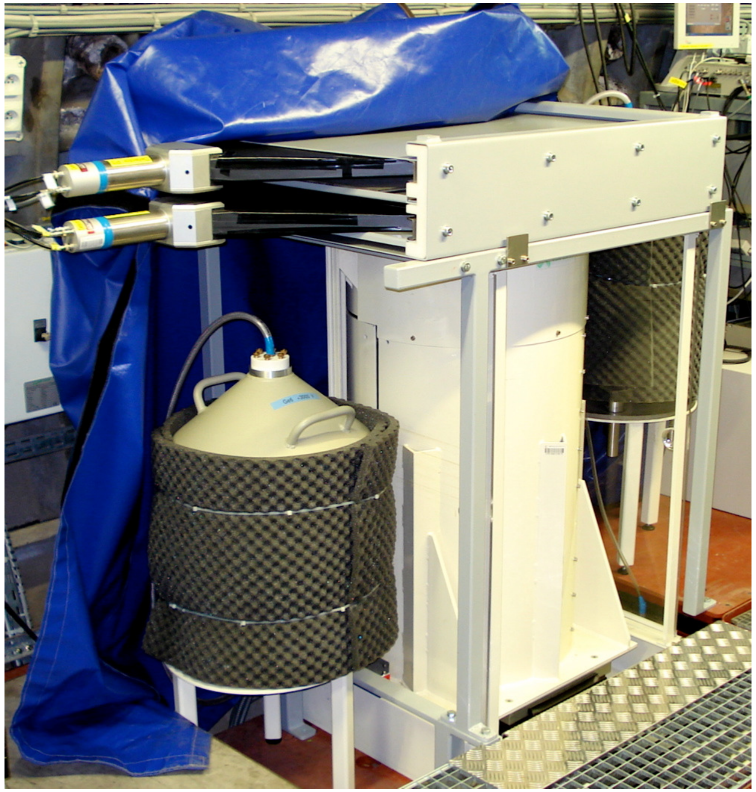
European Commission

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



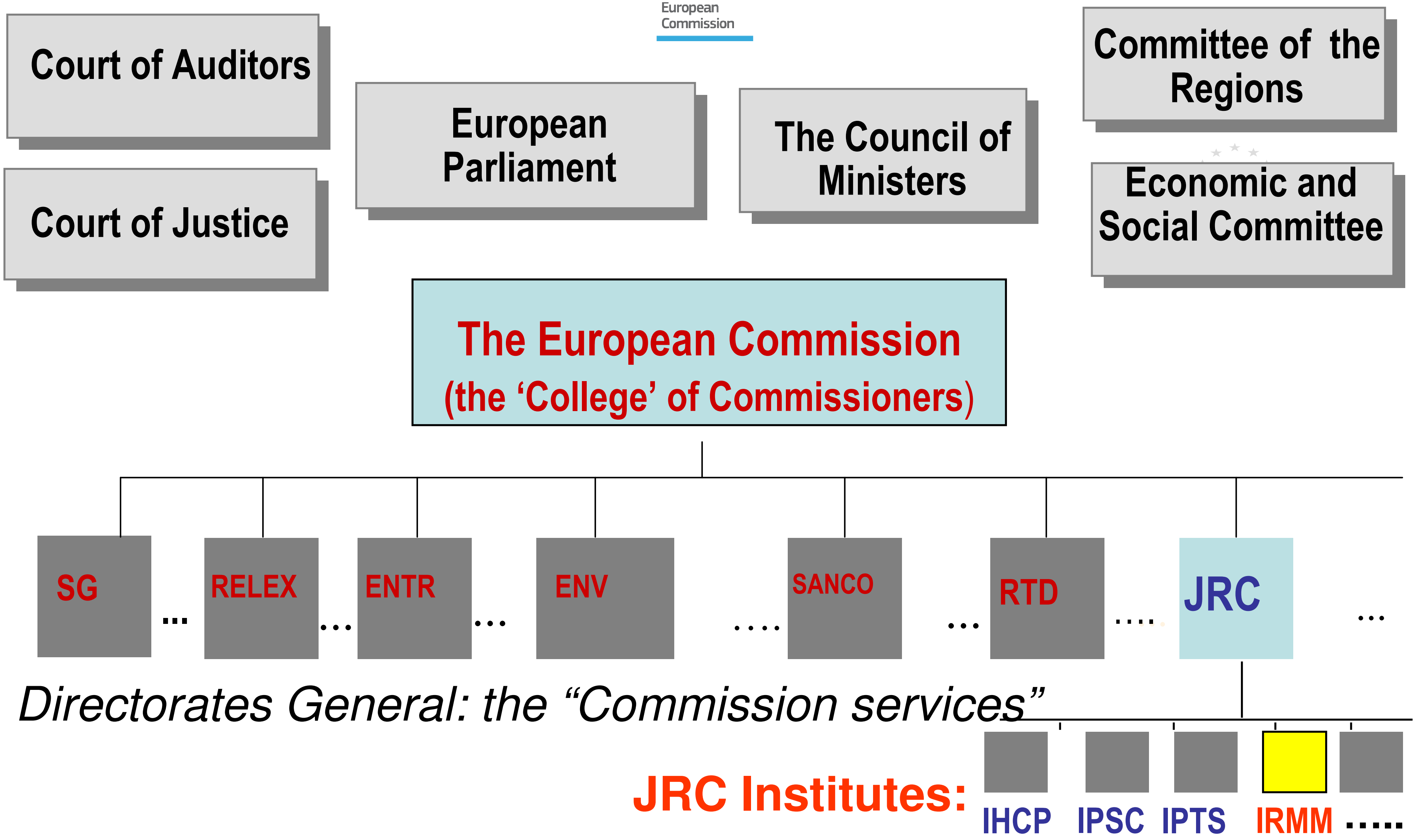
**Mikael Hult**

**www.jrc.ec.europa.eu**

*Serving society*
*Stimulating innovation*
*Supporting legislation*

# The EU Institutions

European Commission






Joint Research Centre

# Joint Research Center

European Commission

***IRMM =***

**Institute for Reference Materials and Measurements**

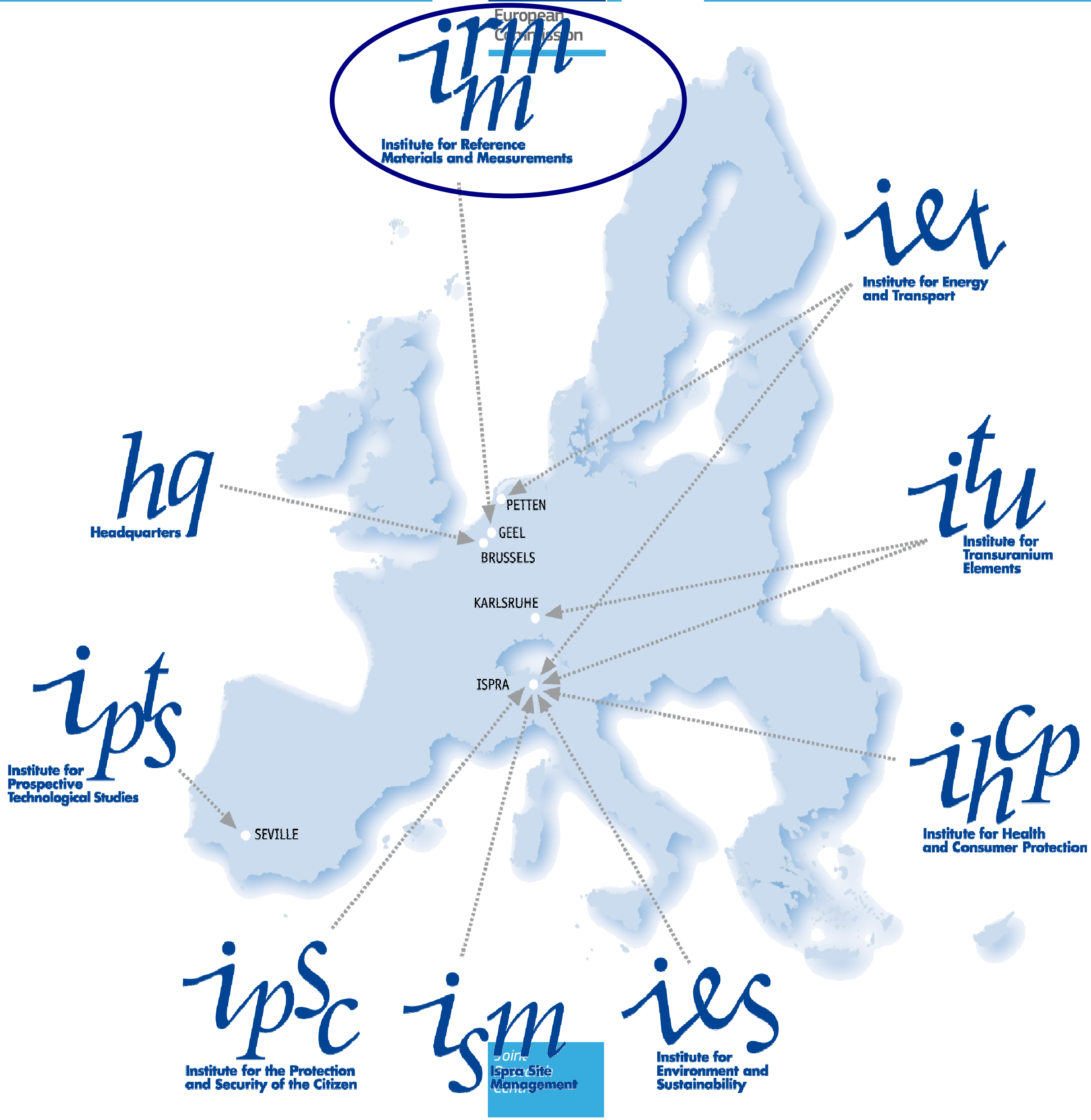
Mission: To promote a common and reliable European measurement system in support of EU policies

## 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.*

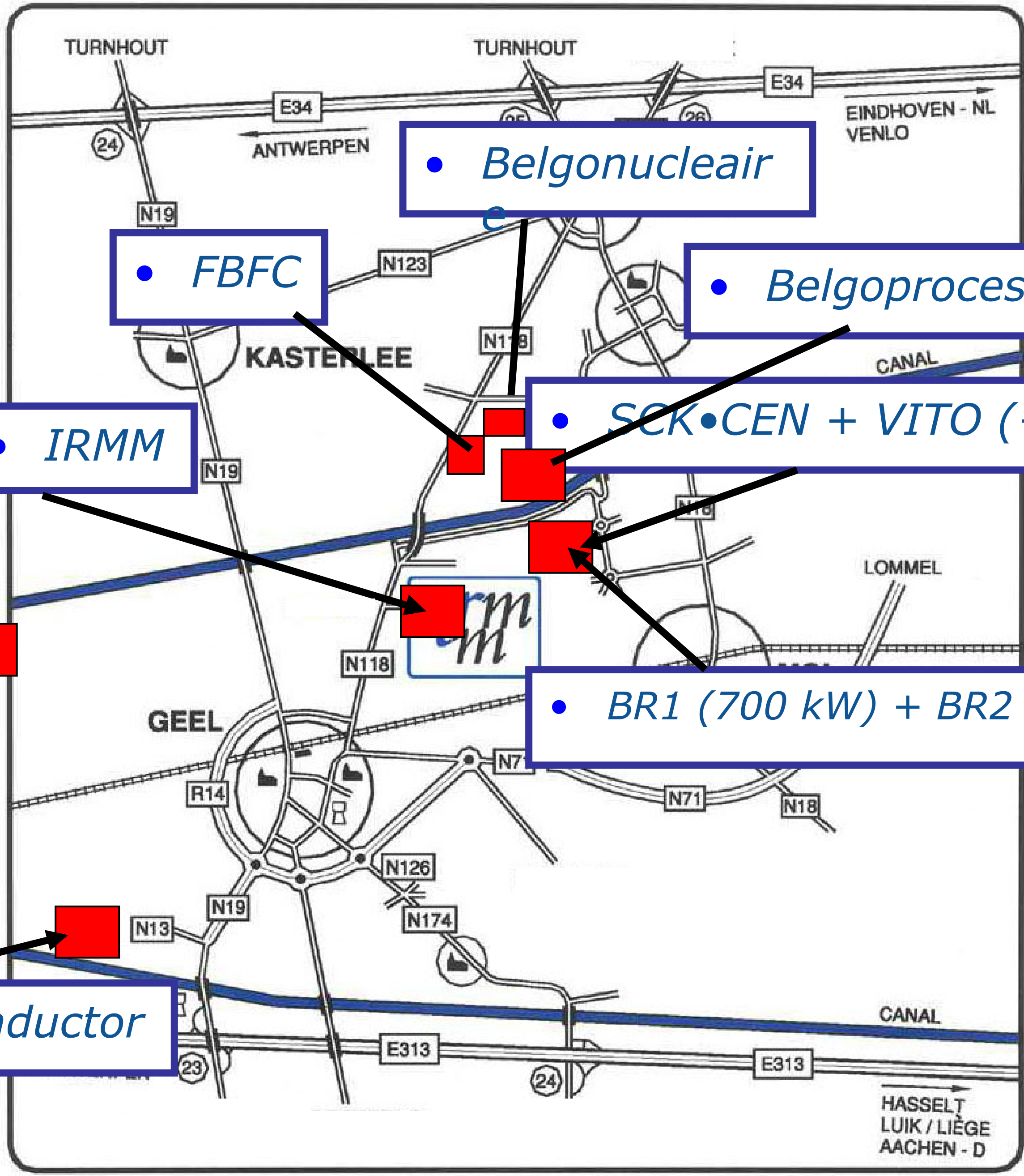
# Joint Research Center

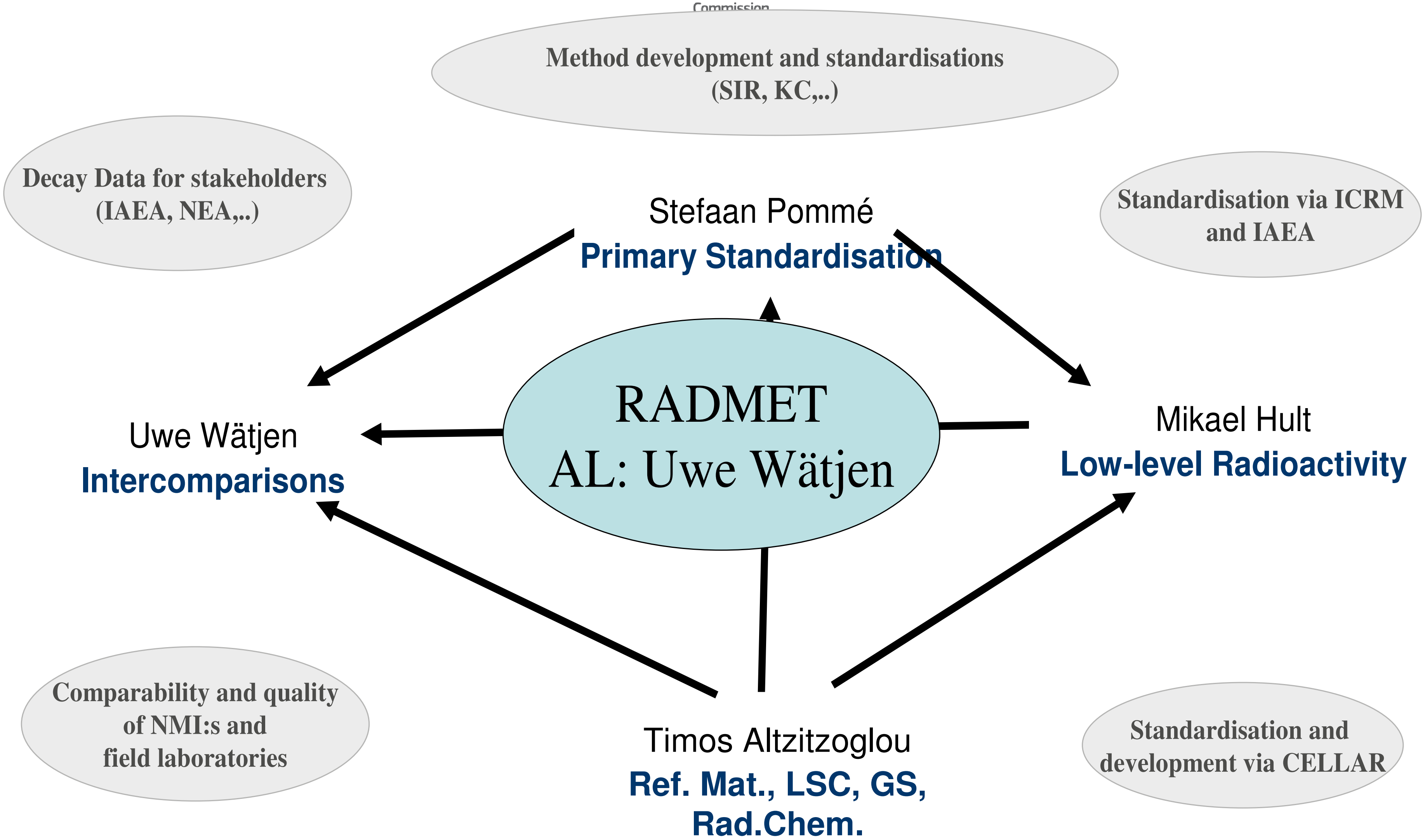


**5 km**

Formerly CBNM – Central Bureau for Nuclear Measurements

- Belgonucleaire
- FBFC
- Belgoprocess
- SCK•CEN + VITO (+ HADES)
- IRMM
- Umicore
- BR1 (700 kW) + BR2 (10 MW)
- Canberra Semiconductor




Method development and standardisations
(SIR, KC,..)
Decay Data for stakeholders
(IAEA, NEA,..)
Stefaan Pommé
Primary Standardisation
Standardisation via ICRM
and IAEA
RADMET
AL: Uwe Wätjen
Uwe Wätjen
Intercomparisons
Mikael Hult
Low-level Radioactivity
Comparability and quality
of NMI:s and
field laboratories
Timos Altzitzoglou
Ref. Mat., LSC, GS,
Rad.Chem.
Standardisation and
development via CELLAR

European Commission

# *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:_______

European Commission

# 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)

Joint Research Centre

http://irmm.jrc.ec.europa.eu/icrm2013

Proceedings published in a special issue of Applied Radiation and Isotopes

Joint Research Centre

# Key Projects 2012

European Commission

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

**EMRP – MetroRWM**
Metrology for Radioactive Waste Management
Oct 2011 – Oct 2014

**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)
Oct 2009 – Oct 2012

**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)
2011(2012) – ~2014

# History of ILCs at IRMM



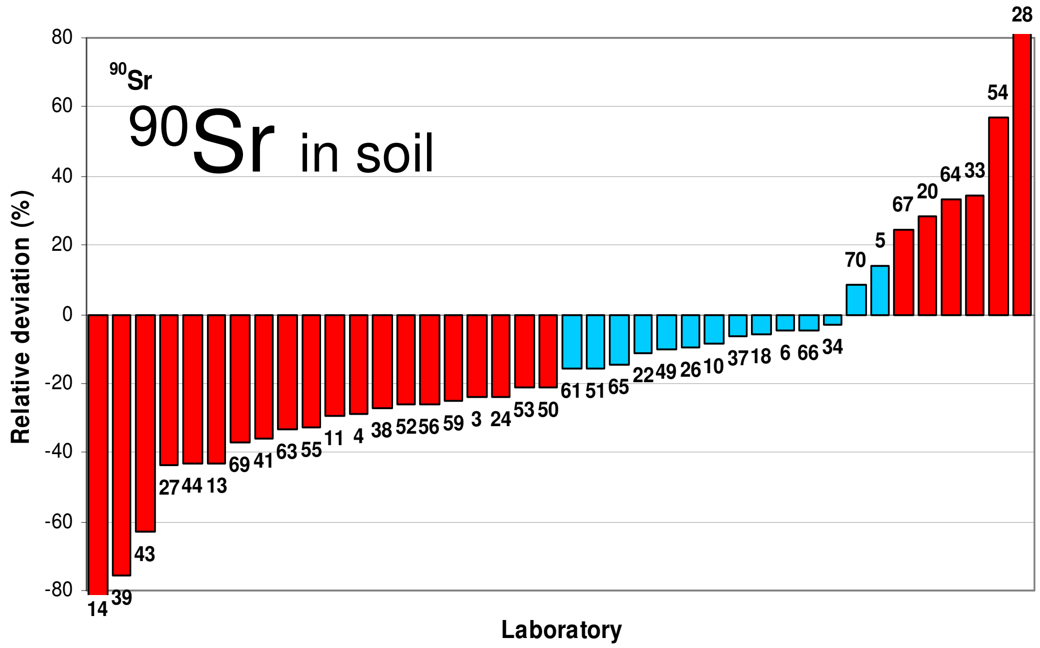
| | | |
|---|---|---|
| 2003 | **Air filter** | **$^{137}Cs$** |
| 2005 | **Milk powder** | **$^{134/137}Cs$, $^{40}K$, $^{90}Sr$** |
| 2008 | **Water** | **$^{238/234}U$, $^{226/228}Ra$** |
| 2010 | **Soil** | **$^{40}K$, $^{137}Cs$, $^{212/214}Bi$, $^{212/214}Pb$, $^{226}Ra$, $^{230/232}Th$, $^{234/235/238}U$, $^{238/239/240}Pu$, $^{90}Sr$** |
| 2011 | **Bilberry** | **$^{90}Sr$, $^{137}Cs$, $^{40}K$** |
| 2012 | Water | Gross alpha/beta activity |
| 2013/2014 (?) | **New Air filter** | **$^{137}Cs$+...?** |



# International comparisons for field laboratories

European Commission

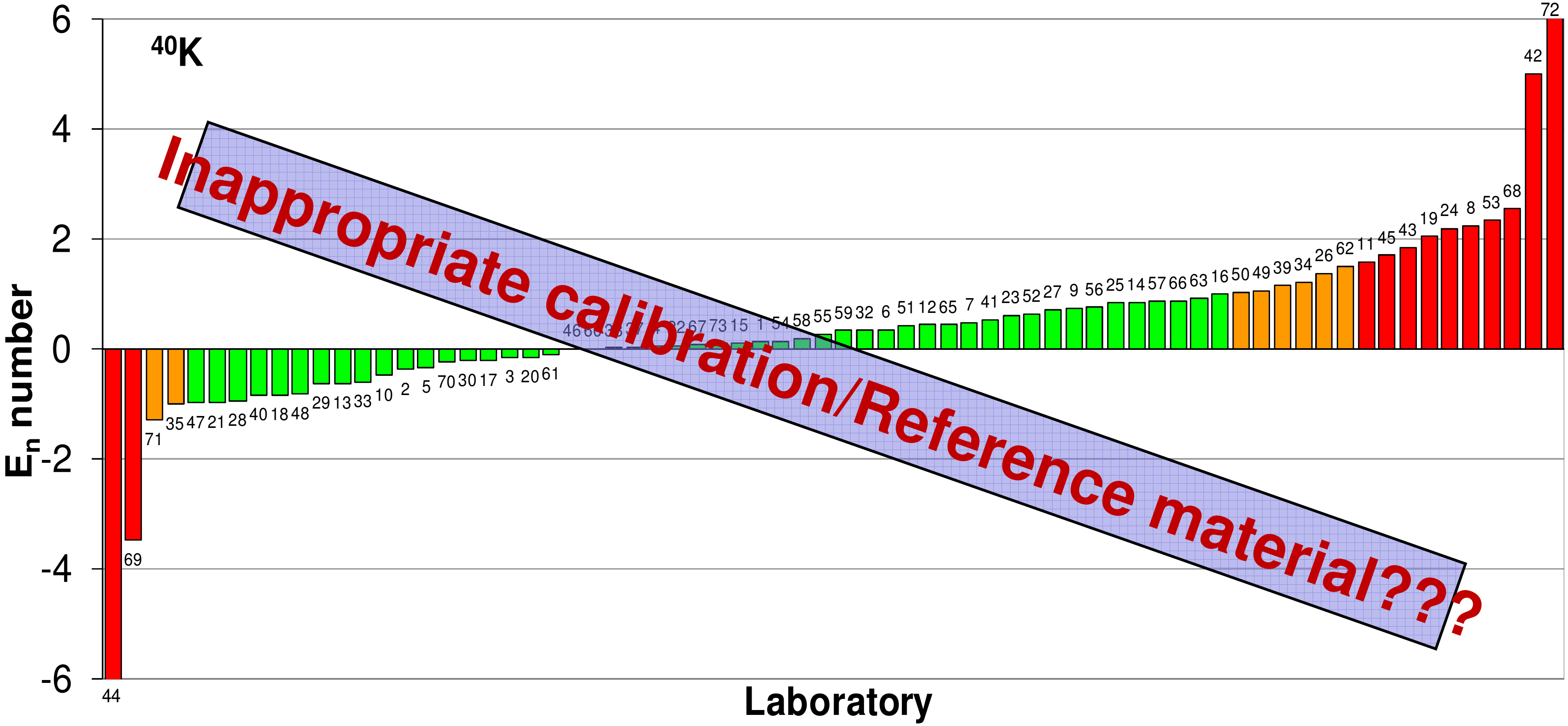
- Organizing comparisons for laboratories monitoring environmental radioactivity in the member states and neighbouring countries of the EU
- → see example of results:




- Evaluation completed: $^{137}$Cs, $^{40}$K, $^{90}$Sr in bilberry powder; 88 labs, comparison report being drafted, completion by end 2012
- Comparison in execution: gross alpha/beta activity in water

Joint Research Centre

# $^{40}K$ in soil intercomparison




70 results

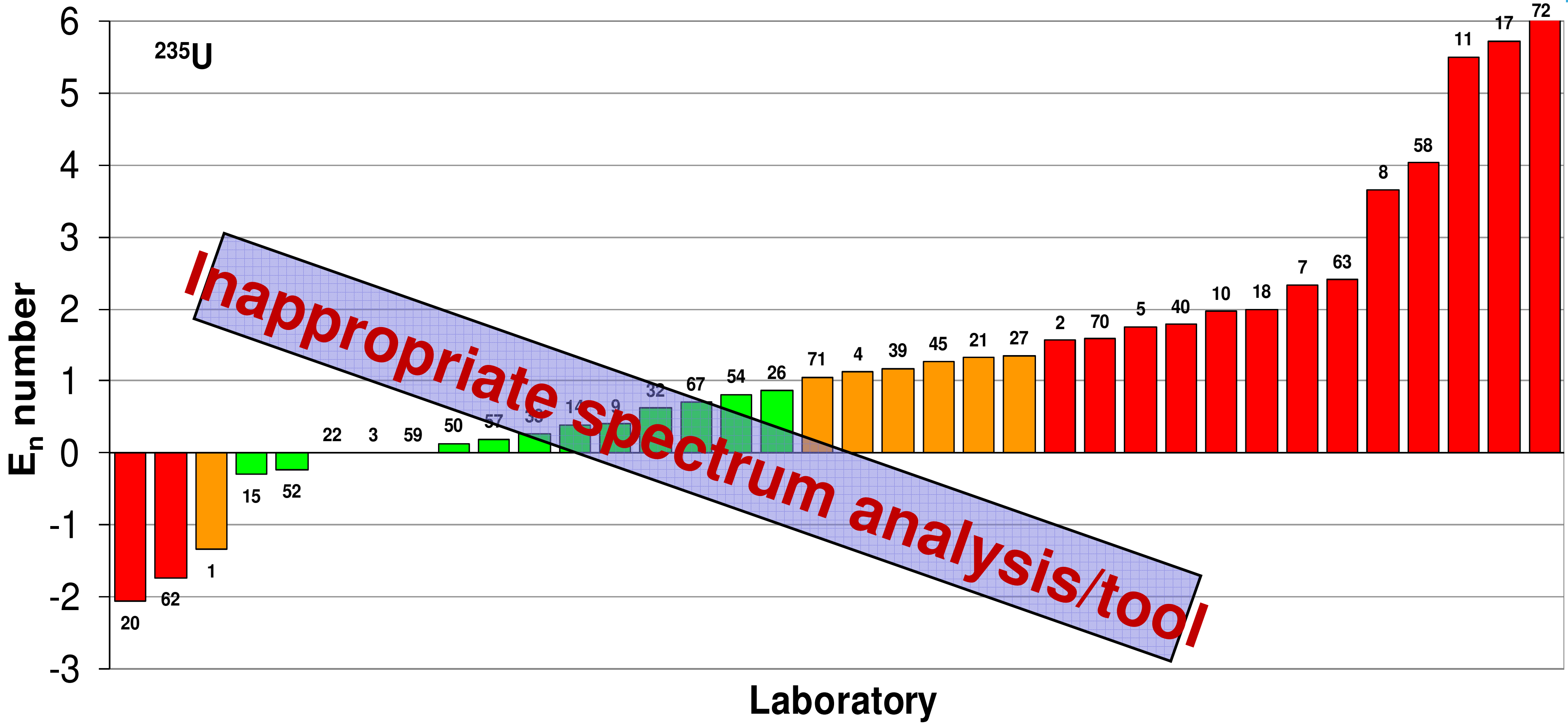
Relative deviations:
- 89% of results within 20% from reference

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



Joint Research Centre

# $^{235}U$ in soil intercomparison




38 results

Relative deviations:

- 26% of results within 20% from reference value

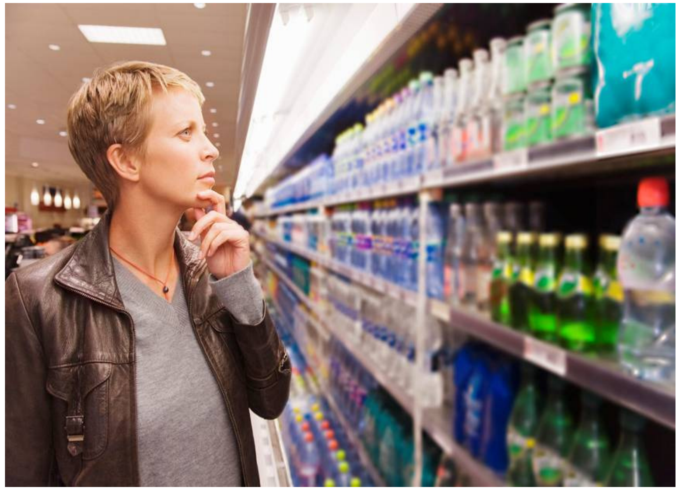
$E_n$ numbers:

- 42% compatible
- 16% warning signal
- 42% action signal

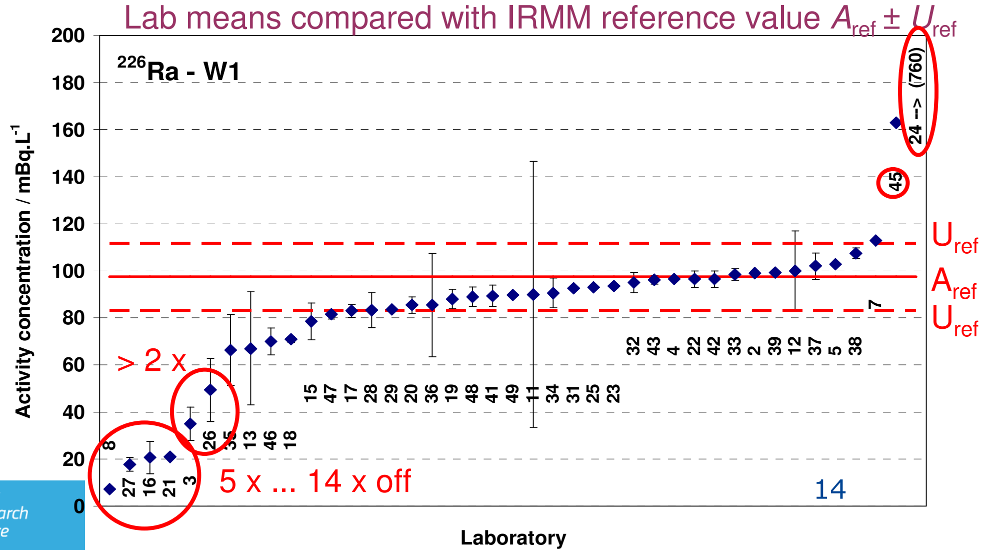
# From recent ILC: radioactivity in mineral water

European Commission

- 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

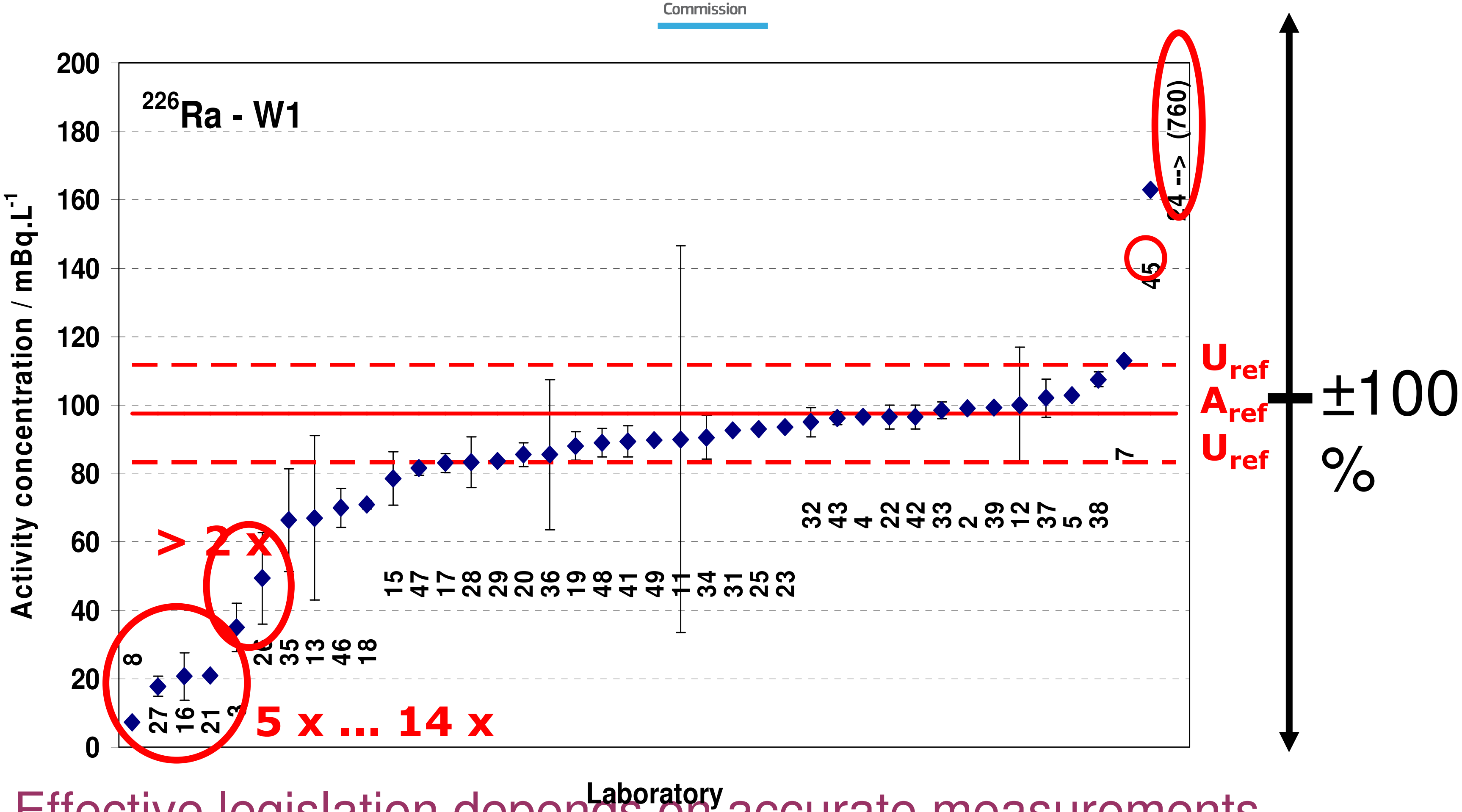


- 14 % of all radium results are off by a factor of two or more
- The comparison clearly demonstrates that a number of monitoring labs need to **<u>improve their analysis procedures</u>** for radium in order to correctly identify drinking water sources for which remedial action is necessary




Joint Research Centre

European Commission




- Effective legislation depends on accurate measurements
- IRMM provides the tools to measure properly and in a harmonised way

# WP objective 4

European Commission

## 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 ($^{238}U$)
  - **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**

# JRP-i13: MetroNORM

European Commission

## Metrology for processing materials with high natural radioactivity

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

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

European
Commission

# Radionuclide metrology laboratory of IRMM

*Primary standardisation laboratory of radioactivity*

*$4\pi\,\beta\text{–}\gamma$ coincidence counting systems*

*$4\pi\gamma$ counting*

*$4\pi\,\beta\text{–}\gamma$ 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**

European Commission

# Radionuclide metrology laboratory of IRMM

*Secondary standardisation laboratory*

- *ionisation chambers*
- *gamma-ray spectrometry*
- *radiochemistry laboratory*
- *in the underground low-level radioactivity laboratory HADES:*

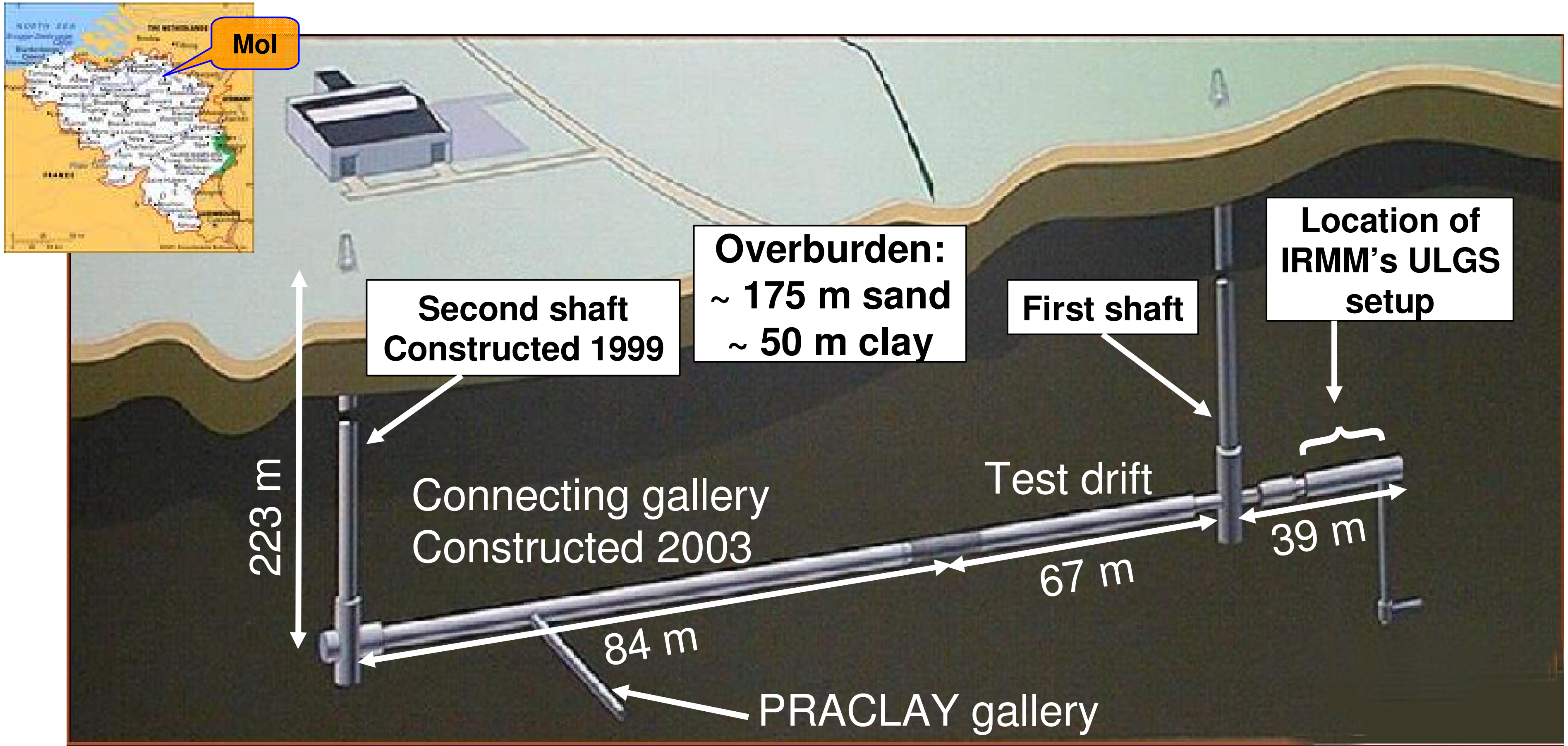
*gamma-ray spectrometry with detection limits of the order of mBq/kg*

# HADES

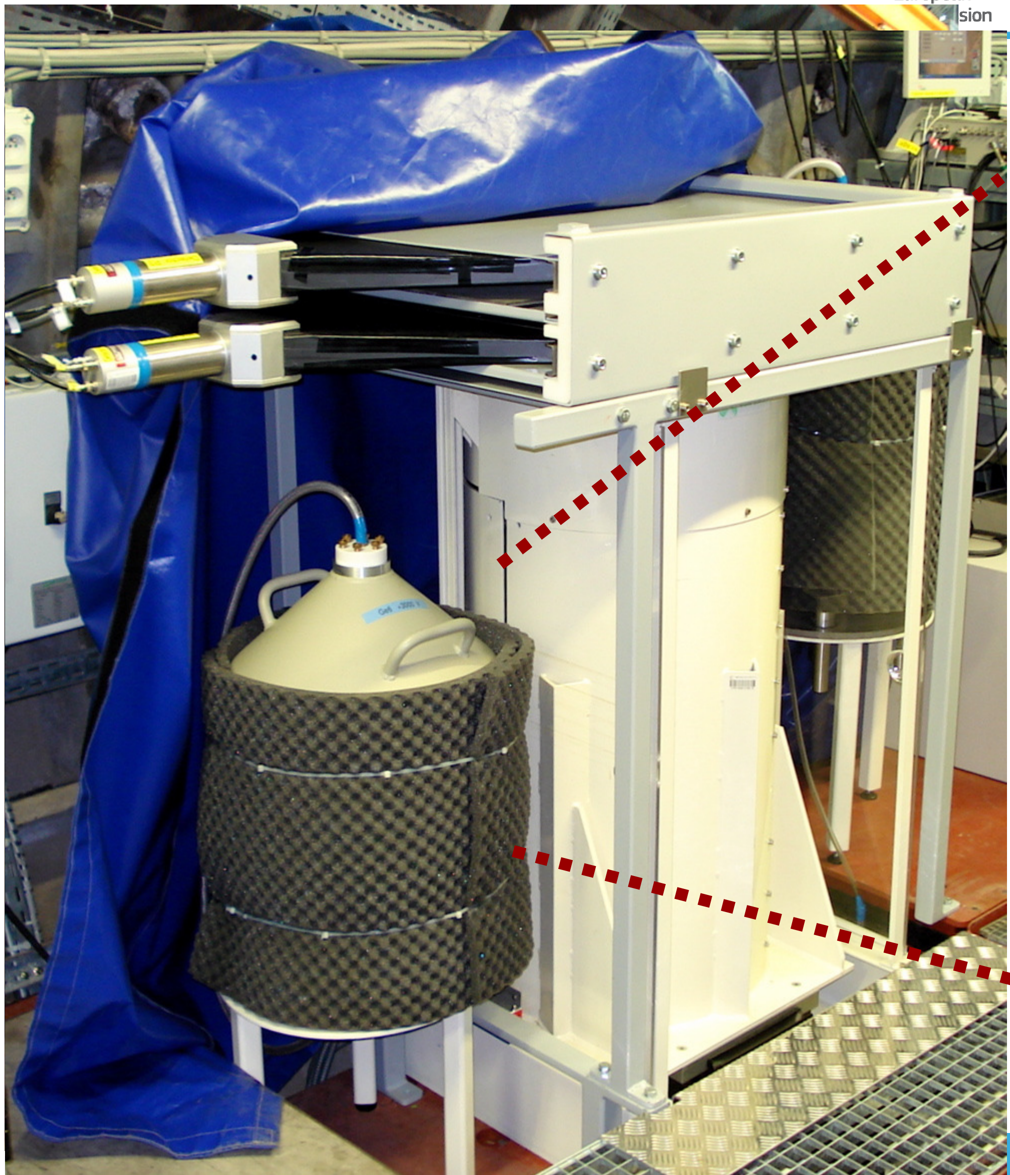


## HADES = High Activity Disposal Experimental Site – Operated by EURIDICE* and located at SCK•CEN in Mol

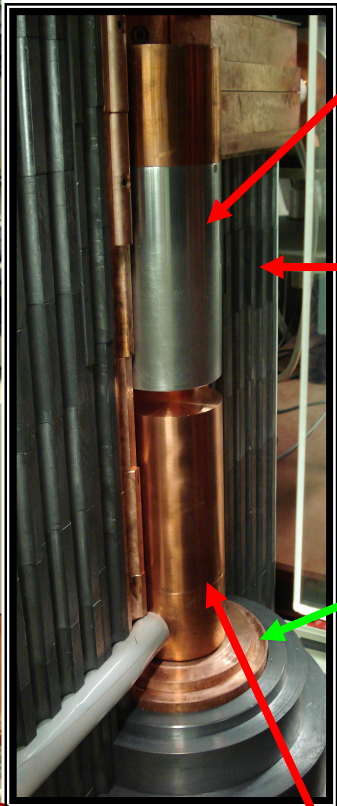
*European Underground Research Infrastructure for Disposal of nuclear waste In Clay Environment



# 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

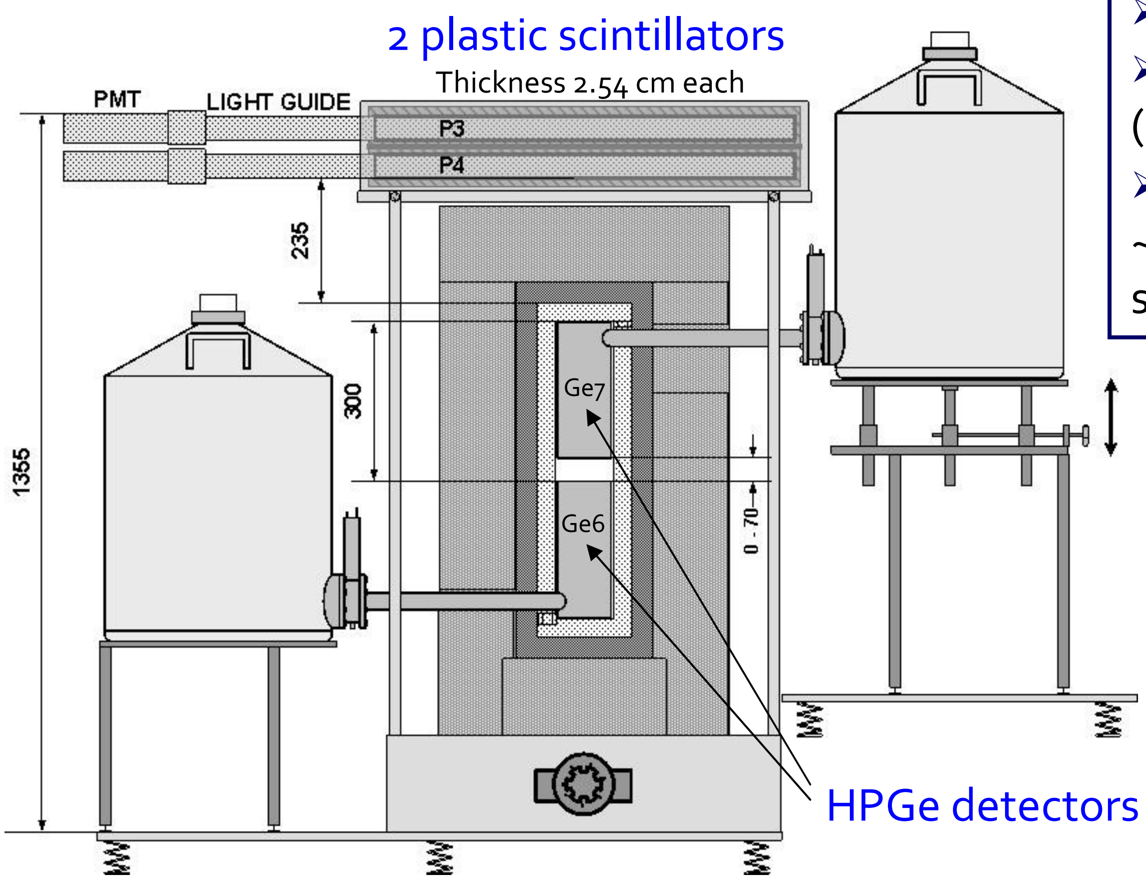
Cu lining = radiopure copper, 3.5 cm

**Ge-6**

**Detector mass ~ 1.9 kg each**

European Commission

# The “Sandwich” spectrometer

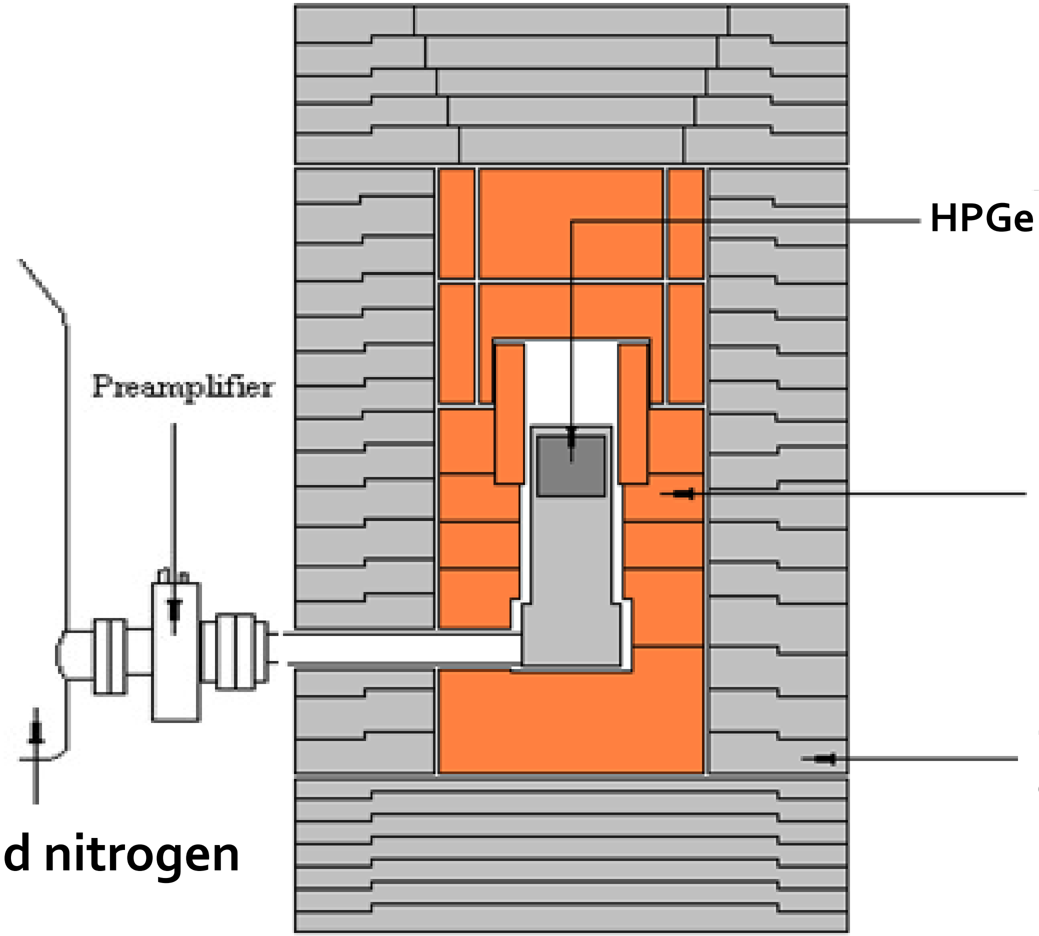



- Increased solid angle
- Doubled FEP efficiency (compared to single HPGe)
- μ contribution to Bkg reduced by ~30% thanks to the plastic scintillators

European Commission

# Detector shielding

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



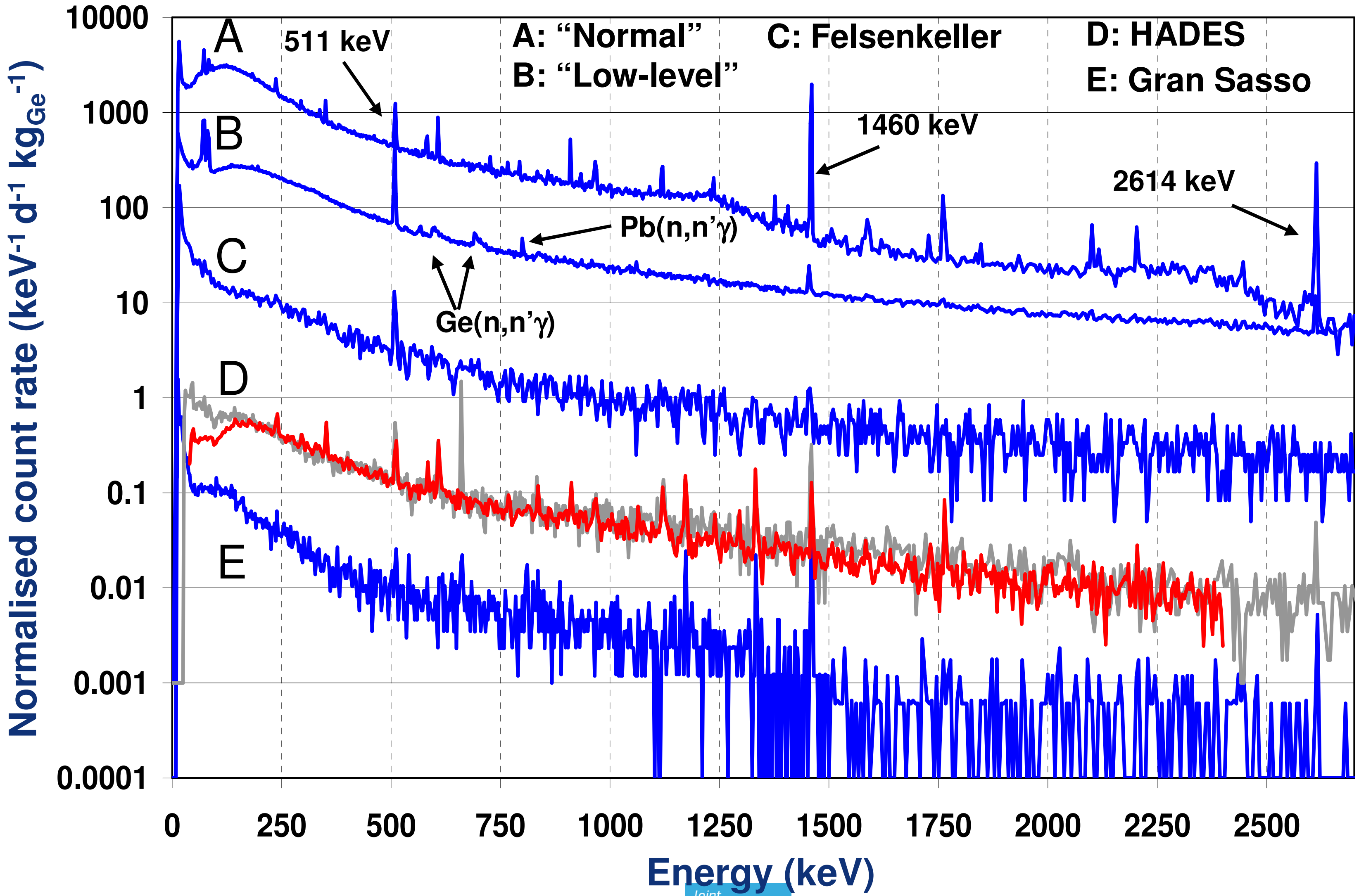

Liquid nitrogen


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

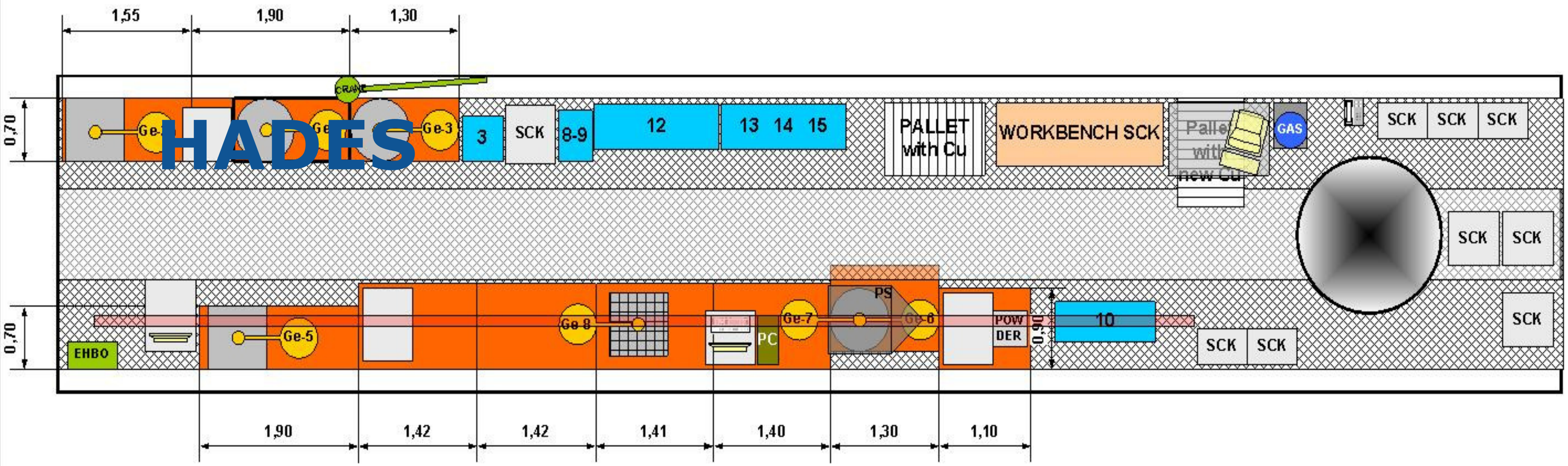
Joint Research Centre

# Background Comparison – Gamma-ray spectrometry

European Commission




Joint Research Centre




**HADES= 0.05 muons/m²s**

| | |
|---|---|
| | Detector |
| | Detectorshield |
| | Cupboard |
| | Electronics rack |
| | Platform |
| PS | Plastic Scintillator |
| UPS | Battery |

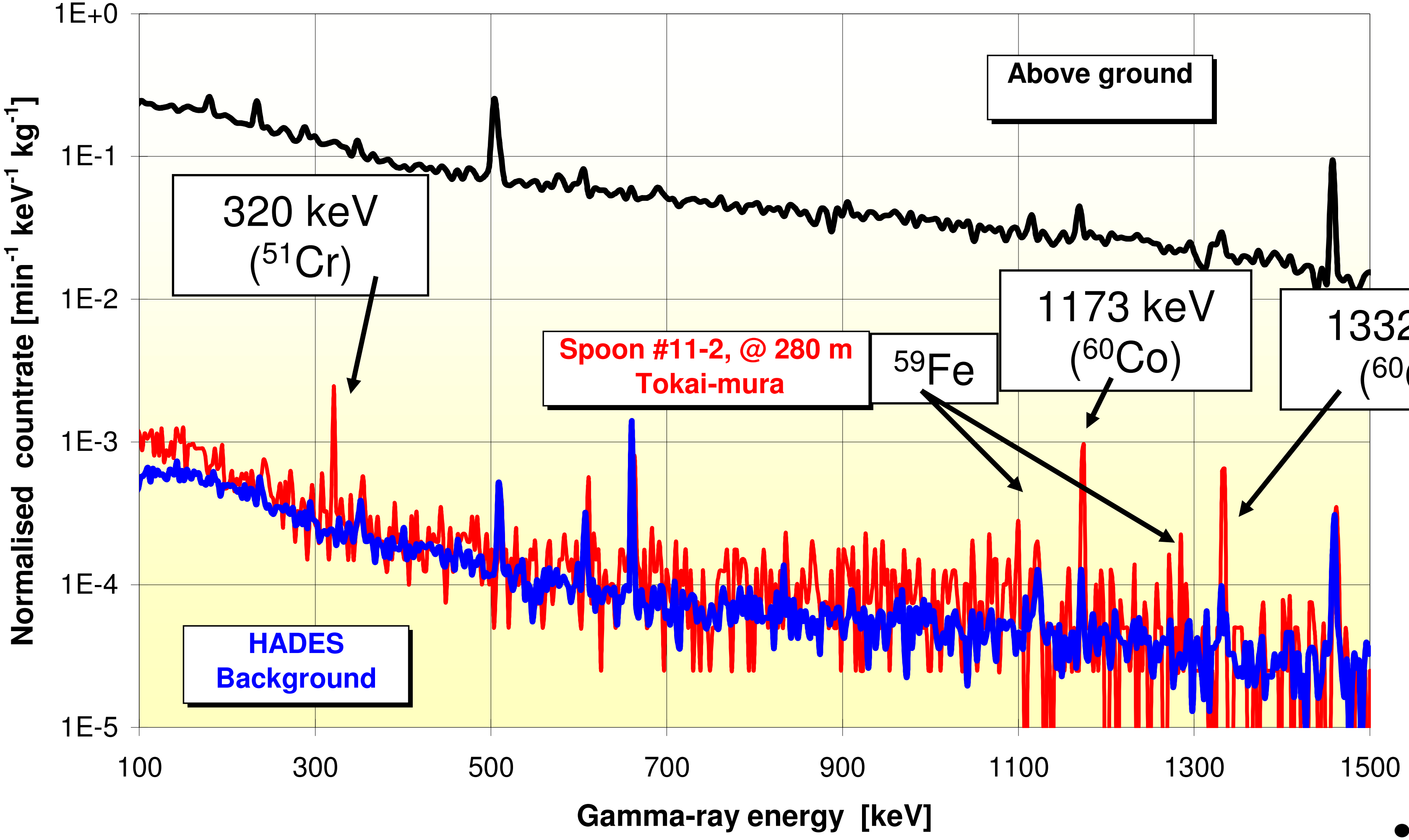
SCALE: 1/50
1.0 m

**10 HPGe detectors**

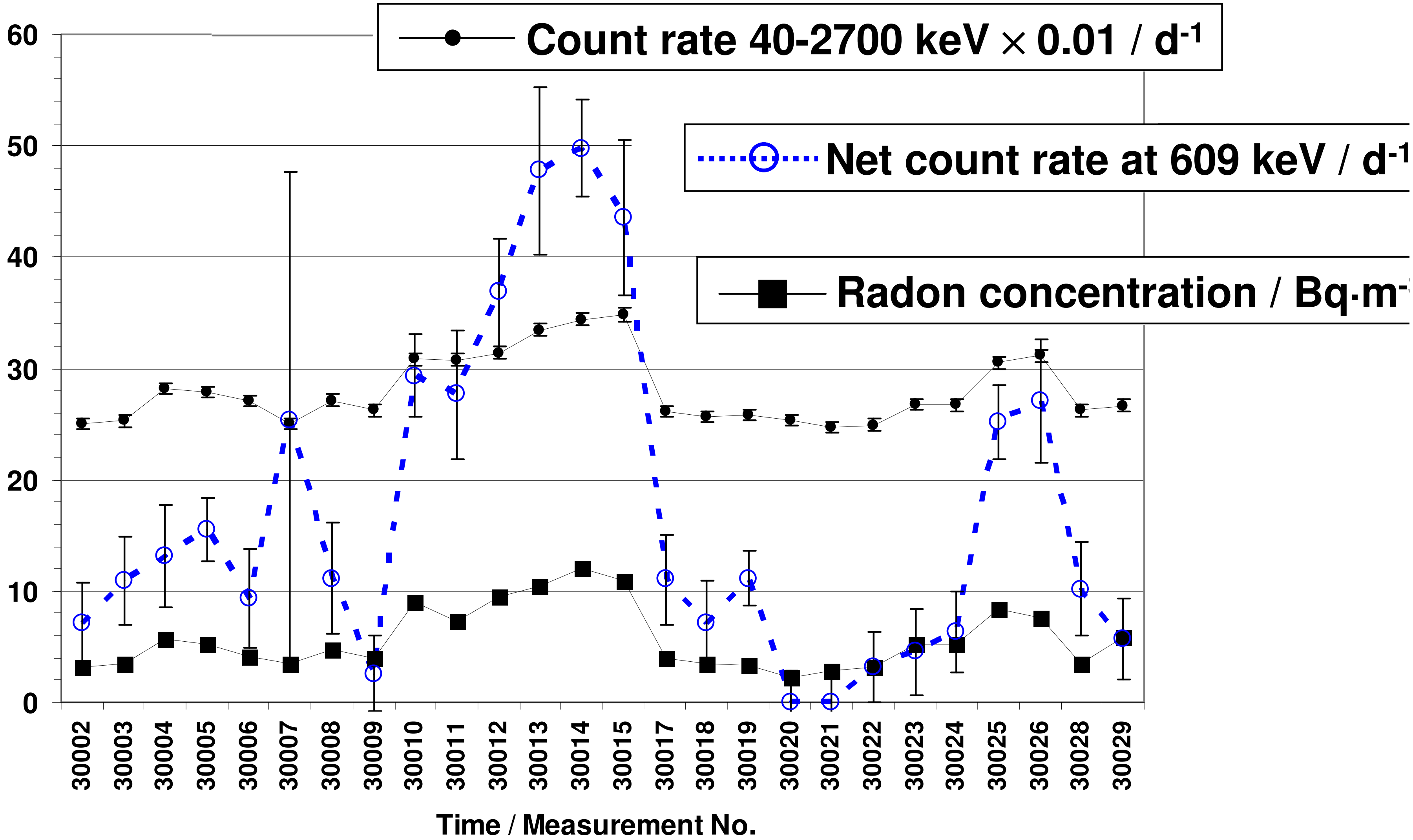
**(soon 11, maybe 12)**

**+3 NaI + 4 PS**

European Commission



European Commission



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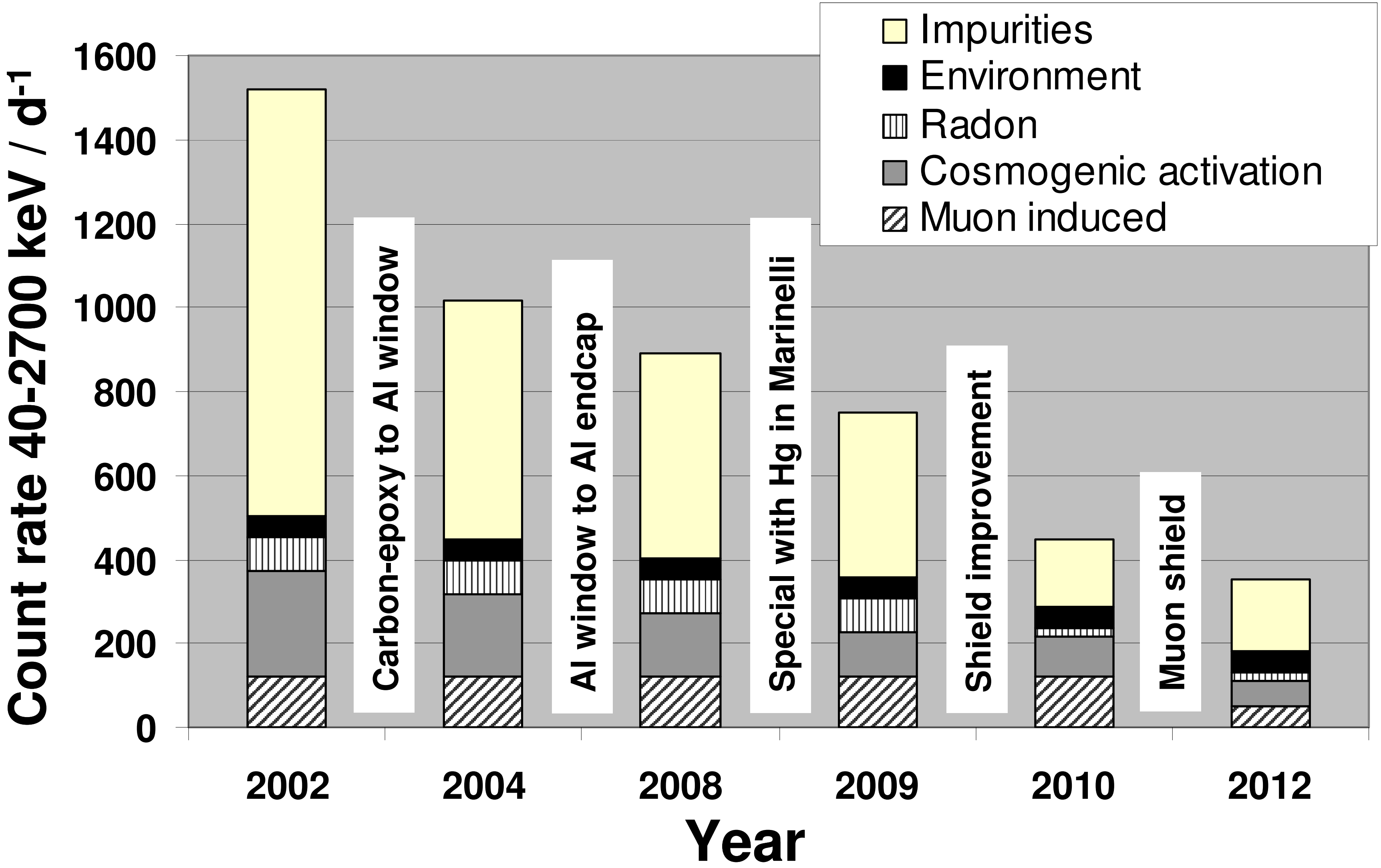
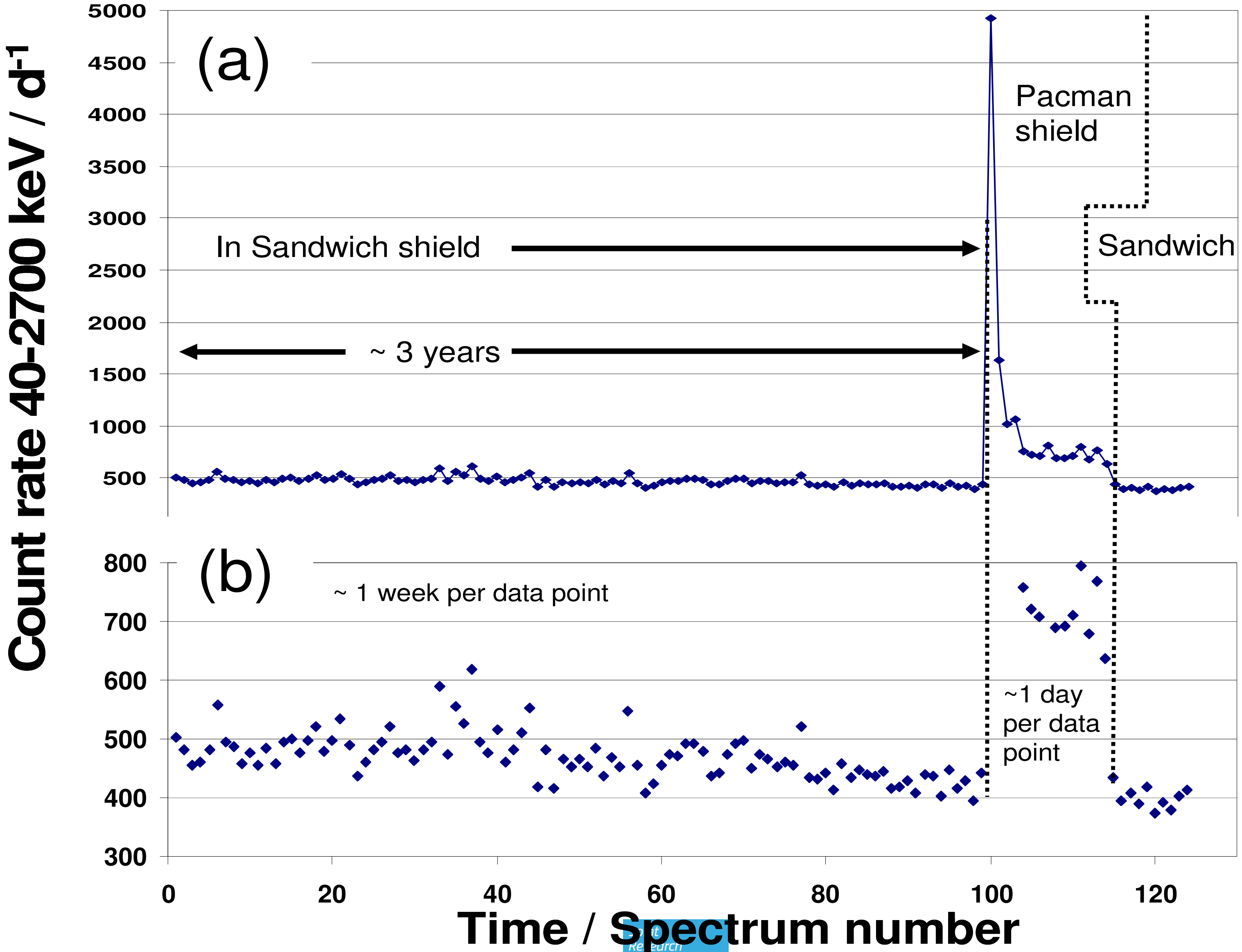
# Detector Ge-5




Count rate 40-2700 keV / d⁻¹
(a)
Pacman shield
In Sandwich shield
Sandwich
~ 3 years
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
(b)
~ 1 week per data point
~1 day per data point
800
700
600
500
400
300
0
20
40
60
80
100
120
Time / Spectrum number

European Commission

# A synergetic process…

**Policy**

Outcome: technology transfer, e.g. radiation protection, **EU policies**

Fundamental physics
search for rare events

***IRMM =***
**Institute for Reference Materials and Measurements**

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

Search for radiopure materials

Ultra Low-level
γ–ray Spectrometry
**ULGS**

**Research**

Joint Research Centre

# Low energy problems

European Commission

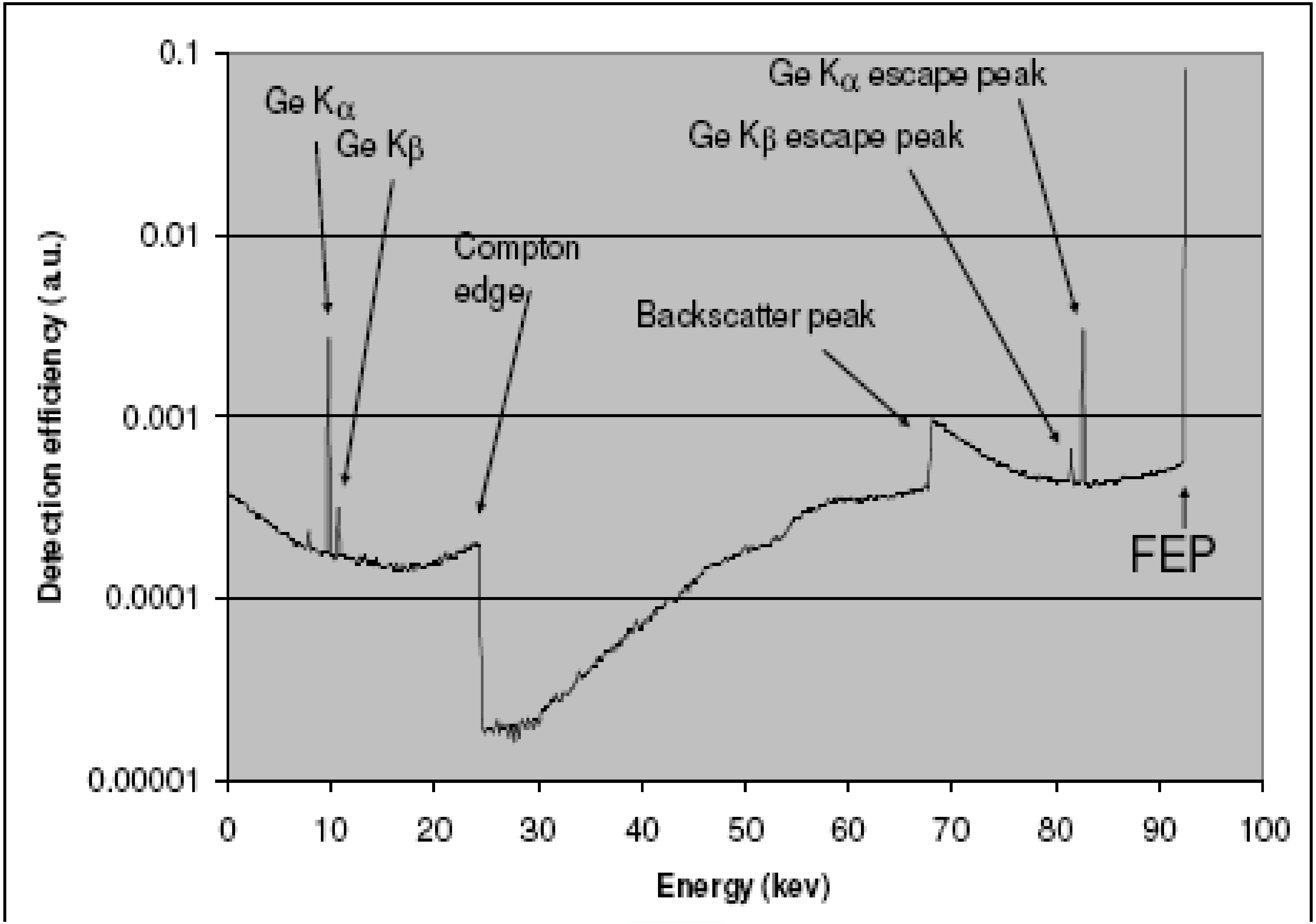
## Problems manifest themselves in numerous situations

- Decay data (see e.g. $^{234}$Th)
- Intercomparisons; more scatter at lower energies (e.g. $^{210}$Pb, $^{109}$Cd, $^{235}$U, $^{142}$Am,… also $^{133}$Ba and $^{152}$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.

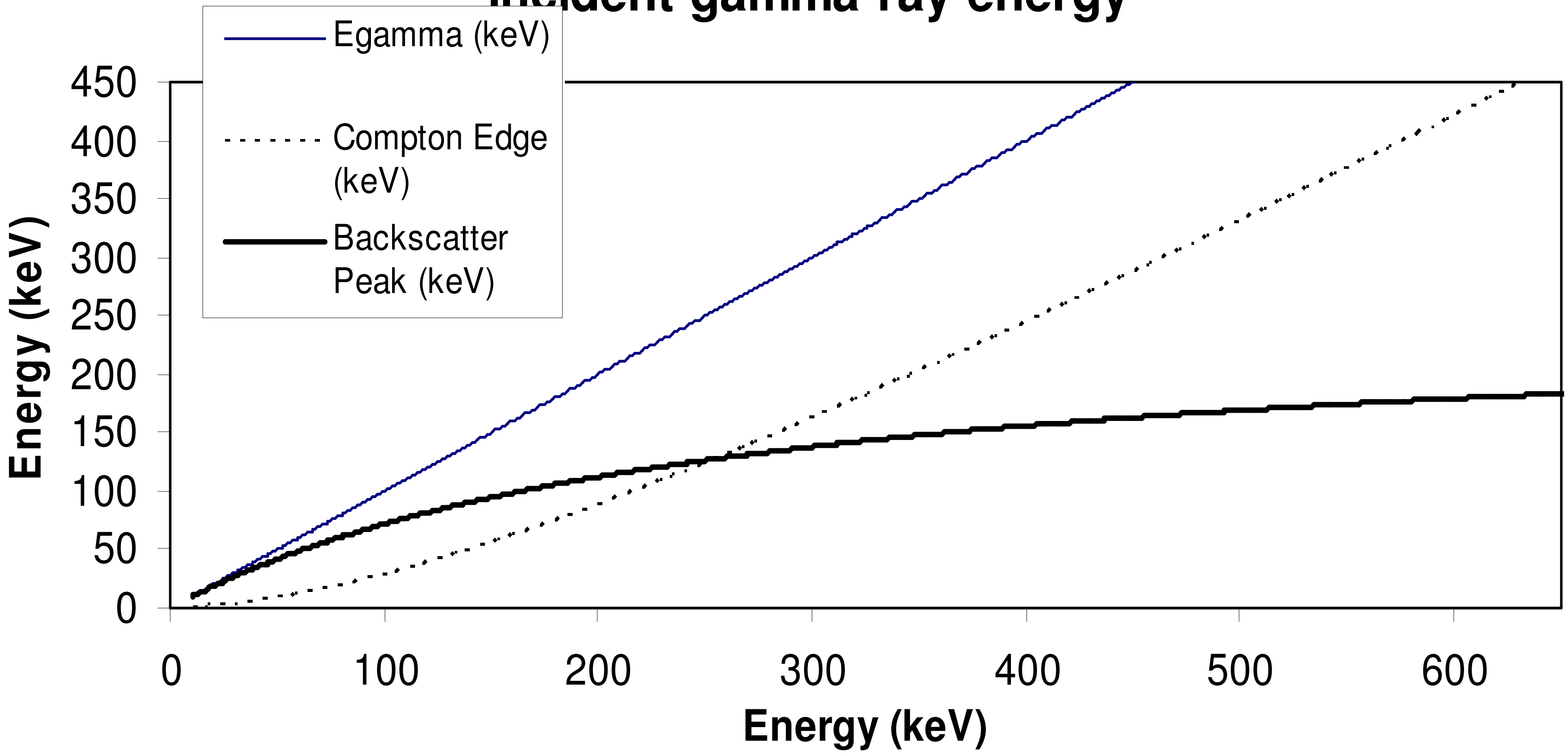
D

# Detector response of 92.5 keV

Useful with Monte Carlo simulation, can “isolate” contriutions



# Compton Edge and Backscatter peak as a function of incident gamma-ray energy



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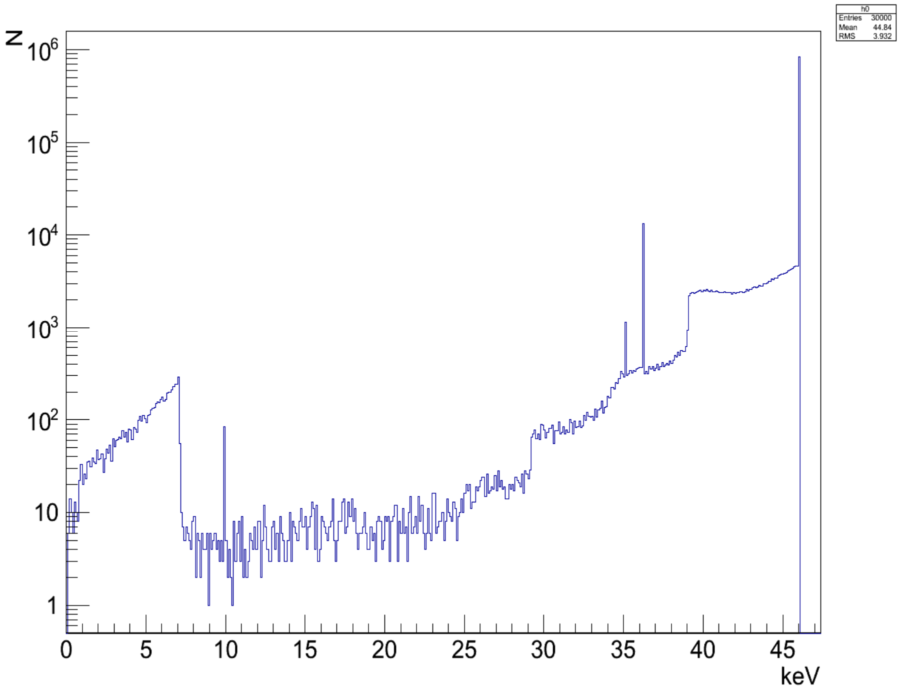
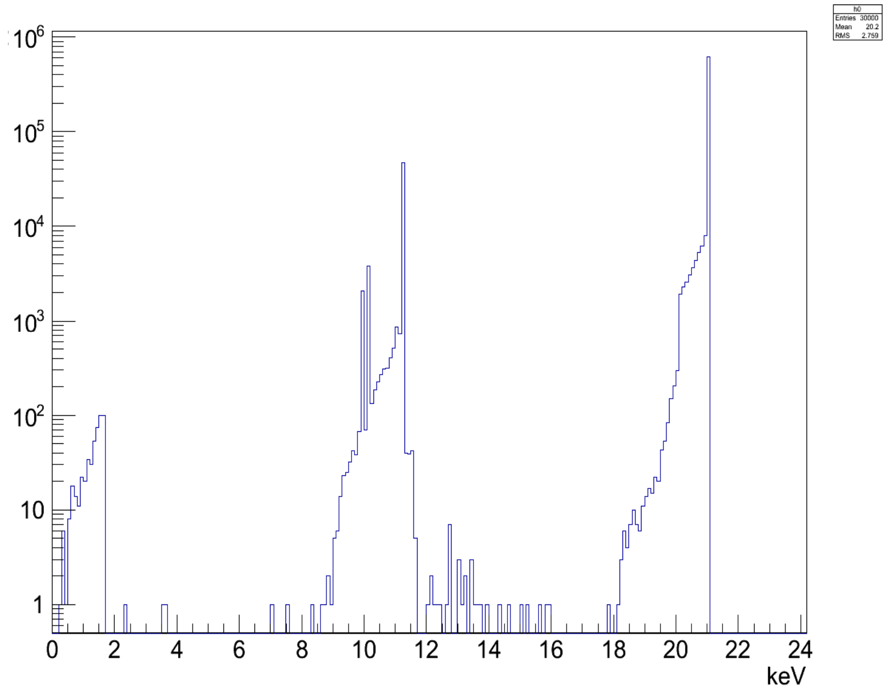
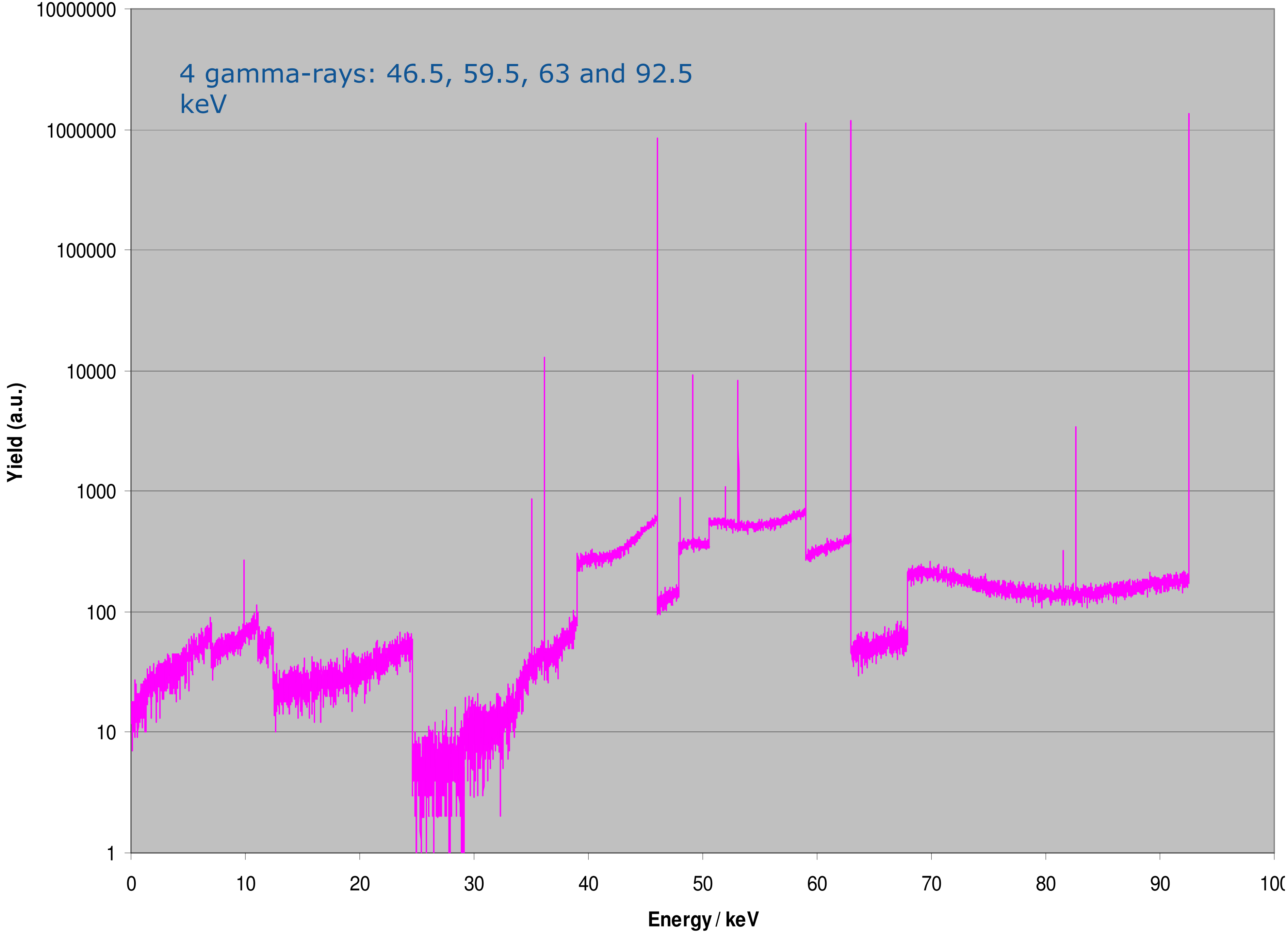
## 46 keV




## 21 keV




4 gamma-rays: 46.5, 59.5, 63 and 92.5
keV
Yield (a.u.)
10000000
1000000
100000
10000
1000
100
10
1
0
10
20
30
40
50
60
70
80
90
100
Energy / keV

European Commission

# Problems with thin deadlayers

- Beta particles can reach the sensitive volume
- Higher background at low energy
- Coincidence summing with X-rays

European Commission

# Focus on $^{238}U$

- 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.

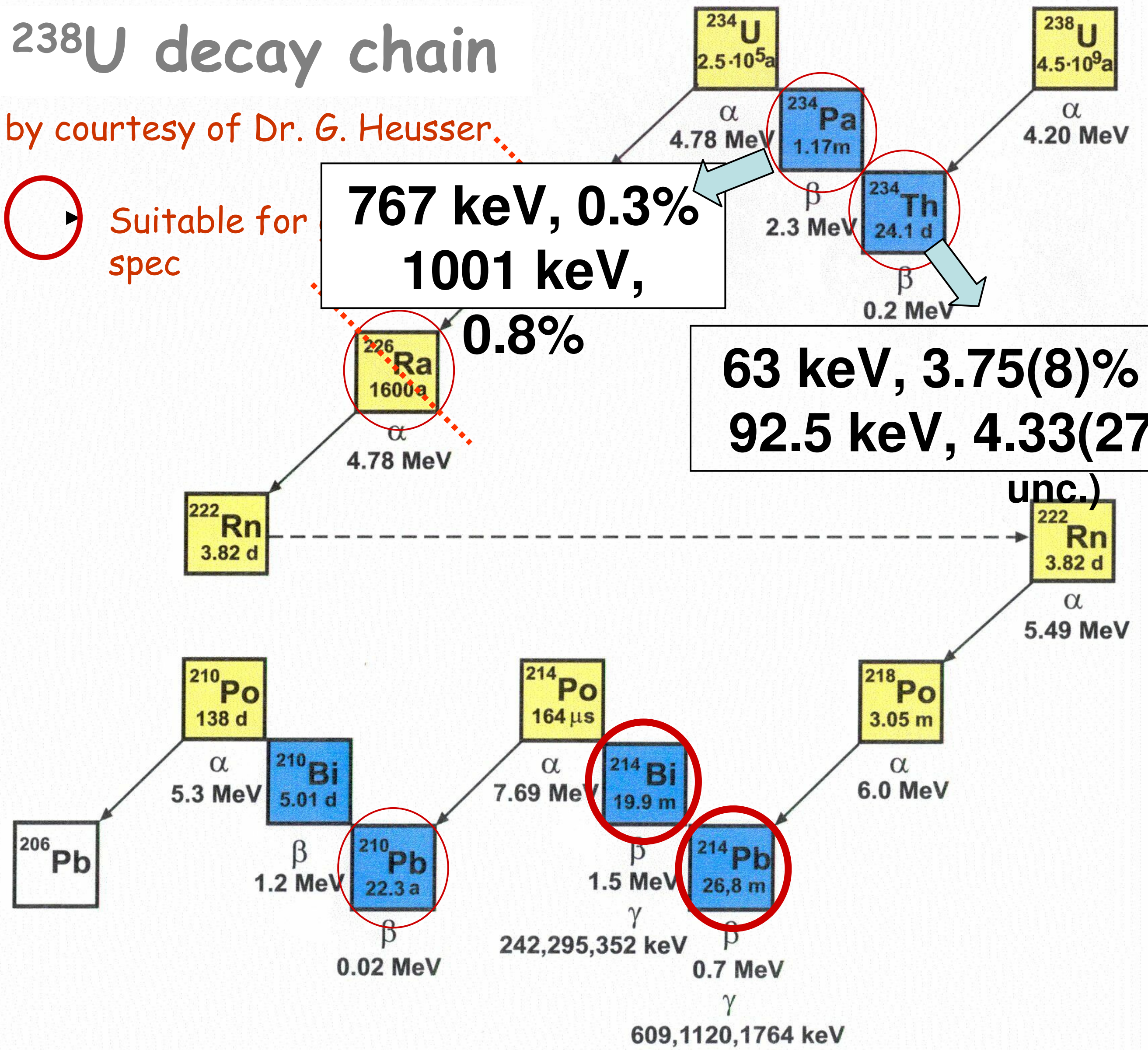
# $^{238}U$ decay chain

by courtesy of Dr. G. Heusser

Suitable for 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?

European Commission

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

**Std.dev: 0.45**

**Rel Std. dev. 11%**

**(Max-min)/average: 30%**

European Commission

# 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**

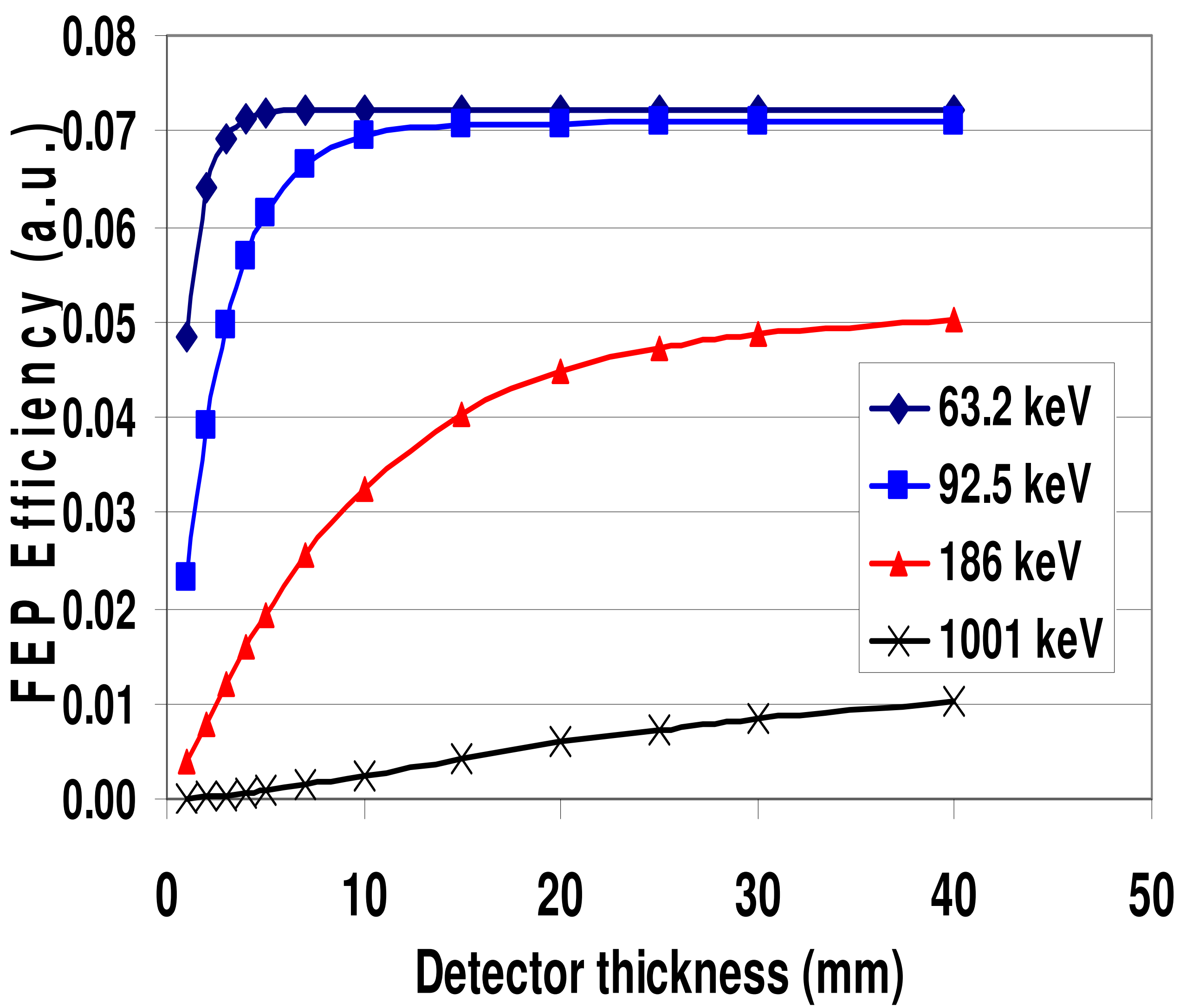
    **⇒ Use "All purpose detectors" with care**

# More Problems / optimisation

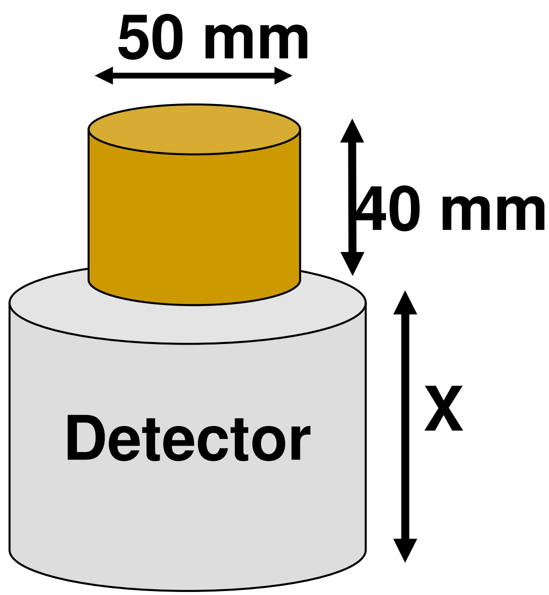
European Commission

- **Optimising sample size and geometry**
- **Subtraction of interfering peaks**
  - **(93.3 keV Th $K_{\alpha 1}$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**

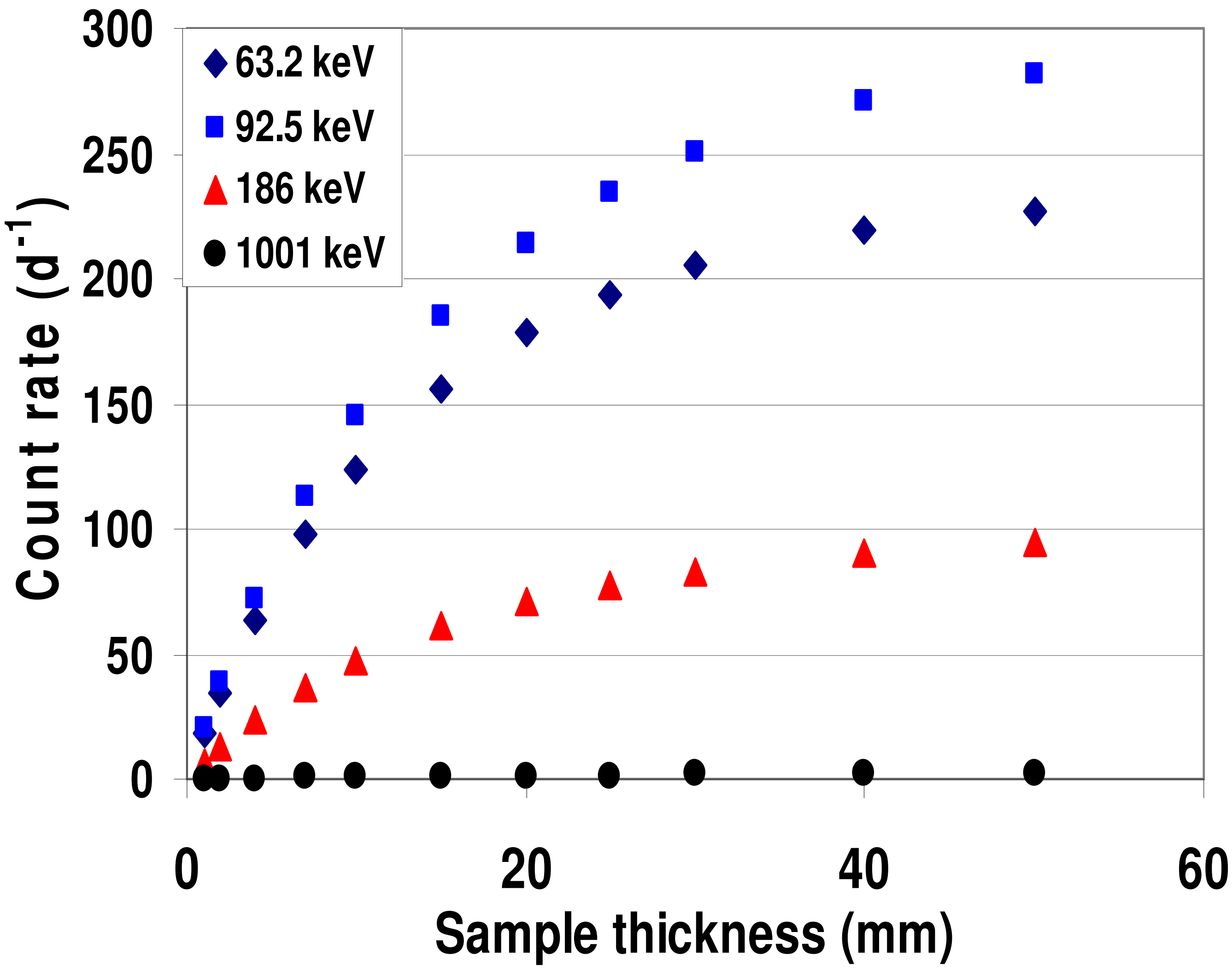



**Sample:**
**Density 1.3 g/cm³**
**Dried soil**

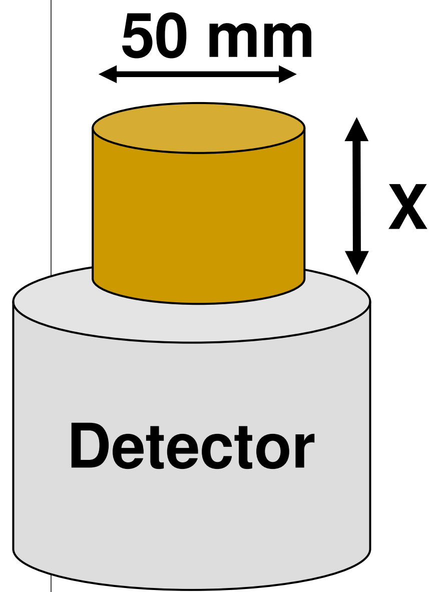


# FEP count rate from U-238 decay

European Commission




**Sample:**
Density 1.5 g/cm$^3$
Dried soil
Detector: 50% rel. eff. BEGe



# *Thank you for your attention!*