IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

Leda : a new γ-γ coincidence spectrometer

Low-level radioactivity measurement in environmental samples

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ICRM

INTERNATIONAL COMMITTEE FOR RADIONUCLIDE METROLOGY

Laboratory of Environment Radioactivity Measurement

- Which samples ? biota, soils, waters, aerosol filters…
- Which radionuclides ?
  - **Present at trace levels** in the French **environment** (nuclear weapon tests, Chernobyl accident, nuclear facilities discharges) : ^{58}Co, ^{60}Co, ^{110m}Ag, ^{137}Cs…
  - Potentially **released** in case of an **incident** or an **accident** : ^{131}I, ^{134}Cs…

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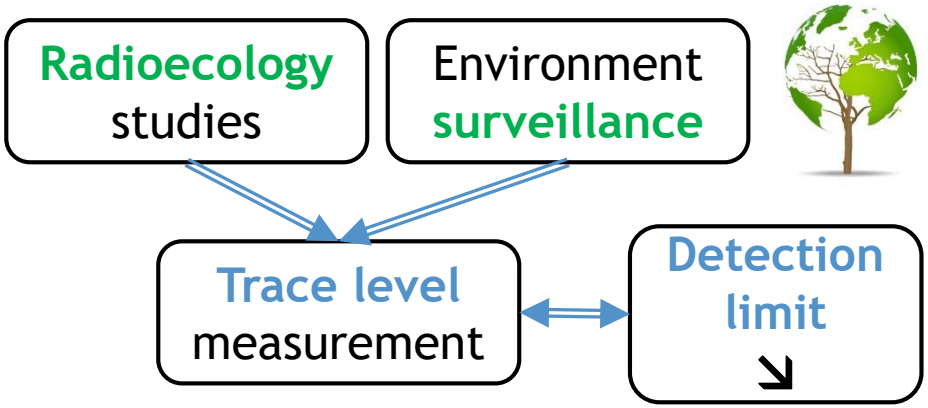

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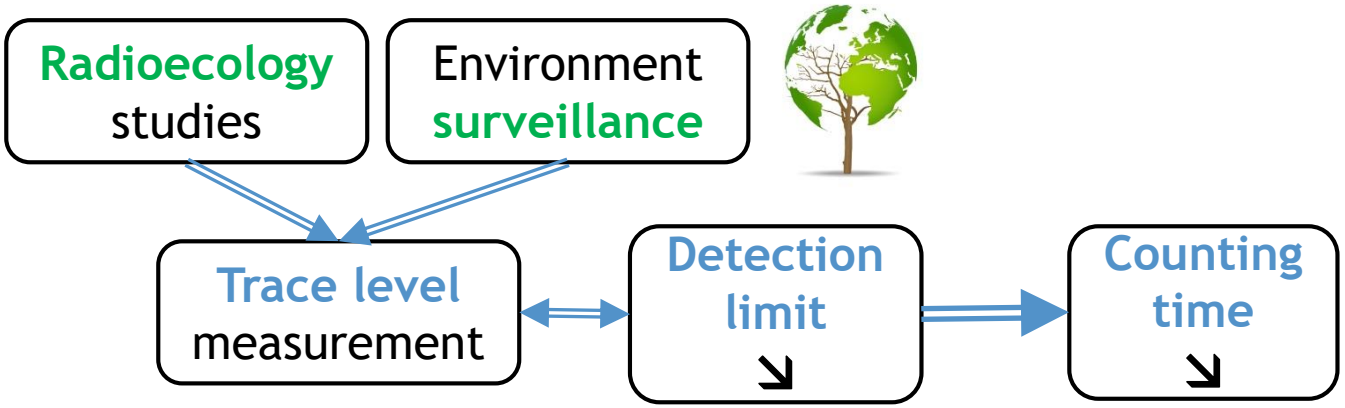

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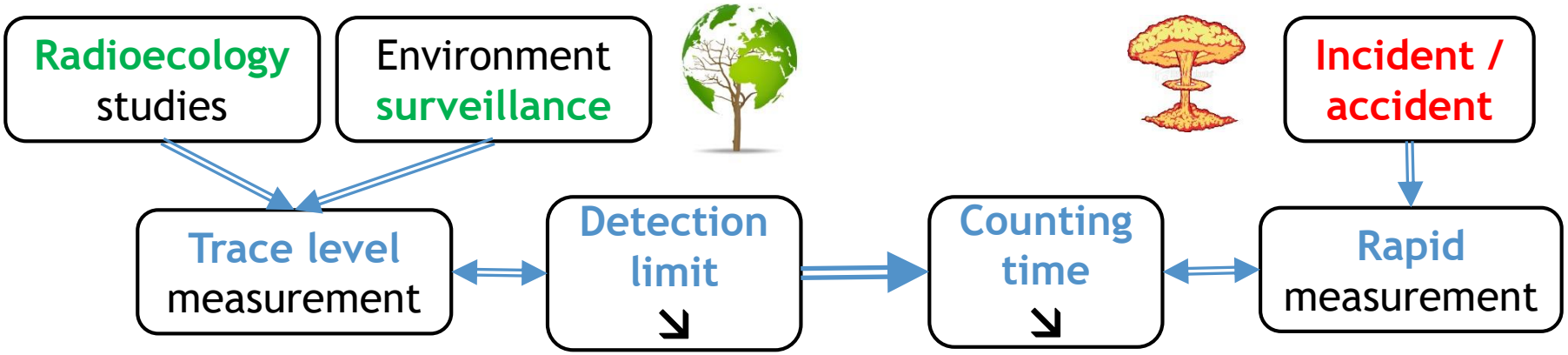

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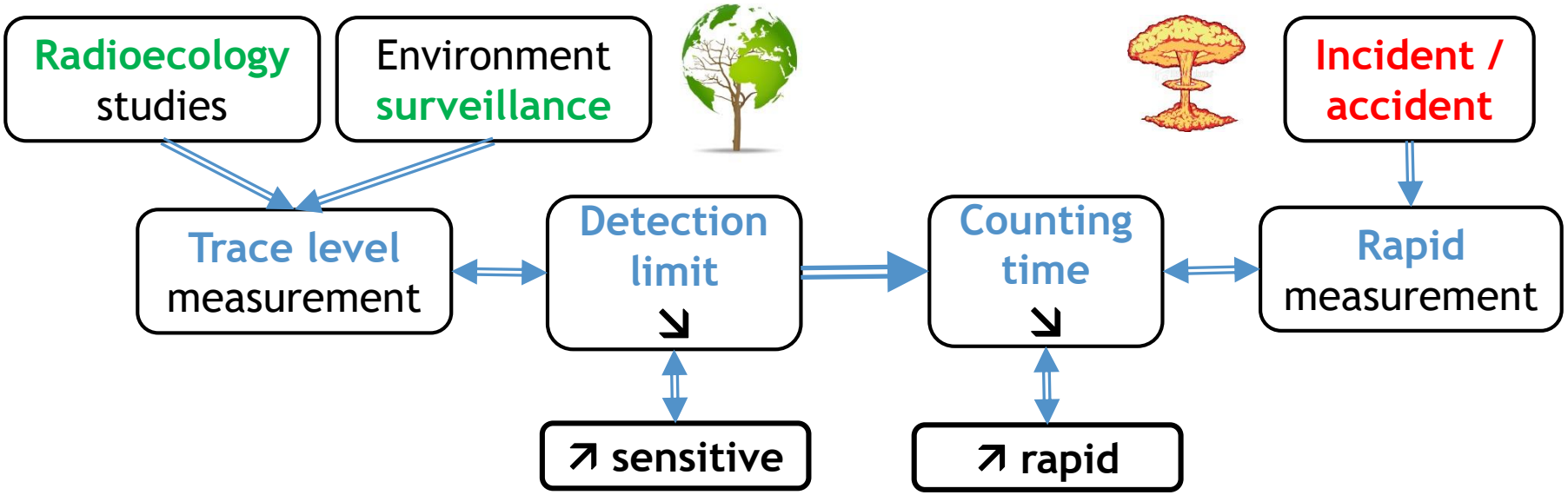


Continuous improvement of materials and methods of analysis

Detection limit improvement

Enhancement of the signal-to-noise ratio (SNR)

Signal ↗ : Detector size, detector efficiency, sample activity concentration…

Background ↘

- intrinsic : Low background detector
- telluric :
  - lead shield
  - shielded room
- radon :
  - ventilation
  - nitrogen flushing
- cosmic rays :
  - shallow or deep facilities
  - **anti-cosmic devices**
- sample : **anti-Compton set up**

Home made anti-cosmic devices

anti-Compton set up

Principle of coincidence measurement

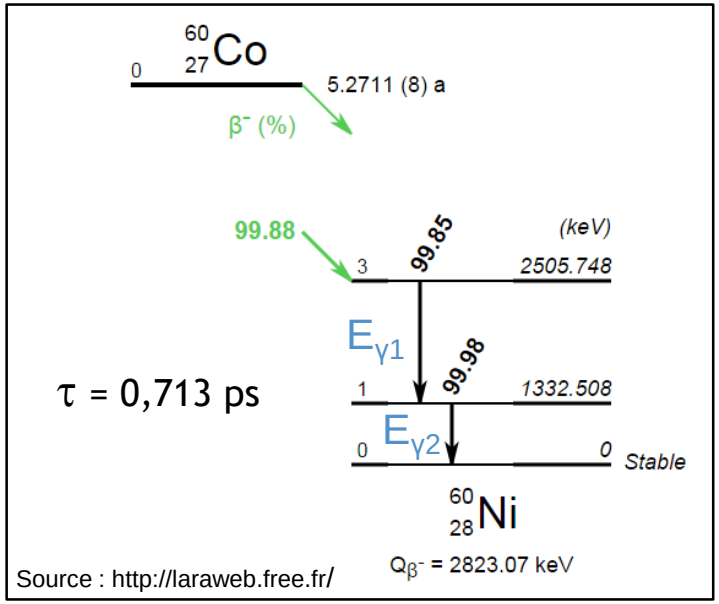
Coincidence emission

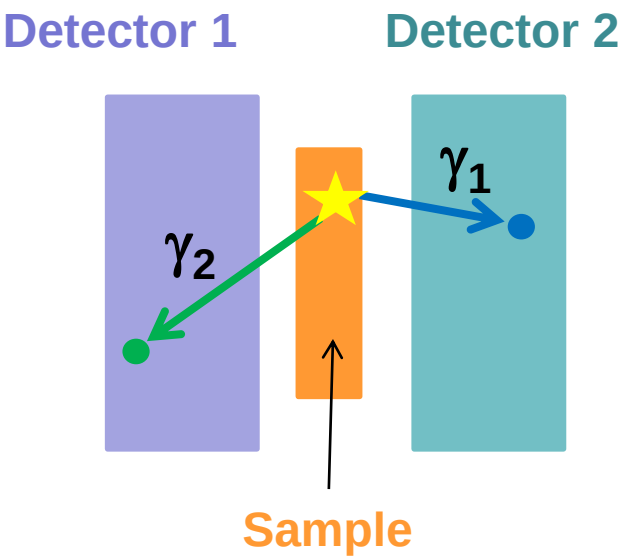
- Cascade decay scheme
- Short-lived intermediate levels

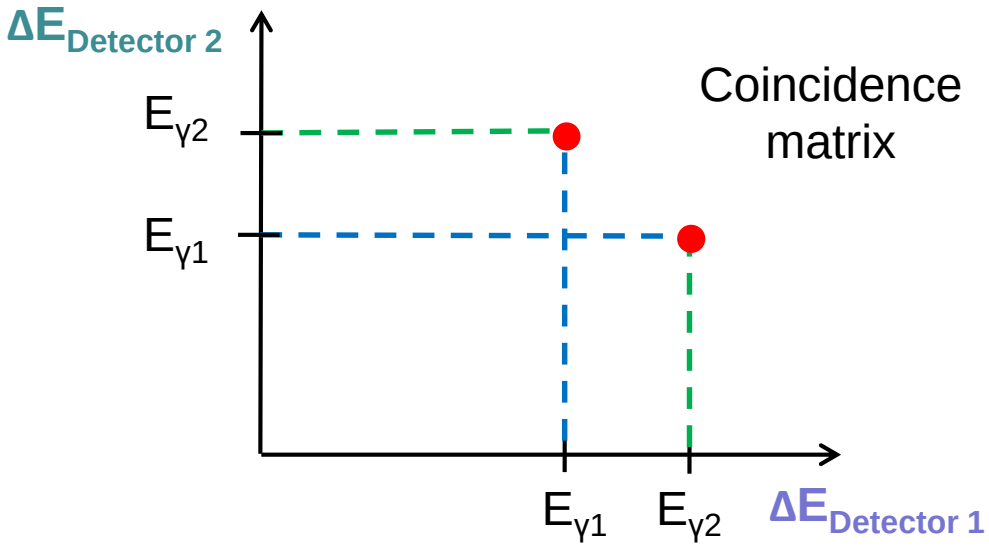
Coincidence detection

Characteristic fingerprints

- Identification of the radionuclide
- Quantification of the radionuclide

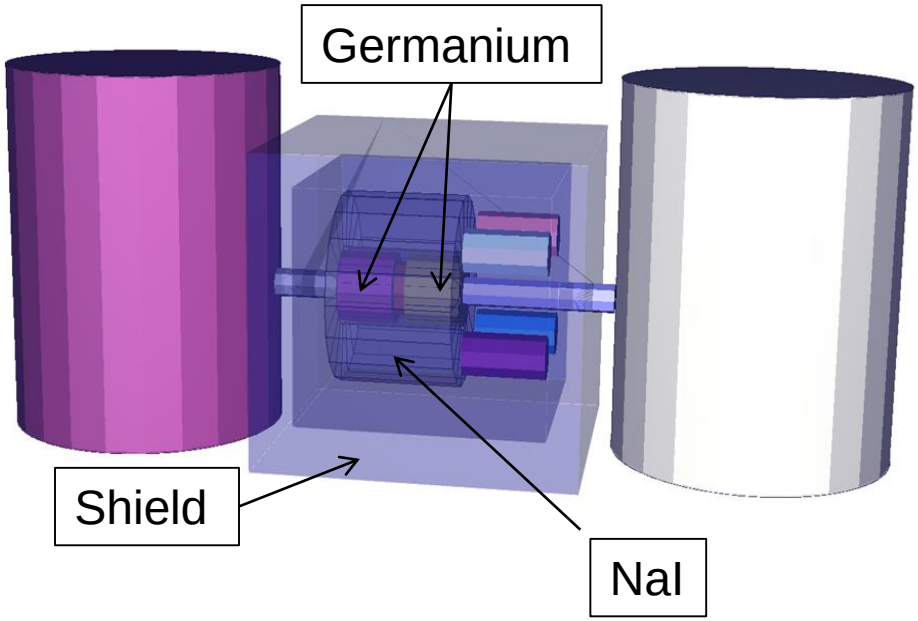






Measurement set-up : Leda

- Designed by Monte Carlo simulations (MCNP_CP)
  - 2 HPGe : Ge/Ge coincidence, resolution, efficiency
  - NaI(Tl) annulus: “4 π”, coincidence, anti-coincidence



- Shielded room
  - under 3 m slab of borated concrete
  - 10 cm lead + 5 mm copper
  - ➔ telluric radioactivity /100
  - ➔ low background lead shield (5 cm)

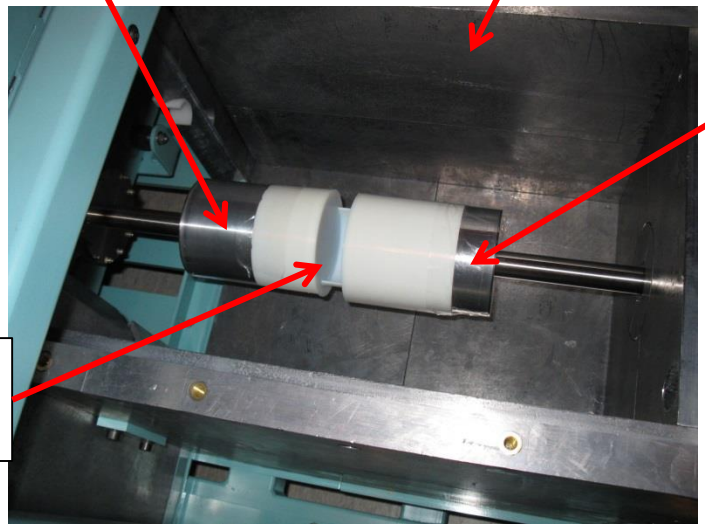
- Digital electronics (Pixie-4, Xia)
  - List mode file
  - Time and energy information
  - Data analysis algorithm
  - ➔ Multiple analysis channels

Measurement set-up : Leda

Mobile detector : BEGe 5030 Canberra **Castor**

Lead shield : **Clytemnestra**

***Leda** and the swan*, Cesare da Sesto



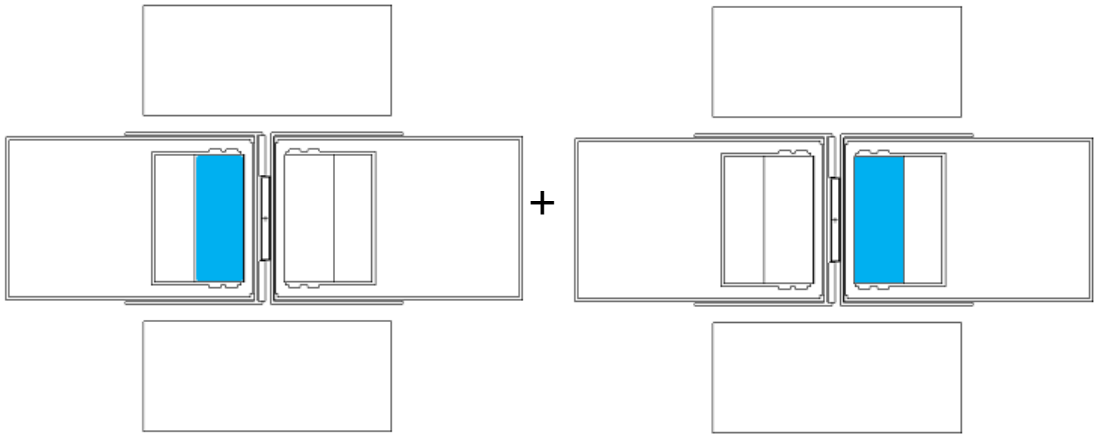
Fix Detector : BEGe 5030 Canberra **Pollux**

Sample holder

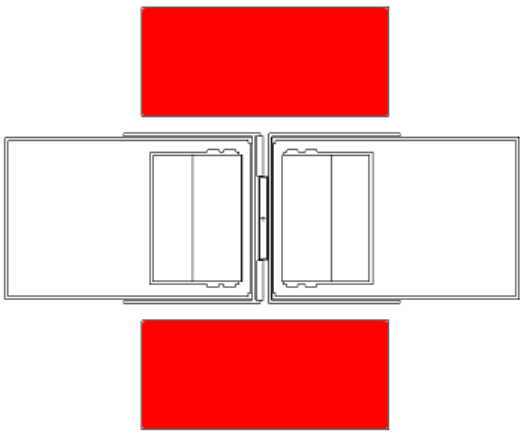
Scintillator : NaI(Tl) Scionix **Helene**

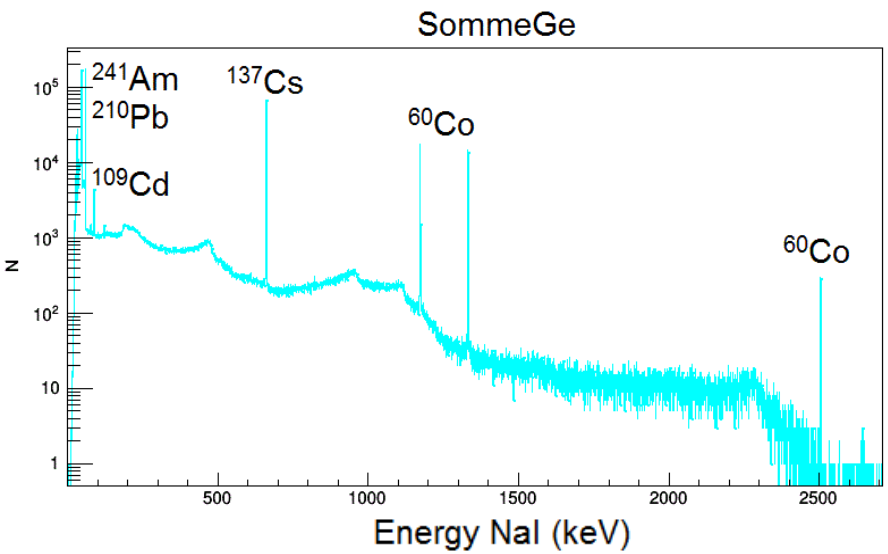
Analysis channel : classics

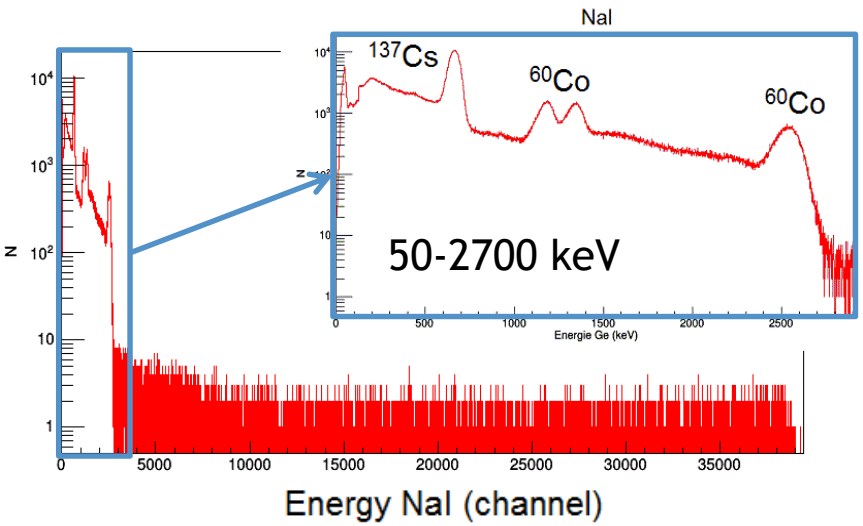
GeSum



NaI

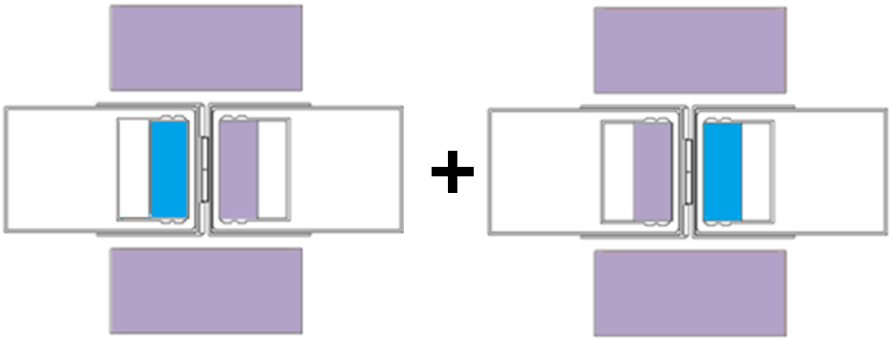


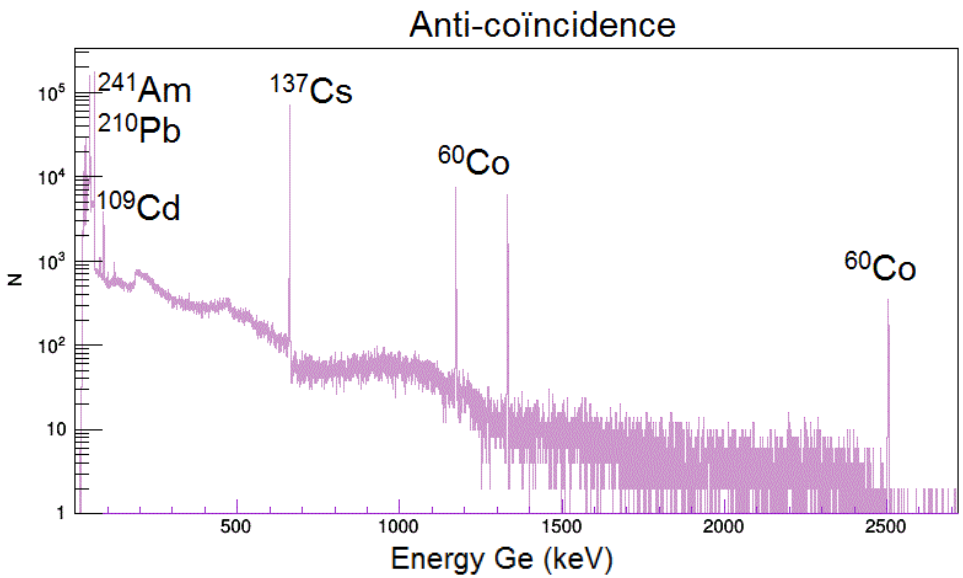




- Channels used if the radionuclide of interest is present in large amount.

Analysis channel : anti-coincidence



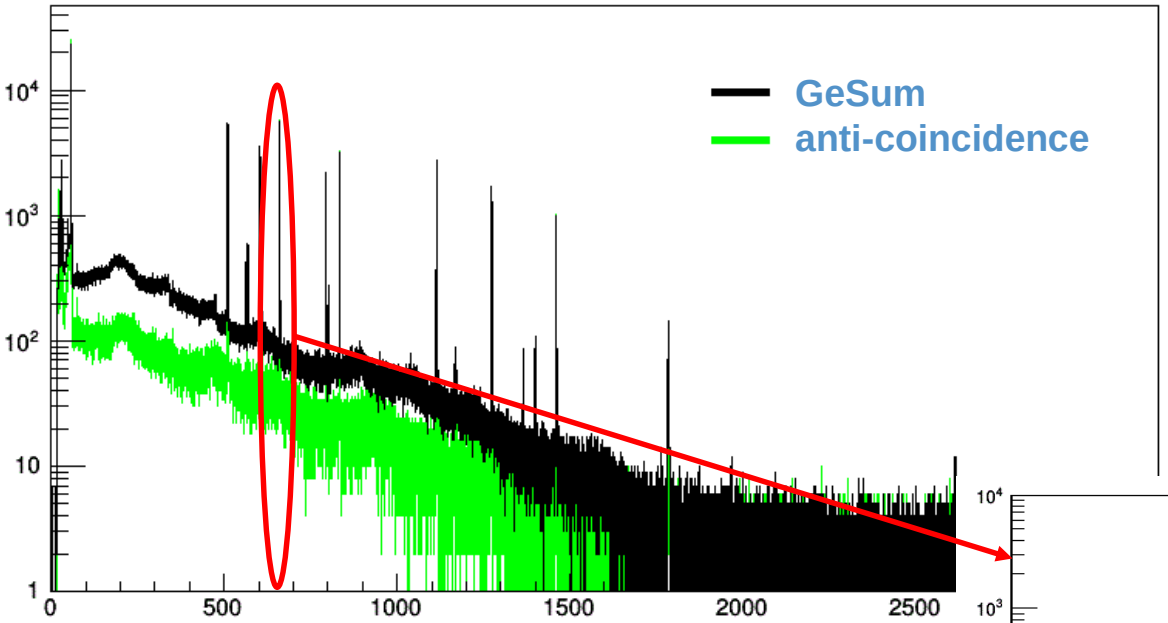


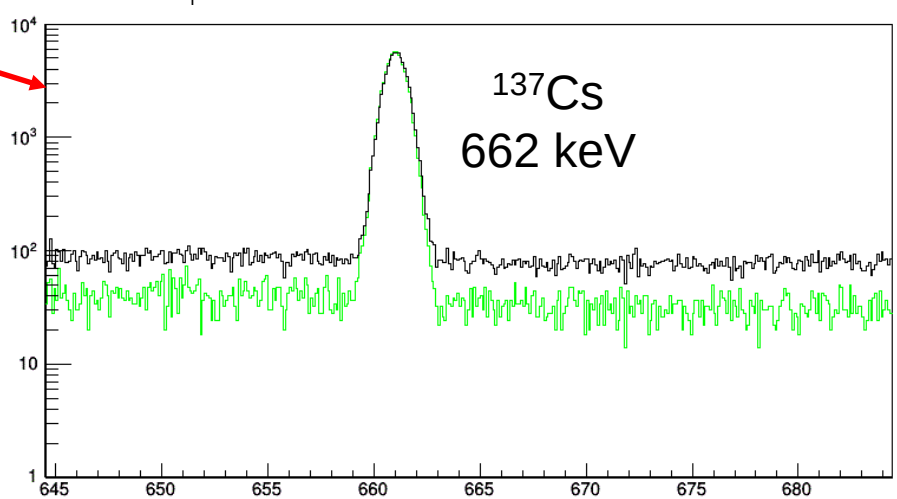
- Channel used for “single” emitters (^{137}Cs, ^{131}I, ^{241}Am…)
  - Compton continuum reduction ➔ anti-Compton measurement
  - Cosmic background decrease ➔ anti-cosmic measurement

Analysis channel : anti-coincidence

Example : ^{137}Cs

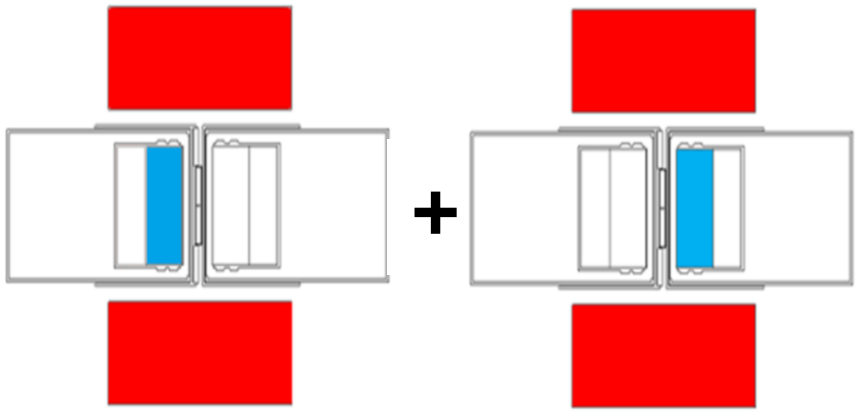
- IAEA proficiency test sample ALMERA 2016 04-03
- Dry vegetal with ^{40}K, ^{134}Cs, ^{137}Cs, ^{241}Am, ^{22}Na

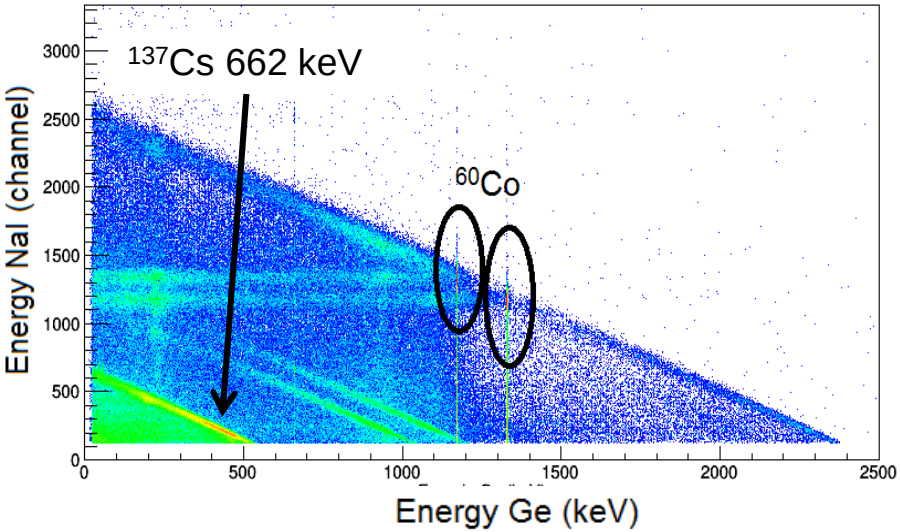


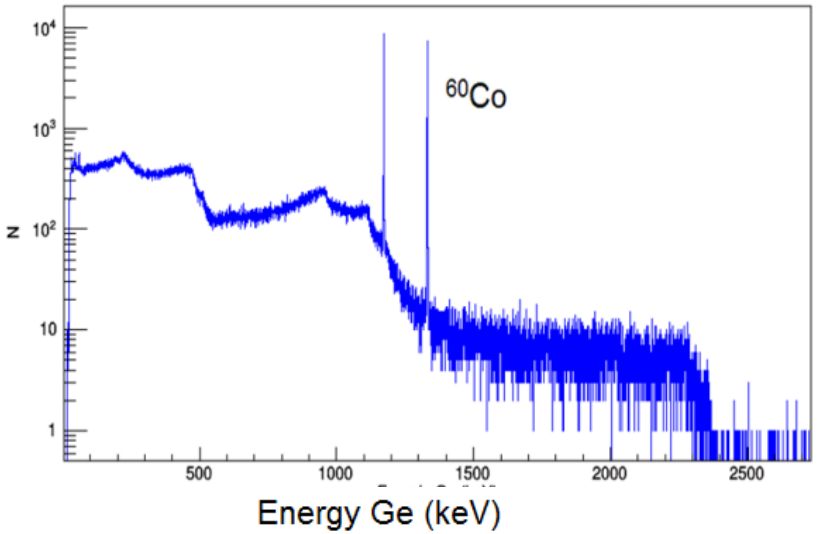


Signal-to-noise ratio X 3

Analysis channel : Ge/NaI coincidence

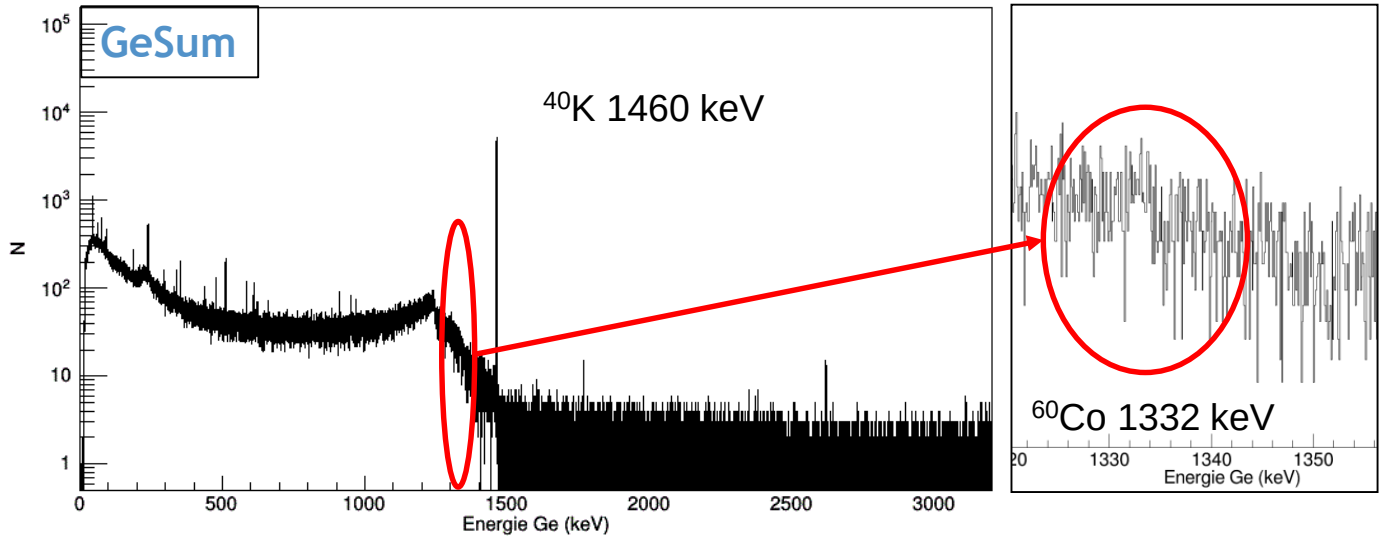




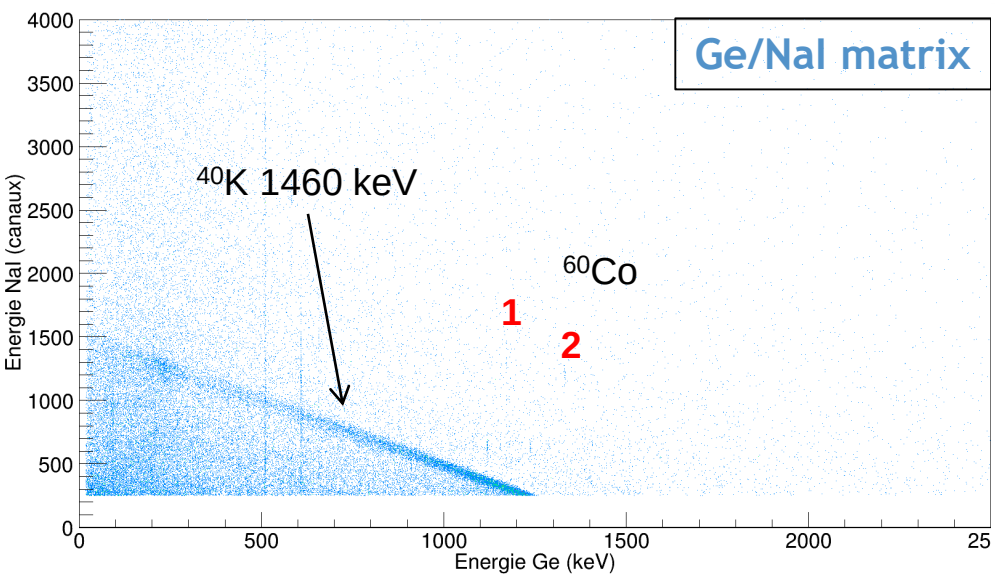


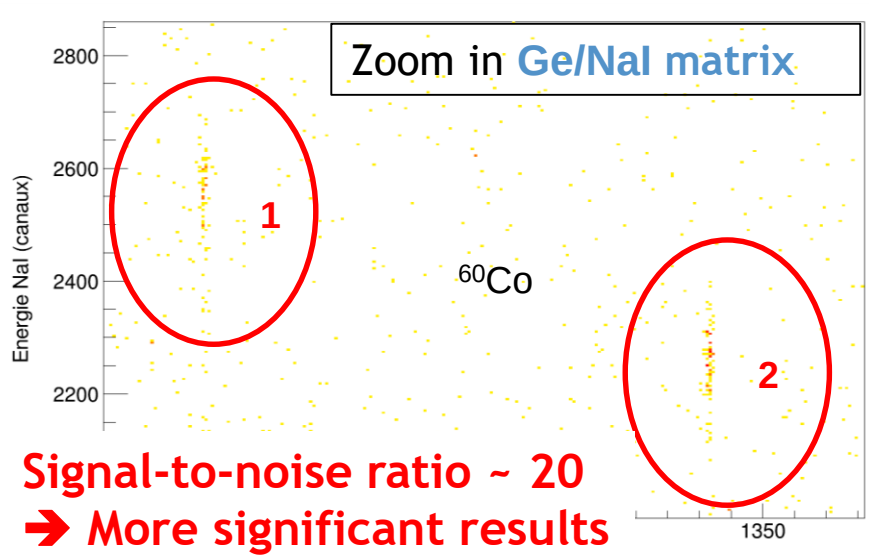
- Channel used for coincidence emitters with “high” energy
  - Matrix if only 2 photons in coincidence (e.g. ^{60}Co…)
  - Spectrum if more than 2 photons in coincidence (e.g. ^{110m}Ag…)

Example : ^{60}Co in a biological sample (^{40}K)



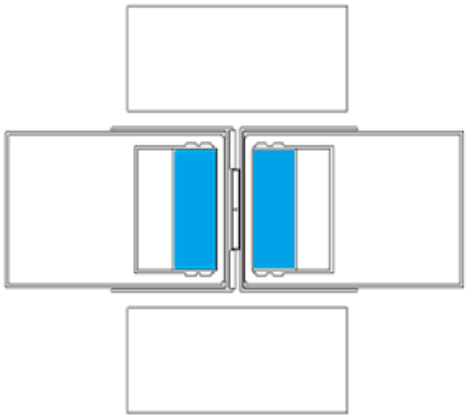
Detection limit

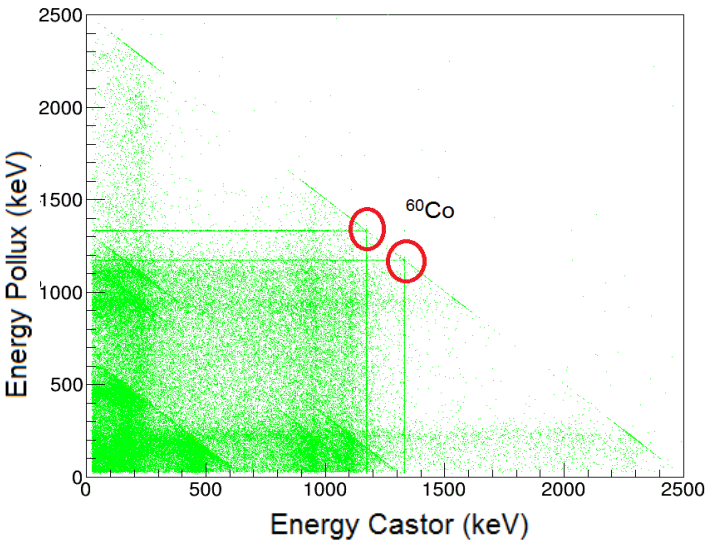


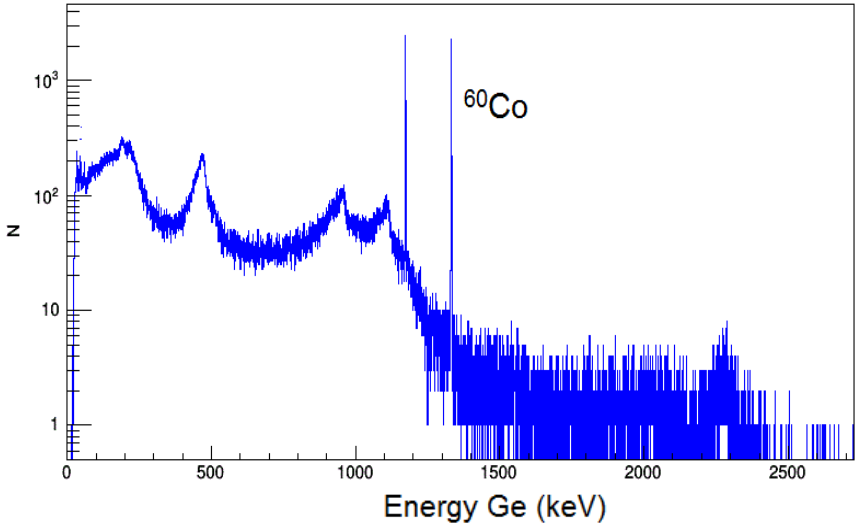


Signal-to-noise ratio ~ 20
➔ More significant results

Analysis channel : Ge/Ge coincidence

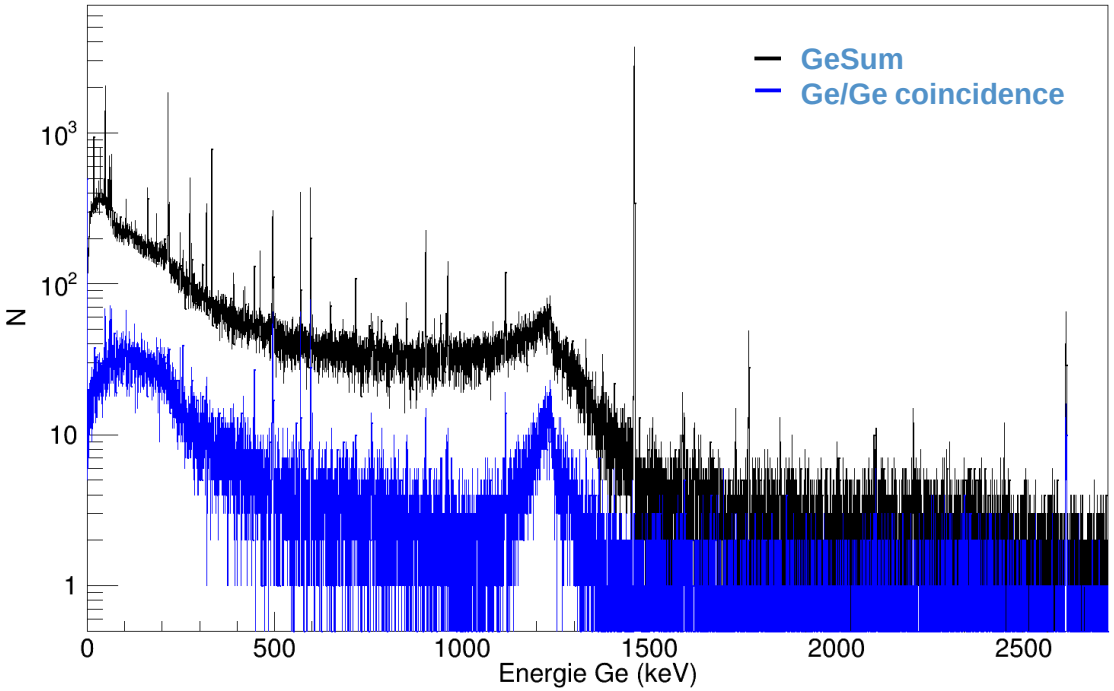


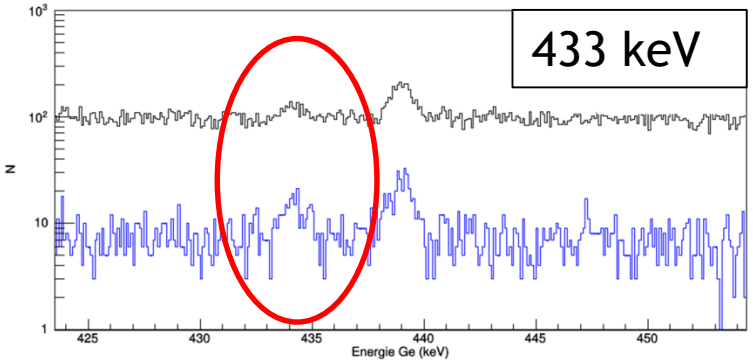


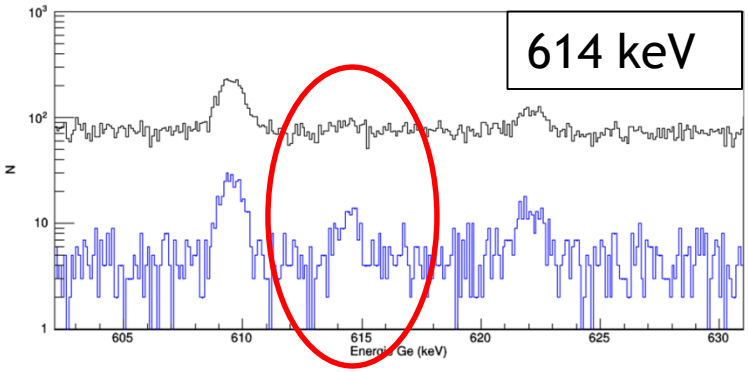


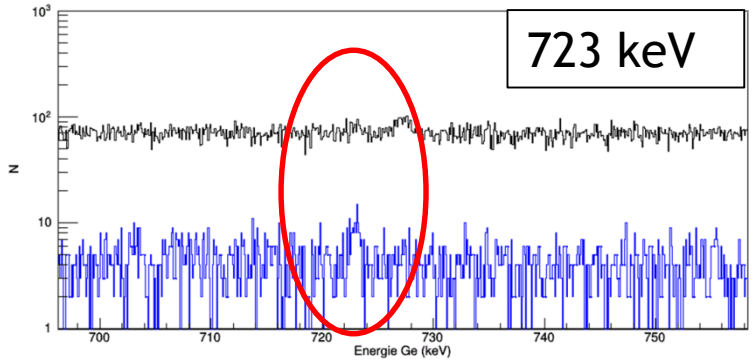
- Channel used for coincidence emitters with one photon energy < 1 MeV
  - Matrix if only 2 photons in coincidence (e.g. ^{134}Cs…)
  - Spectrum if more than 2 photons in coincidence (e.g. ^{108m}Ag…)

Example : ^{108m}Ag in a biological sample (^{40}K)









GeSum spectrum :
only 1 peak @ 433 keV with SNR ~ 0.1

Ge/Ge spectrum :
- 3 peaks
- peak @ 433 keV with SNR ~ 1

Net number of events : $n_{n,i}$

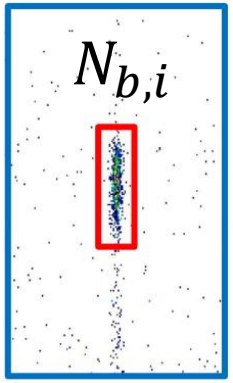
- For each peak or fingerprint i
- Background estimation

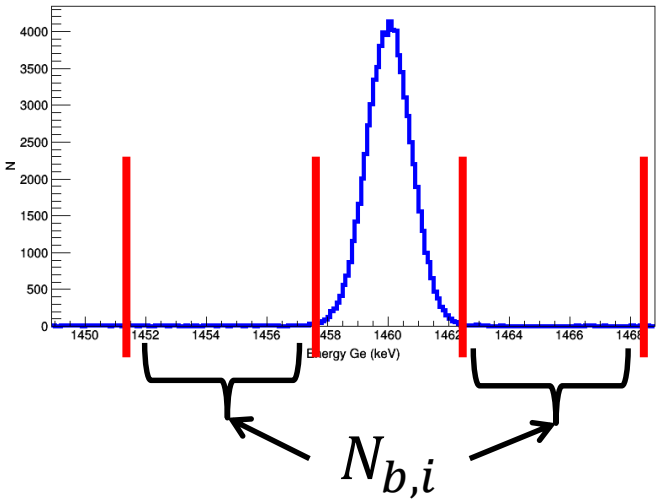
$$n_{b,i} = \frac{N_{b,i}}{\chi}$$

χ : areas/widths ratio

- $n_{n,i} = n_{g,i} - n_{b,i}$

n : net ; g : gross ; b : background





Mass activity : A_i

$$A_i = \frac{n_{n,i}}{C_i . t . m} . FC \quad [\text{Bq/kg}]$$

Calibration coefficient

- Standard source (^{60}Co, ^{88}Y…)
- MCNP_CP (^{108m}Ag, ^{152}Eu, …)

Counting time [s]

Sample mass [kg]

Correction factors

- Decay
- Self-absorption

Efficiency calibration

Standard sources

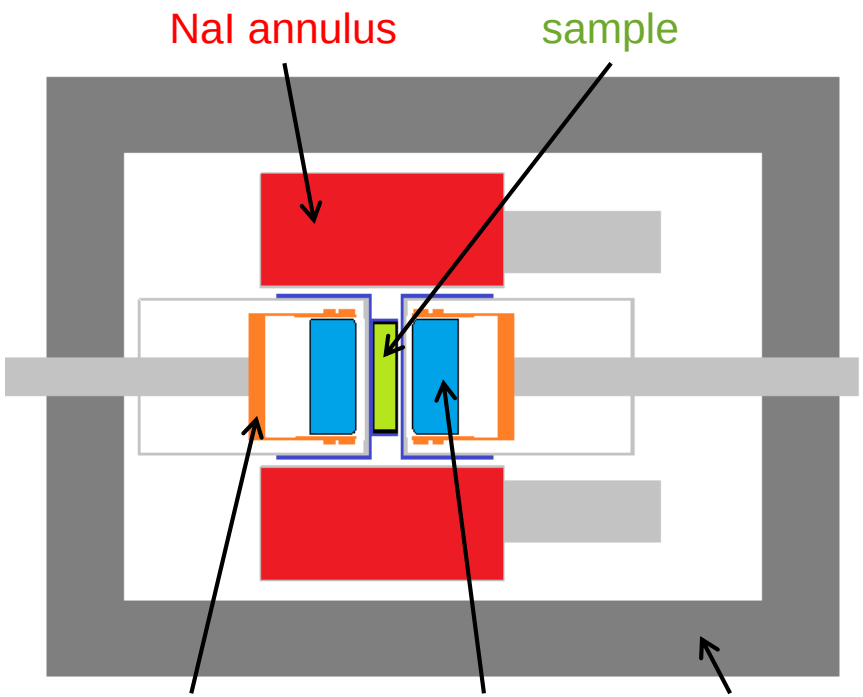
- Same counting geometry : 17, 30 and 60 mL
- Water equivalent resin
- Mixture of gamma emitters : ^{60}Co, ^{137}Cs…

$$C_i = \frac{n_{n,i}(source)}{A(source) \cdot t(source)}$$

Monte Carlo simulation : MCNP_CP

- MCNPX extension by A. Berlizov
- Decay scheme from ENSDF
- List mode ➔ same algorithm

$$C_i(\text{MCNP CP}) = \frac{n_{n,i}(simulated\ matrix)}{NPS}$$



Berlizov A. (2007) ACS Symposium Series 945, 183-197

Leda : samples measurement

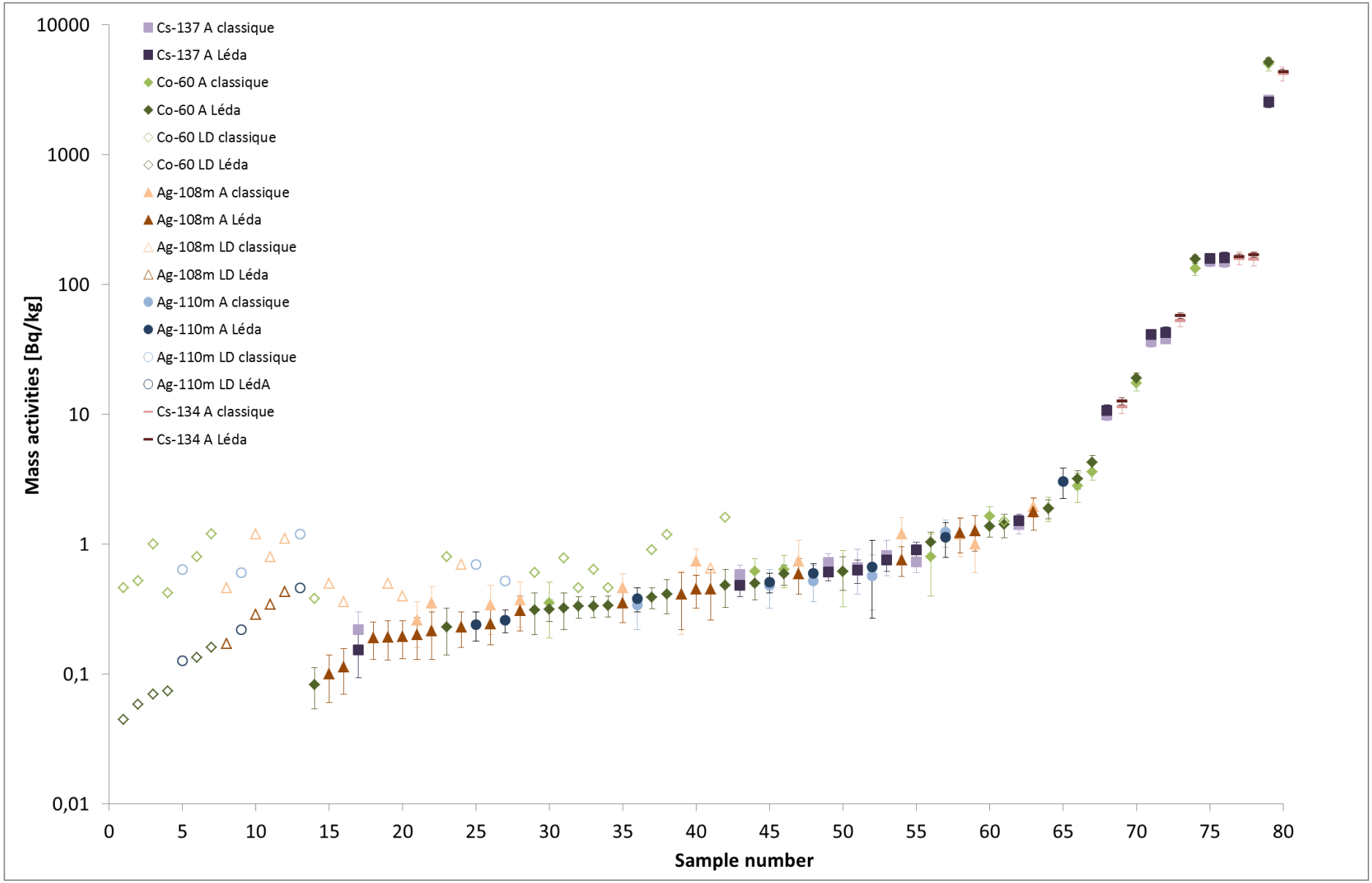
- 80 samples (biological samples, soils, waters…); counting time = 1 day

- Analysis channel depending on :
  - Sample type (if known) ➔ major radionuclide (e.g. ^{40}K in biological samples)
  - Decay scheme of the radionuclide of interest
    - ➔ coincidence emitter ?
    - If so, number and energies of coincidence emissions

- Radionuclides :
  - ^{108m}Ag : Ge/Ge coincident spectrum
  - ^{110m}Ag : Ge/NaI coincident spectrum
  - ^{60}Co : Ge/NaI matrix
  - ^{134}Cs : Ge/Ge matrix or spectrum
  - ^{137}Cs : anti-coincidence spectrum

- Comparison between classic spectrometry and measurement with Leda:
  - Mass activities
  - Detection limits

Leda : samples measurement



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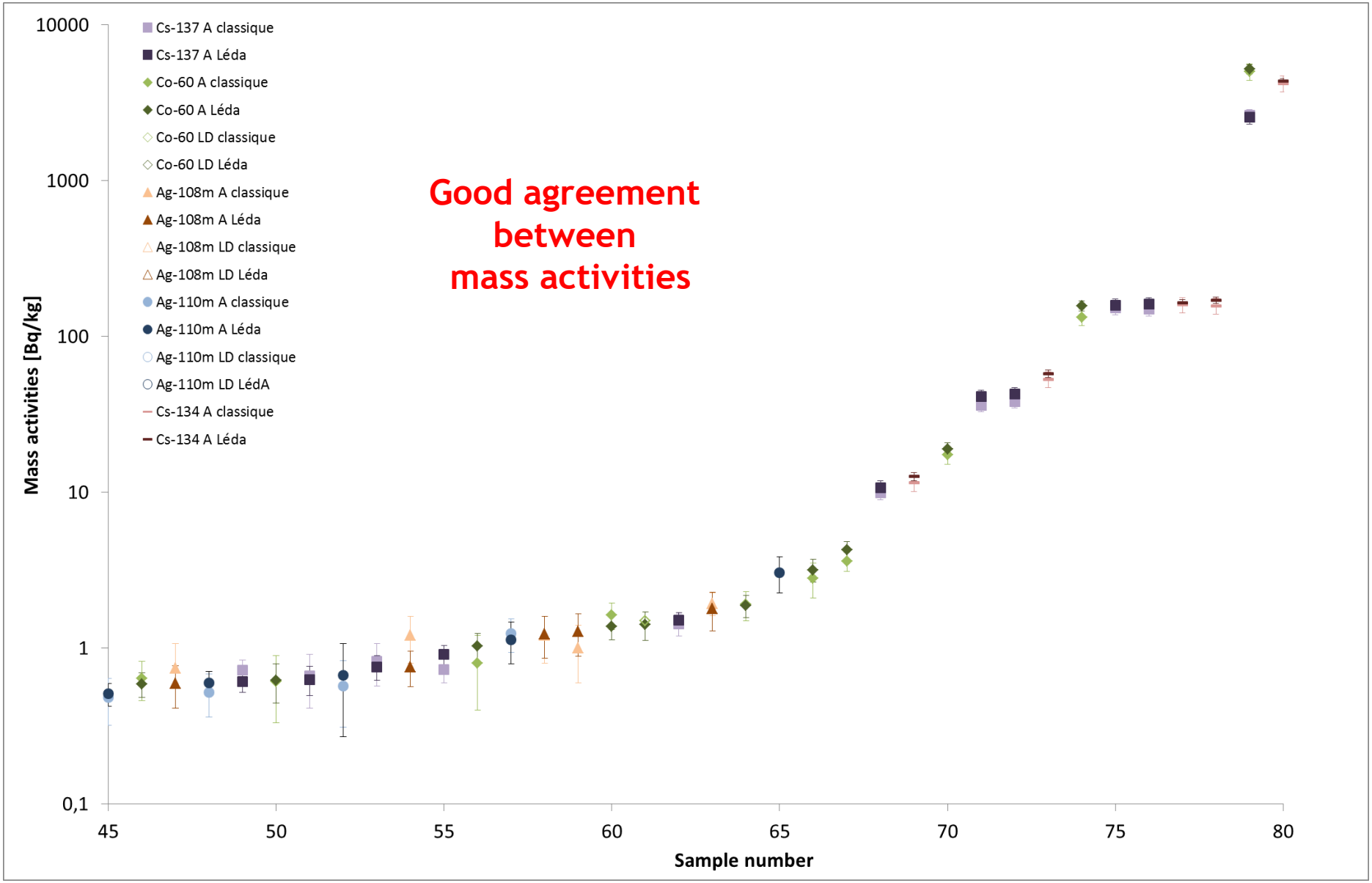

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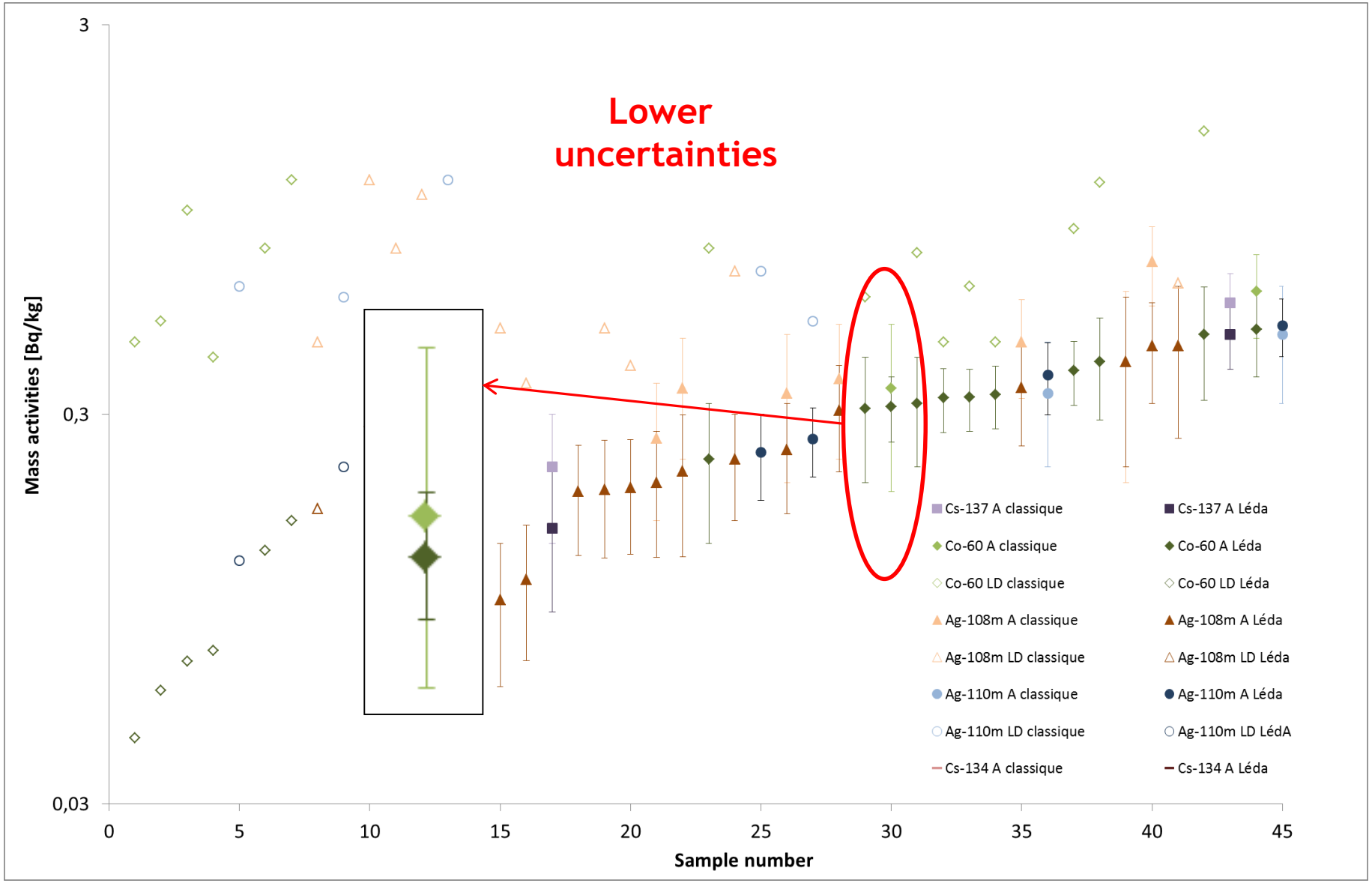

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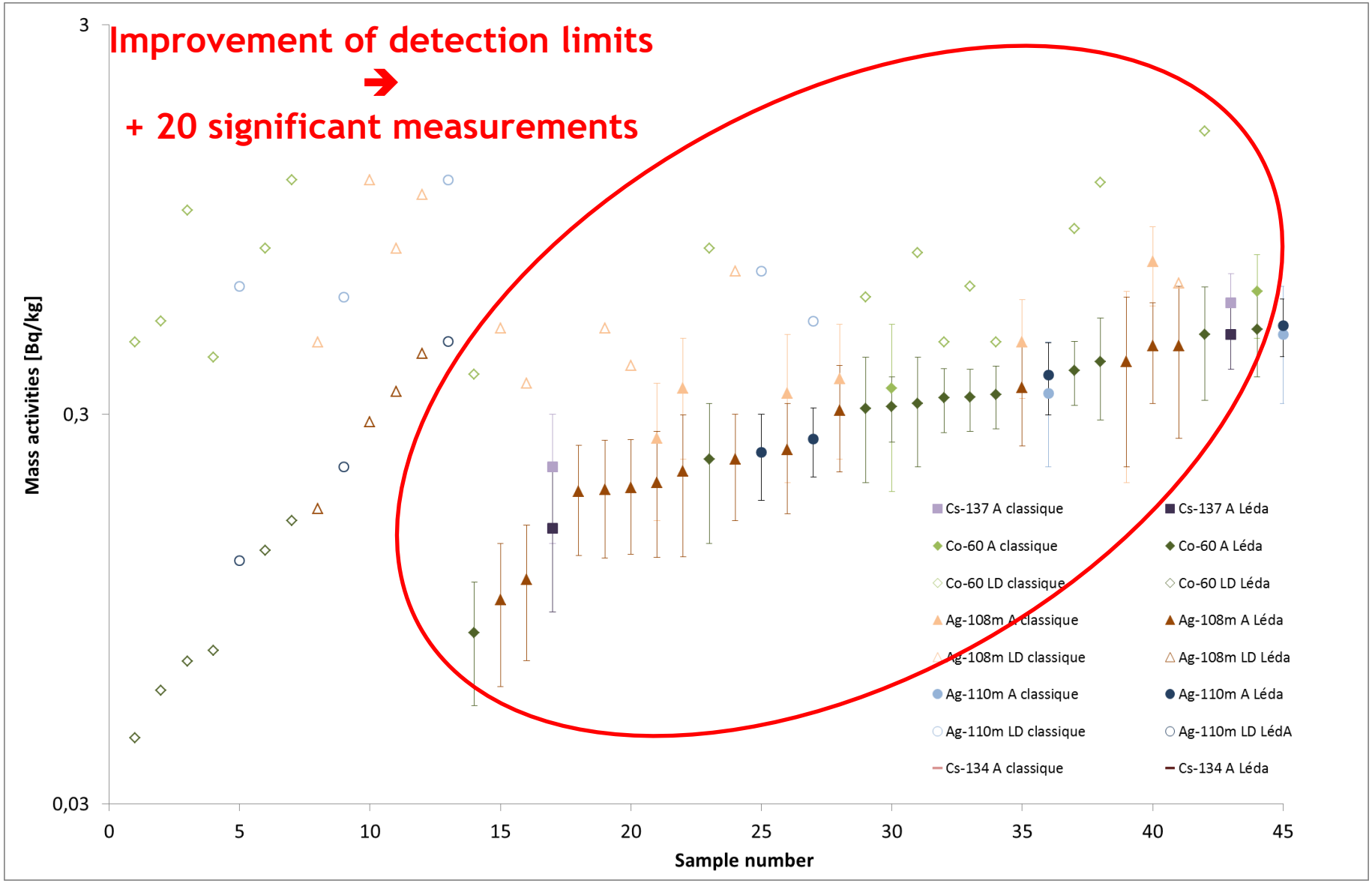

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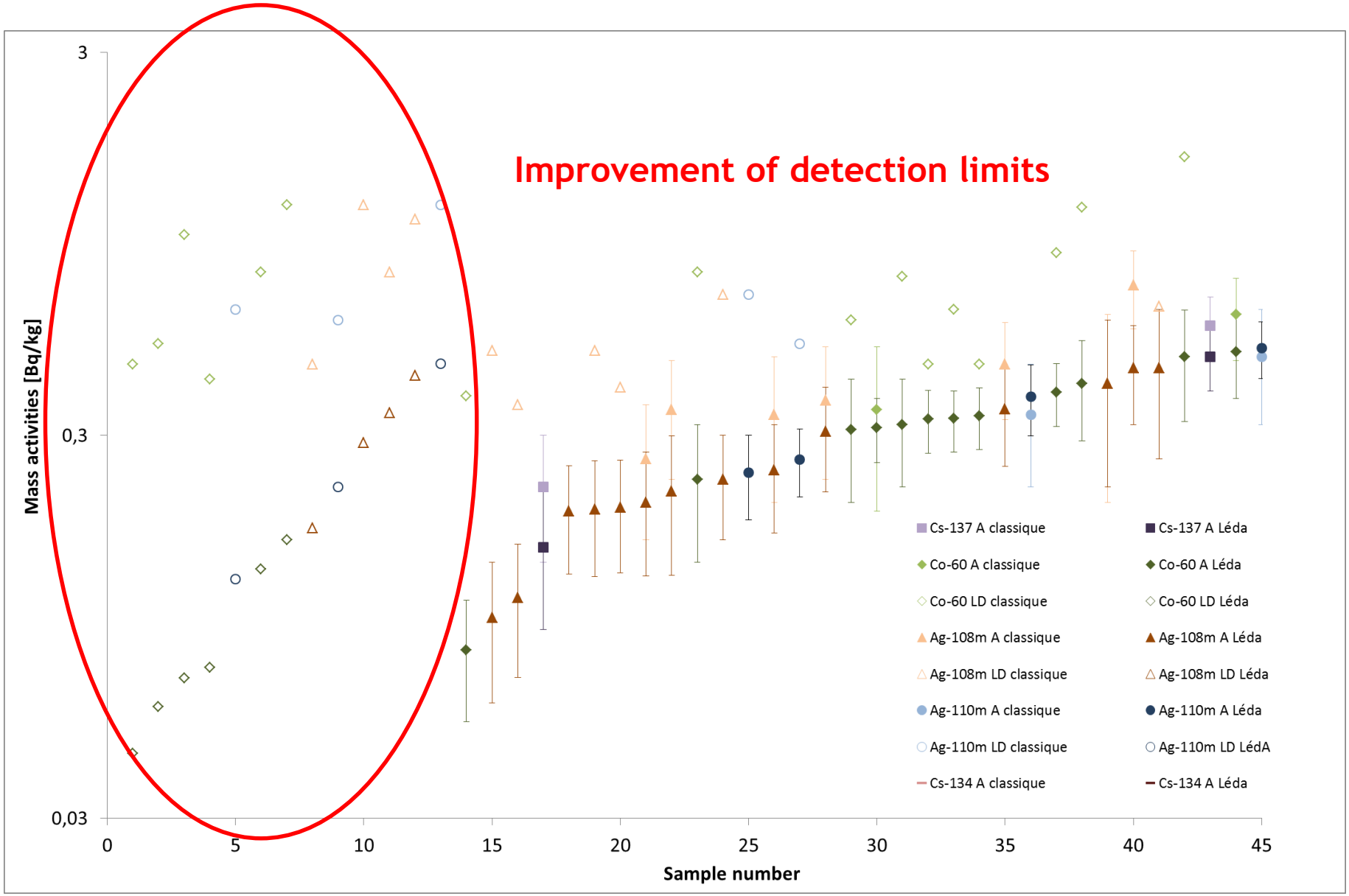

Conclusion

Leda a new coincident spectrometer

- **Designed** by Monte Carlo simulations
- Coincident measurement based on the analysis of the **list mode file**
- Calibration can be performed by MCNP_CP if **no standard source**

Performances compared to classical γ spectrometry

- More **precise** : reduction of the **uncertainties** by a factor of up to 3
- More **sensitive** :
  - improvement of the **detection limits** by a factor of up to 12
  - for all radionuclides
  - even for samples in which a radionuclide is present in major concentration
    - Compton continuum reduction for single emitters (e.g. ^{137}Cs, ^{241}Am)
    - coincidence matrix for coincidence emitters (e.g. ^{60}Co, ^{108m}Ag, ^{134}Cs)
- More **rapid** : gain on the **counting time** by a factor of 4

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Thank you for your attention.