
1. Effective legislation depends on accurate measurements
2. IRMM provides the tools to measure properly and in a harmonised way
Reference data in policy-relevant domains

Participation in EMRP (Art. 169) projects:

- **JRP MetroFission** (Metrology for new generation nuclear power plants):
  IRMM tasks: neutron metrology and decay data ($^{238}\text{U}$)

- **JRP MetroRWM** (Metrology for radioactive waste management):
  IRMM tasks: development of reference materials for free release systems,
  improved half-lives of waste-relevant radionuclides

- **JRP MetroMetal** (Ionizing radiation metrology for the metallurgical industry):
  IRMM tasks: characterisation of reference materials, MC simulations, comparisons

- Member of consortium for proposed **JRP MetroNORM**

**EU nuclear safety standards (BSS) require actions on NORM**
Metrology for processing materials with high natural radioactivity

JRP-Coordinator: Franz-Josef Maringer, BEV/PTP (Austria)

<table>
<thead>
<tr>
<th>WP No</th>
<th>Work Package Name</th>
<th>Active JRP-Participants (WP leader in bold)</th>
</tr>
</thead>
<tbody>
<tr>
<td>WP1</td>
<td>Reference materials and standard sources</td>
<td>CMI, BEV/PTP, CEA, CIEMAT, ENEA, IST,</td>
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<td></td>
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<td>JRC, MKEH, NPL, STUK, REG(SURO)</td>
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<tr>
<td>WP2</td>
<td>Design of measurement systems</td>
<td>NPL, BEV/PTP, CEA, CMI, ENEA, IJS, STUK,</td>
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<td></td>
<td></td>
<td>REG(SURO)</td>
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<tr>
<td>WP3</td>
<td>Development of measurement procedures</td>
<td>JRC, BEV/PTP, CIEMAT, CMI, ENEA, IJS, IST,</td>
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<tr>
<td></td>
<td></td>
<td>MKEH, NPL, NRPA, STUK, REG(BOKU), REG(SURO)</td>
</tr>
<tr>
<td>WP4</td>
<td>Improvement of NORM related data</td>
<td>CEA, BEV/PTP, CIEMAT, CMI, ENEA, IST,</td>
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<td></td>
<td></td>
<td>JRC, MKEH, NPL, REG(BOKU)</td>
</tr>
<tr>
<td>WP5</td>
<td>On-site testing</td>
<td>IJS, BEV/PTP, CMI, ENEA, IST, JRC, NPL,</td>
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<td></td>
<td></td>
<td>NRPA, STUK, REG(BOKU)</td>
</tr>
<tr>
<td>WP6</td>
<td>Creating Impact</td>
<td>ENEA, all partners</td>
</tr>
<tr>
<td>WP7</td>
<td>Management and Coordination</td>
<td>BEV/PTP, all partners</td>
</tr>
</tbody>
</table>
Radionuclide metrology laboratory of IRMM

Primary standardisation laboratory of radioactivity

$4\pi \beta-\gamma$ coincidence counting systems

$4\pi \gamma$ counting

$4\pi \beta-\gamma$ sum counting

$4\pi e^-, \beta, \gamma$, X-ray counting (unique CsI sandwich detector)

defined solid angle alpha-particle counting

liquid scintillation counting:
- CIEMAT/NIST method
- TDCR method
Radionuclide metrology laboratory of IRMM

Secondary standardisation laboratory
  ionisation chambers
  gamma-ray spectrometry
  radiochemistry laboratory
  in the underground low-level radioactivity laboratory HADES:
  gamma-ray spectrometry with detection limits of the order of mBq/kg
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
The Sandwich Spectrometer

Increased solid angle

Ge-7

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

+14.5 cm lead, 20 Bq/kg

Cu lining = radiopure copper, 3.5 cm

Ge-6

Detector mass ~ 1.9 kg each
The “Sandwich” spectrometer

- Increased solid angle
- Doubled FEP efficiency (compared to single HPGe)
- $\mu$ contribution to Bkg reduced by $\sim 30\%$ thanks to the plastic scintillators
Detector shielding

- Minimised empty space inside the shielding
- Nitrogen flushed inside the shielding
- Dust covers

15-20 cm Pb of which the inner 2-5 cm low in $^{210}\text{Pb}$ (< 3 Bq/kg)

Lead from several hundreds years old buildings in Gent
Background Comparison – Gamma-ray spectrometry

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

- 511 keV
- 2614 keV
- 1460 keV
- Ge(n,n'γ)
- Pb(n,n'γ)

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

Energy (keV)
HADES = 0.05 muons/m²s

10 HPGe detectors
(soon 11, maybe 12)
+3 NaI + 4 PS
Above ground

Spoon #11-2, @ 280 m Tokai-mura

320 keV ($^{51}$Cr)

1173 keV ($^{60}$Co)

1332 keV ($^{60}$Co)

59Fe

HADES Background

Gamma-ray energy [keV] Normalised countrate [min$^{-1}$ keV$^{-1}$ kg$^{-1}$]
Dec. 4, 2012, EAN NORM, Dresden

- Count rate 40-2700 keV \times 0.01 / d^{-1}
- Net count rate at 609 keV / d^{-1}
- Radon concentration / Bq·m^{-2}

Time / Measurement No.
Dec. 4, 2012, EAN NORM, Dresden

(a) Pacman shield
(b) In Sandwich shield

Time / Spectrum number

Count rate 40-2700 keV / d⁻¹

- ~ 3 years
- ~ 1 week per data point
- ~1 day per data point
A synergetic process...

IRMM - Institute for Reference Materials and Measurements

Mission: to promote a common and reliable European measurement system in support of EU policies

Research

ULGS - Ultra Low-level γ-ray Spectrometry

Fundamental physics

Search for rare events

Policy

Outcome: technology transfer, e.g., radiation protection, EU policies
Problems manifest themselves in numerous situations

• Decay data (see e.g. $^{234}\text{Th}$)

• Intercomparisons; more scatter at lower energies (e.g. $^{210}\text{Pb}$, $^{109}\text{Cd}$, $^{235}\text{U}$, $^{142}\text{Am}$, … also $^{133}\text{Ba}$ and $^{152}\text{Eu}$, …)

• Monte Carlo simulations attempting to reproduce measured efficiency (Exemplify w.b.)

• ……

• => part of an ongoing IAEA-CRP. Major review of low-energy gamma-ray spec. in pipe line.
Detector response of 92.5 keV

Useful with Monte Carlo simulation, can “isolate” contributions
Compton Edge and Backscatter peak as a function of incident gamma-ray energy

- $E_{\gamma}$ (keV)
- Compton Edge (keV)
- Backscatter Peak (keV)
4 gamma-rays: 46.5, 59.5, 63 and 92.5 keV
Problems with thin deadlayers

- Beta particles can reach the sensitive volume
- Higher background at low energy
- Coincidence summing with X-rays
Focus on $^{238}\text{U}$

- Gamma-ray spectrometry not the best technique to quantify U-238!

- Still, gamma-ray spectrometry often used since one can get results for many radionuclides in one analysis.

- Sometimes, gamma-ray spectrometry is dangerously simple to use.

- There is no data analysis software that does “it all” for you.

- It is still necessary with some hard work and know-how to obtain robust results and good quality data.
$^{238}$U decay chain

by courtesy of Dr. G. Heusser

- Suitable for gamma spec: 767 keV, 0.3%; 1001 keV, 0.8%
- 63 keV, 3.75(8)% (2% rel unc.)
- 92.5 keV, 4.33(27)% (6% rel unc.)
Decay data – well known?

<table>
<thead>
<tr>
<th>Reported value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.8 (6)%</td>
<td>Nucléide - 2000</td>
</tr>
<tr>
<td>4.80%</td>
<td>Mini Table de Radionucléides, 2007</td>
</tr>
<tr>
<td>4.49%</td>
<td>Genie-2000</td>
</tr>
<tr>
<td>4.1 (7)%</td>
<td>αβγ-Table, Wahl</td>
</tr>
<tr>
<td>4.1 (7)%</td>
<td>PTB-bericht 1998</td>
</tr>
<tr>
<td>4.00 (6)%</td>
<td>Nuclides2000</td>
</tr>
<tr>
<td>3.75 (8)%</td>
<td>DDEP - 2009</td>
</tr>
<tr>
<td>3.7 (2)%</td>
<td>The Radiochemical Manual (1988)</td>
</tr>
<tr>
<td>3.7 (4)%</td>
<td>NNDC</td>
</tr>
<tr>
<td>3.69 (7)%</td>
<td>NDS - 2007</td>
</tr>
<tr>
<td>3.6 (1)%</td>
<td>PTB-Ra-16/3, 1989</td>
</tr>
</tbody>
</table>

Std.dev: 0.45
Rel Std. dev. 11%
(Max-min)/average: 30%
More Problems / optimisation

- Doublets (both 63 keV and 92.5 keV) ⇒ broad peaks
- Suitable detector – size, deadlayer thickness
  - Resolution,
  - Amplifications (also in simulations)
  - background,
  - efficiency
  ⇒ Use “All purpose detectors” with care
More Problems / optimisation

- Optimising sample size and geometry
- Subtraction of interfering peaks
  - (93.3 keV Th K\(_{\alpha1}\) X-ray – mainly from \(^{228}\)Ac – also \(^{235}\)U and \(^{238}\)U)
- Reference Materials (Reference value? Stable? Hot spots?)
- Efficiency Transfer, Monte Carlo simulations
  - Accuracy of model, bin-width, coincidences, algorithm at low-E?
- Extrapolation of efficiency curve
- Eff. Curve coincidence summing corrections
- Background – variations of cosmic rays, radon, contamination (detector, shield, sample), nearby activities

Interference free detection limit in HADES of pure U-sample in swipe sample ~ 1 ng
Sample:
Density 1.3 g/cm$^3$
Dried soil
Sample:
Density 1.5 g/cm³
Dried soil
Detector: 50% rel. eff. BEGe

Count rate (d⁻¹) vs. Sample thickness (mm) for different energies:
- 63.2 keV
- 92.5 keV
- 186 keV
- 1001 keV

Detector: 50 mm
Thank you for your attention!