Università degli Studi di Ferrara

Strategies for assessing high intensity radioactive sources with unknown shield

Master’s degree in Physics

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OUTLINE

- Scientific challenge
- Rationale
  - Indirect method
  - Photopeak method
- Monte Carlo (MC) input and constraints
- Detector efficiency
- Experimental conditions
- Analysis and results
- Conclusion and perspectives

SCIENTIFIC CHALLENGE

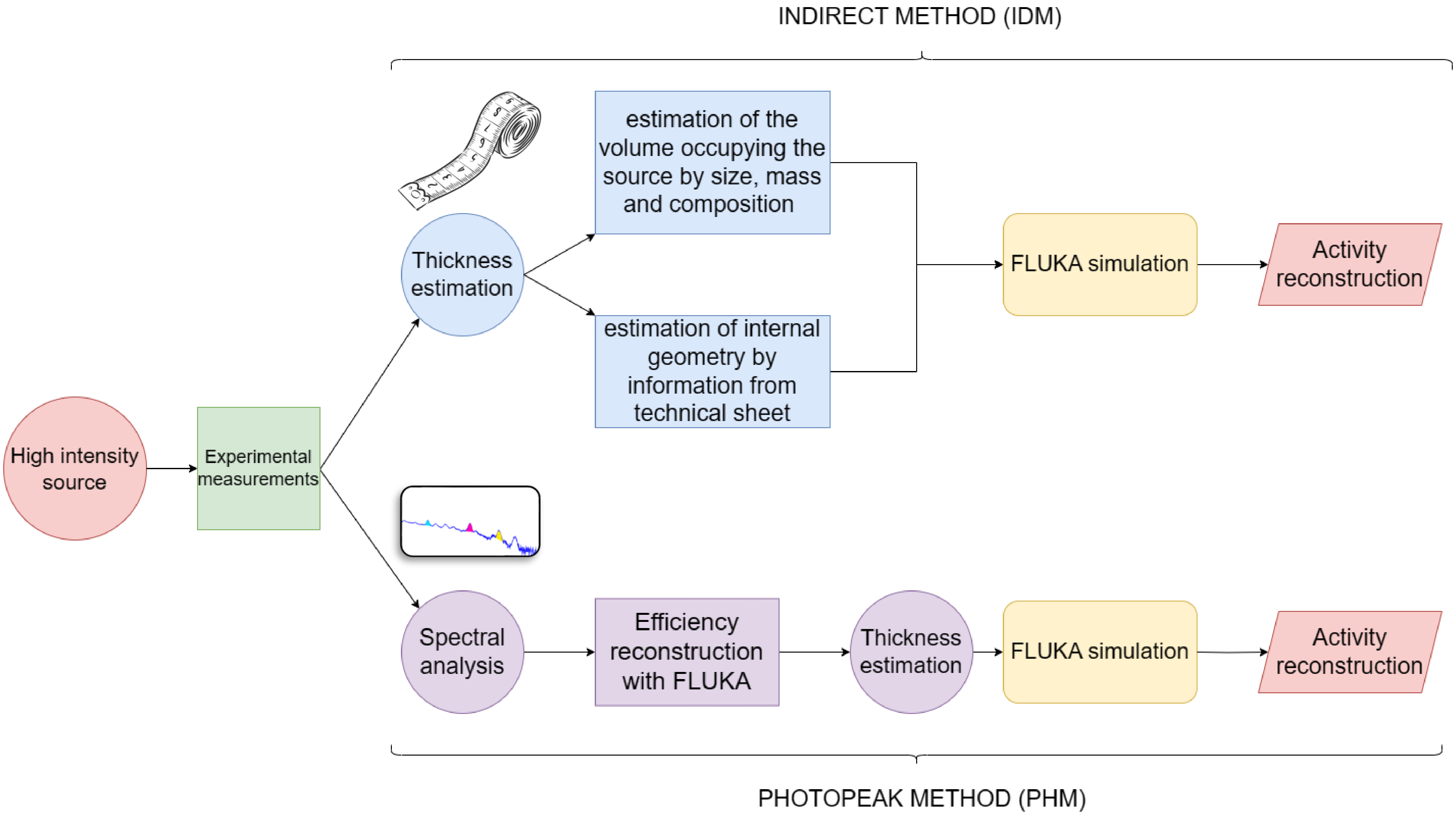
Unorganized site containing 7 unidentified radioactive sources with the following features:

- **high activity** sources such (e.g. ^{226}Ra, ^{60}Co, ^{137}Cs)
- containers with uncertain features, thus **unknown thickness**
- unknown activity, except for dubious past measurements, carried out unclearly and of unknown origin
- need to be **transported** to a final disposal site

How to estimate activity of intense sources with unknown shielding to protect operators from radiological risks?

ID	Radionuclide	Shielding material	Expected activity [Bq]
S2	^{226}Ra	Pb	$(1.06 \pm 0.16) \cdot 10^{6}$
S4	^{60}Co	Depleted Uranium	$(8.6 \pm 1.3) \cdot 10^{8}$
S5	^{137}Cs	Pb	$(7.70 \pm 1.16) \cdot 10^{10}$
S6	^{85}Kr	Stainless steel	$(4.4 \pm 0.7) \cdot 10^{6}$
S7	^{192}Ir and ^{137}Cs	Depleted Uranium	$(2.2 \pm 0.3) \cdot 10^{-3}$
S9	^{226}Ra	Pb	$(5.3 \pm 0.8) \cdot 10^{7}$
S10	^{226}Ra	Pb	/

WORKFLOW: INDIRECT METHOD (IDM) AND PHOTOPEAK METHOD (PHM)



INDIRECT METHOD (IDM)

High intensity source

Experimental measurements

Thickness estimation

estimation of the volume occupying the source by size, mass and composition

estimation of internal geometry by information from technical sheet

FLUKA simulation

Activity reconstruction

Spectral analysis

Efficiency reconstruction with FLUKA

Thickness estimation

FLUKA simulation

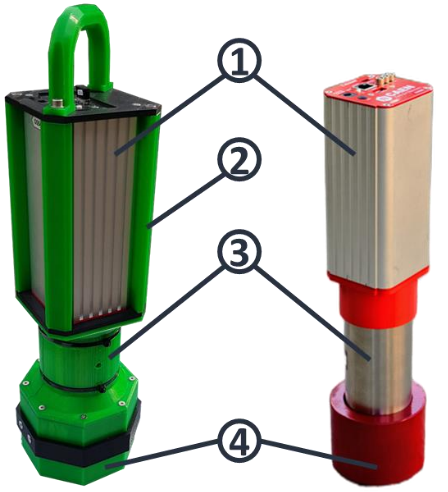
Activity reconstruction

PHOTOPEAK METHOD (PHM)

DETECTOR

- 2’’ $CeBr_3$ scintillation detector
- Volume 103 cm^3
- Resolution of 4.1% at 661.7 keV (^{137}Cs)
- 19 ns photon mean life

$$R[\%] = \frac{FWHM}{E}$$

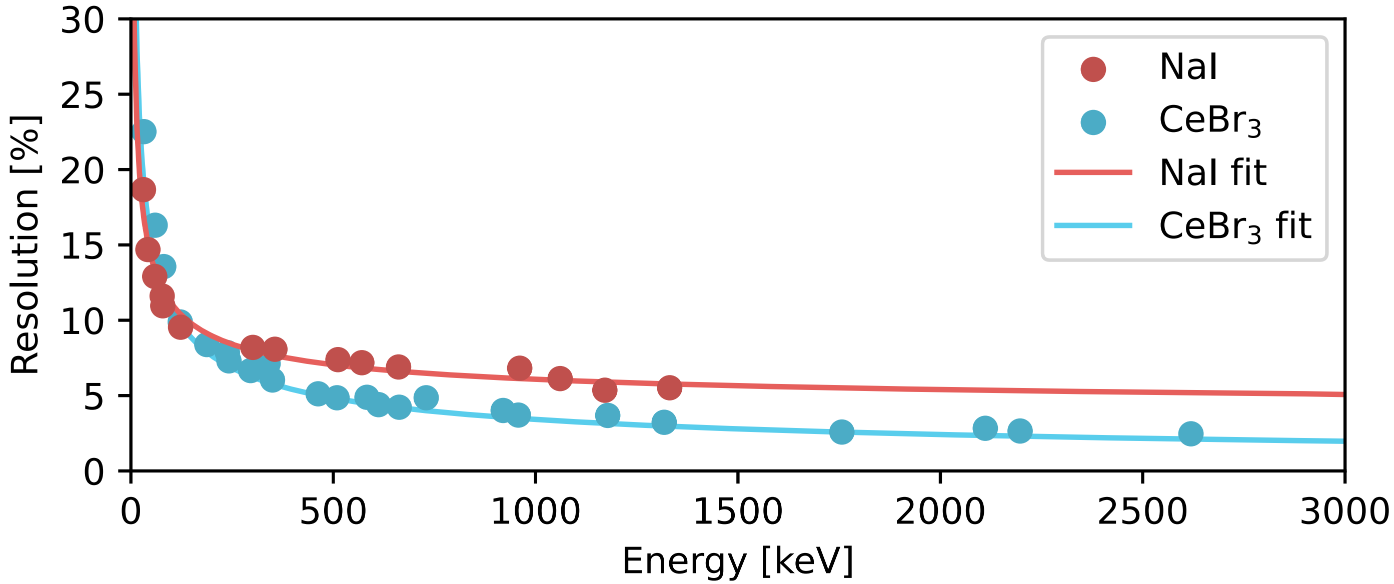


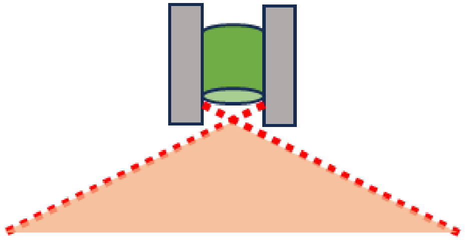
① Multi Channel Analyzer

② PLA coating

③ $CeBr_3$ scintillator

④ Lead shielding



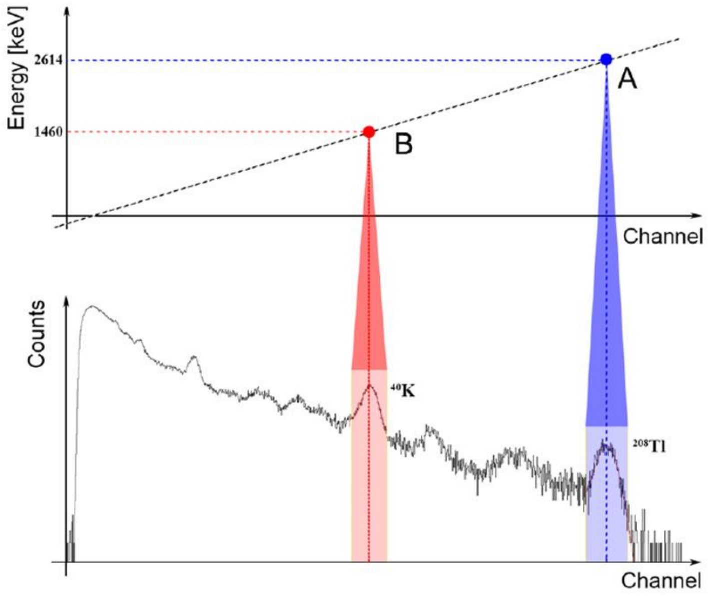


- Partially collimated with a 1.25 cm Pb shield (~160° vision angle) attenuating the background by the 60%
- Multi Channel Analyzer with Bluetooth connection

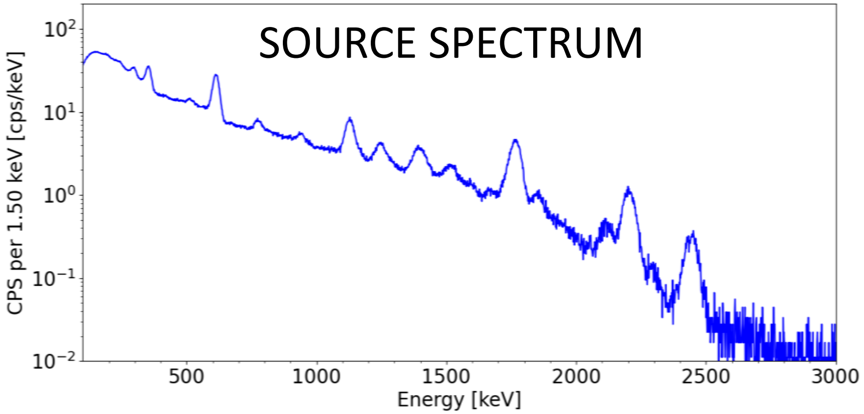
SPECTRAL ANALYSIS

Energetic calibration

- Establishing the relationship between the acquisition channel and the energy



Background subtraction



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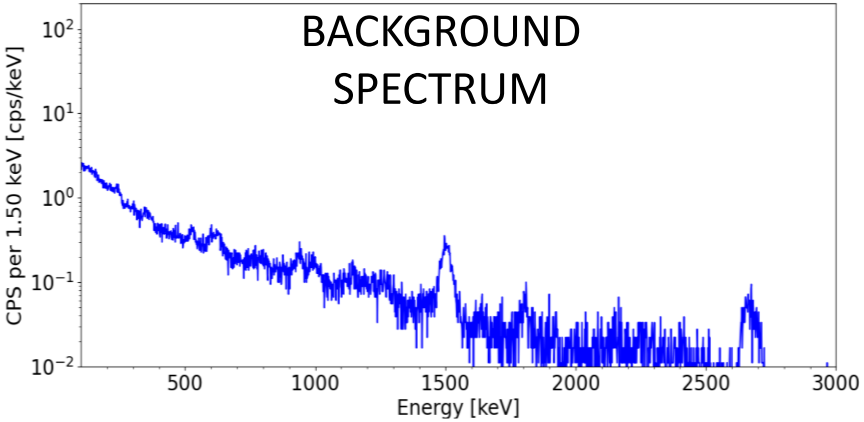
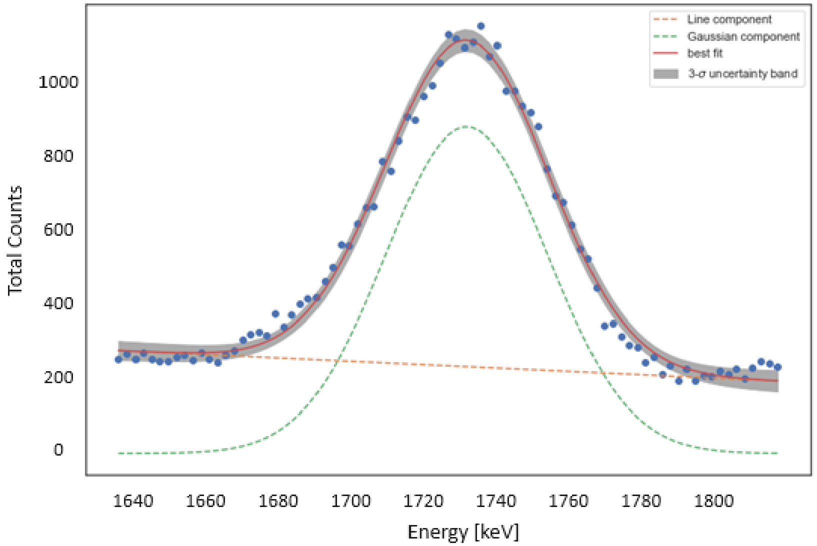
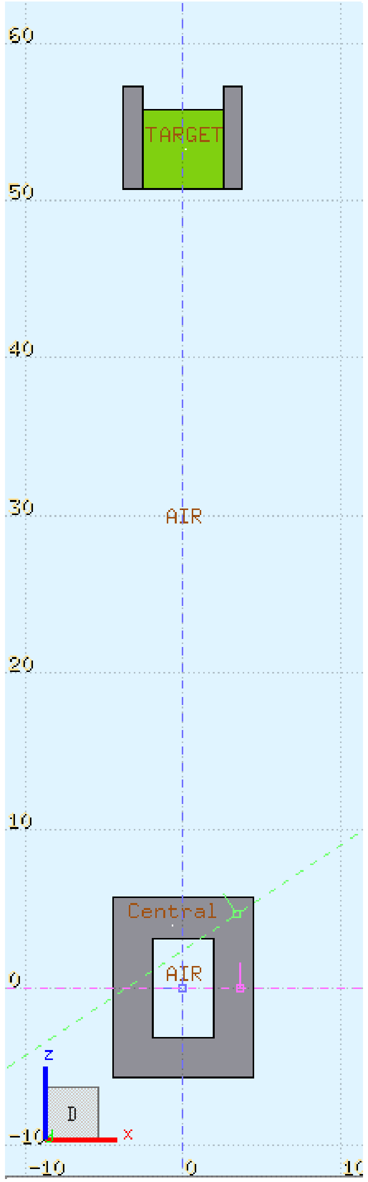


Photopeak fit

- Combination of gaussian and linear fit
- Net count per second calculation **N_{net} [cps]**



MONTE CARLO SIMULATIONS WITH FLUKA

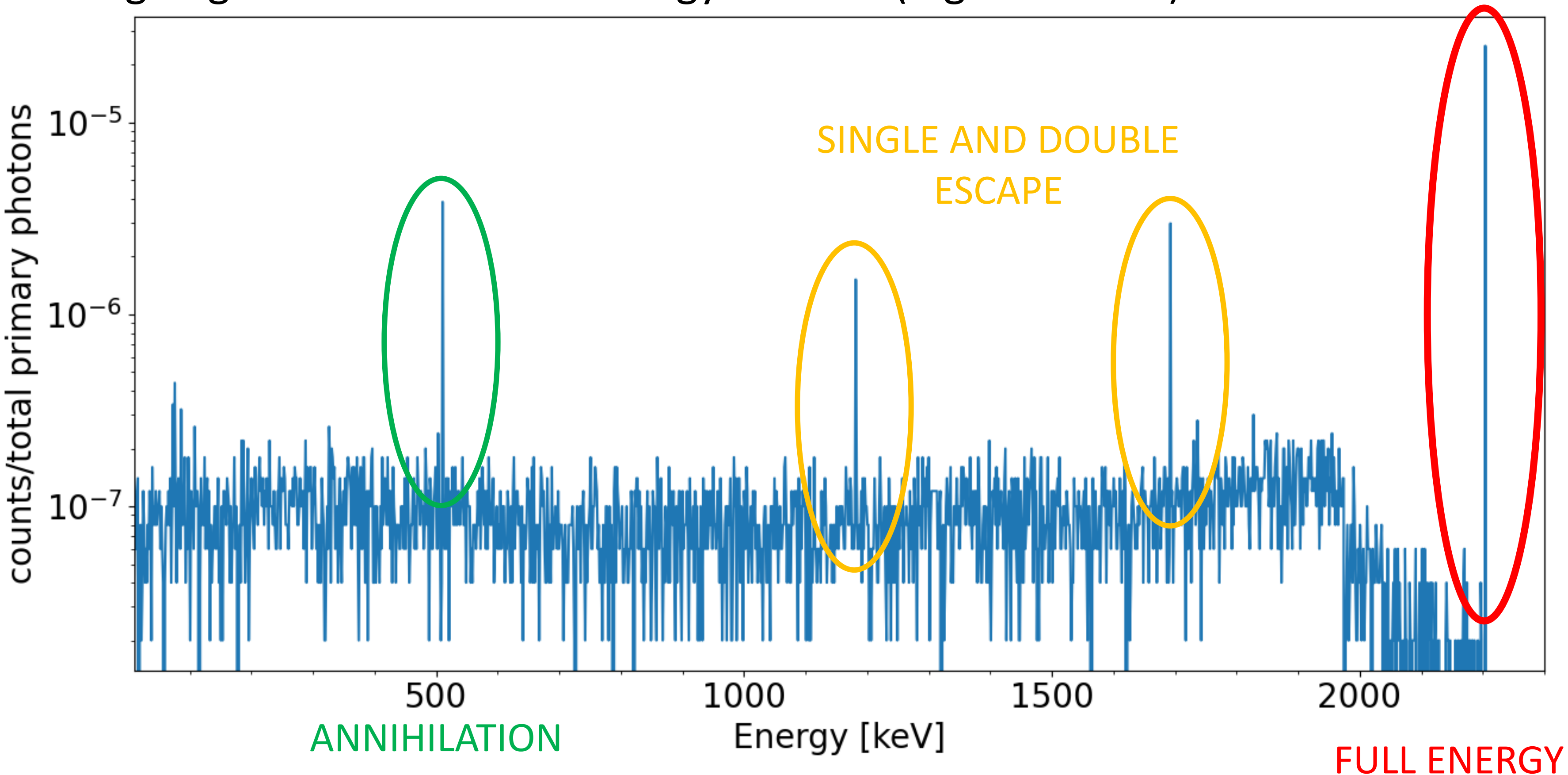


- FLUKA is a MC software for the simulation of transport and interaction of particles with matter

INPUT

- Reconstruction of the experimental setup's geometry, including the detector, shielding, distances, sources, etc.
- The source activity was modelled by specifying the number of photons and assigning a monochromatic energy to them (e.g. **2204 keV**)

OUTPUT



ANNIHILATION LINE

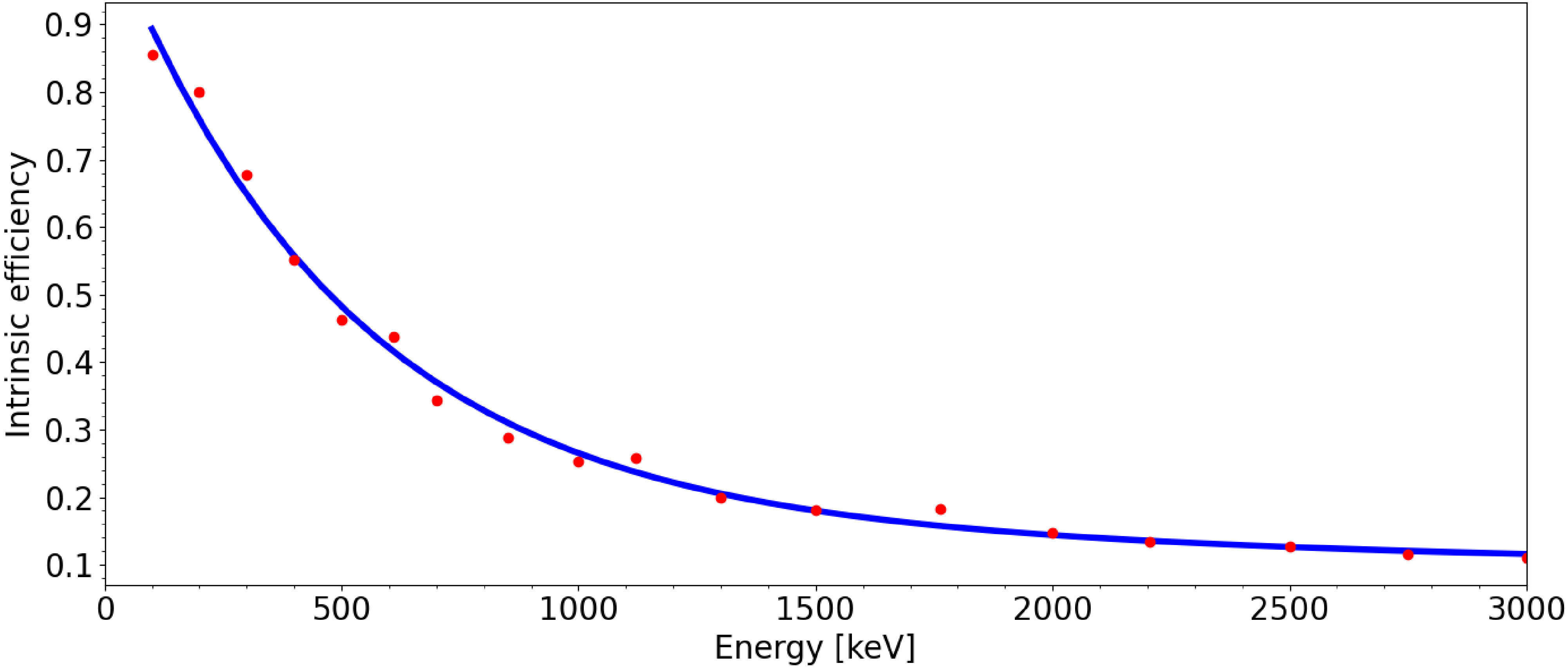
FULL ENERGY PEAK

INTRINSIC EFFICIENCY CURVE

- The **intrinsic efficiency** is related to the detector's ability to record incoming photons
- Simulation of a detector-source setup in a vacuum in the energy range **100-3000 keV**
- The function applied for the fitting as suggested in [1]

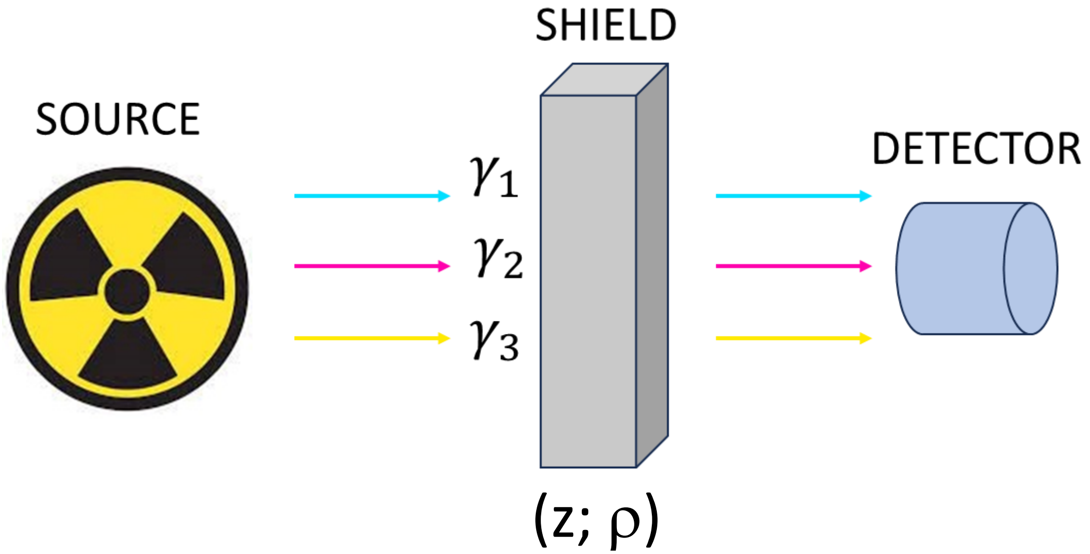
$$\varepsilon_{\mathrm{intrinsic}} = \mathrm{Photons}_{\mathrm{recorded}} / \mathrm{Photons}_{\mathrm{incoming}}$$

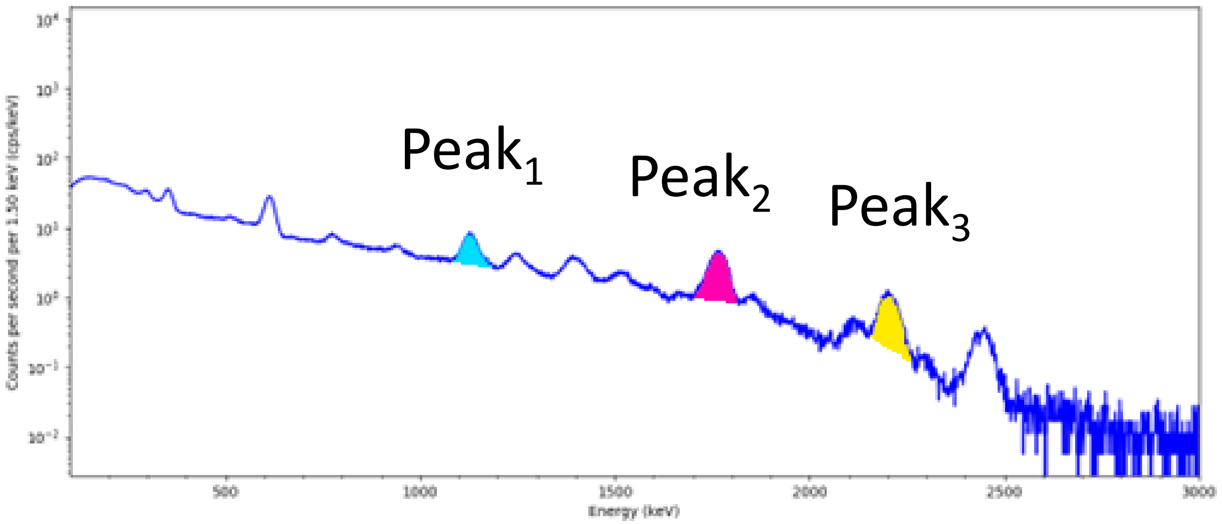
$$f(E) = A_1 e^{-A_2 E} + A_3 e^{-A_4 E}$$



[1] Knoll, G.F., *Radiation detection and measurement*. 2010: John Wiley & Sons.

PHOTOPEAK METHOD FOR ESTIMATING THE THICKNESS





- Consider a source emitting **at least 2 gammas** γ_1, γ_2 with energies E_1 (keV), E_2 (keV) and intensities I_1, I_2
- A uniform shield with density ρ (g/cm^3) and **thickness z (cm)**
- Linear attenuation coefficient λ_1(1/cm), λ_2(1/cm)
- A detector recording the two photopeaks cps Peak$_1$, Peak$_2$
- With intrinsic efficiencies $\varepsilon_1^{\text{int}}, \varepsilon_2^{\text{int}}$

$$\frac{\text{Peak}_1}{\text{Peak}_2} = R_{1,2} = \frac{\varepsilon_1{}^{int} I_1}{\varepsilon_2{}^{int} I_2} e^{\boxed{z}(\lambda_2 - \lambda_1)}$$

Knowing $\varepsilon_1^{\text{int}}, \varepsilon_2^{\text{int}}$ one can calculate: $z = \frac{1}{\lambda_2 - \lambda_1} \ln\left(R_{1,2} \cdot \frac{\varepsilon_1{}^{int} I_2}{\varepsilon_2{}^{int} I_1}\right)$

Estimation of z studying photopeak ratio

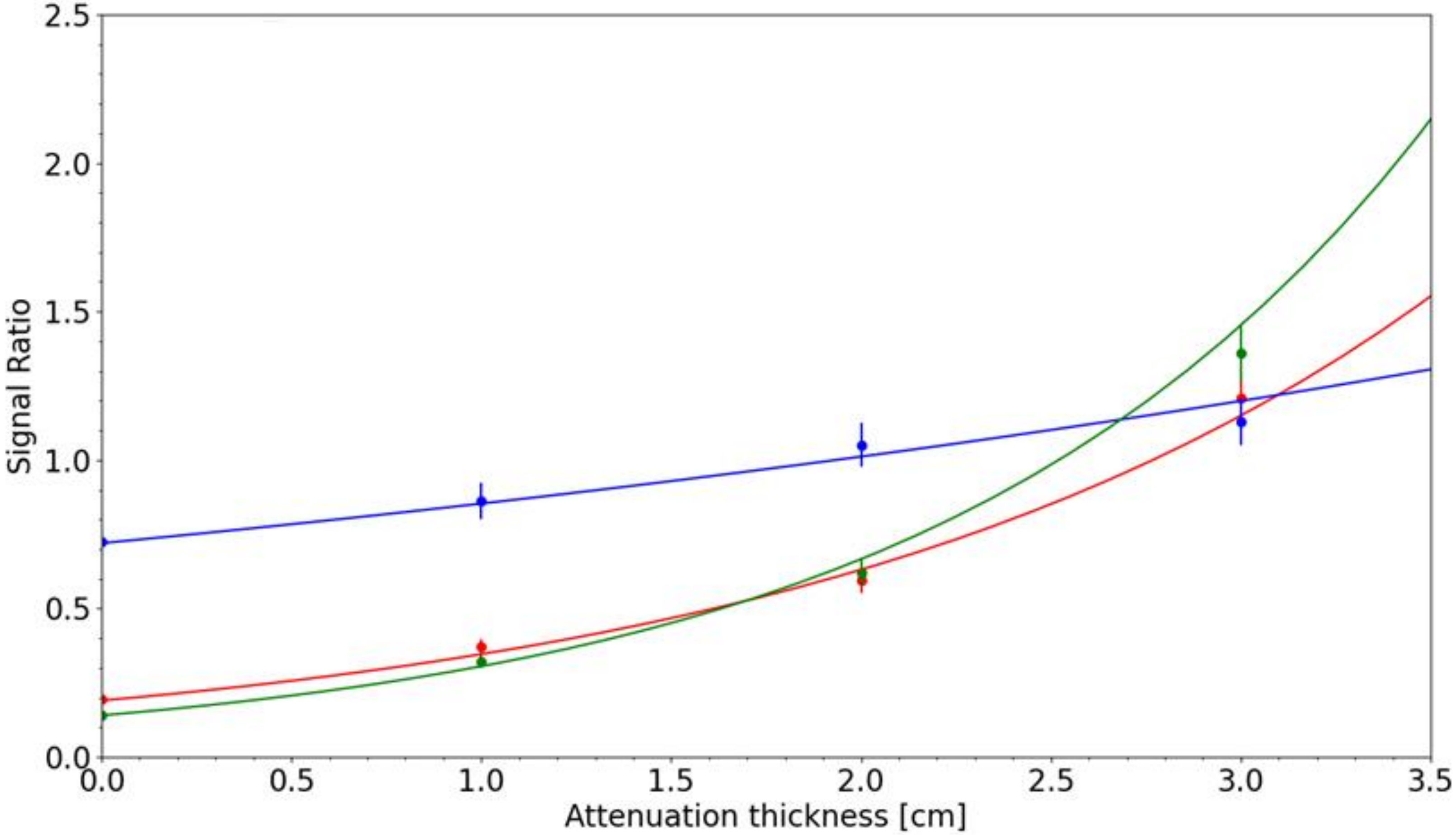
SIGNAL RATIO AS A FUNCTION OF ATTENUATION (^{226}Ra): analytical vs MC

- Analytical calculation of the ratios R1, R2, R3 as a function of the attenuation
- Simulation with FLUKA of the same ratios at 4 different thickness z = 0, 1, 2, 3 cm

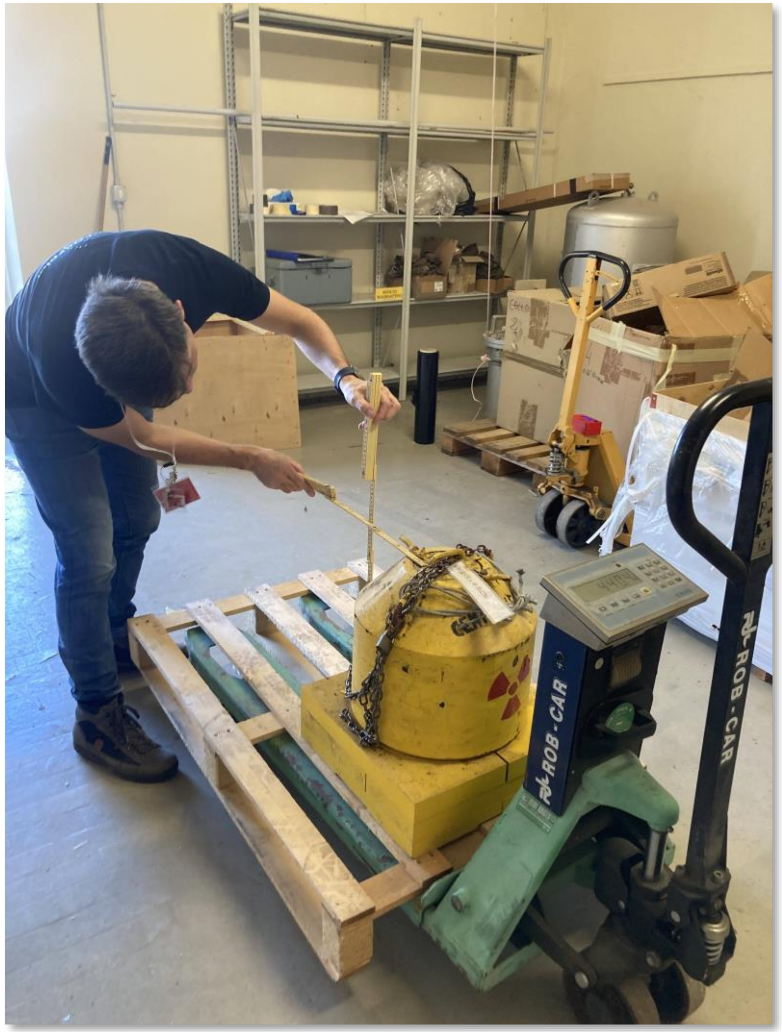
R1 = S2 (E =1764 keV) / S1 (E = 609 keV)

R2 = S3 (E =2204 keV) / S2 (E = 1764 keV)

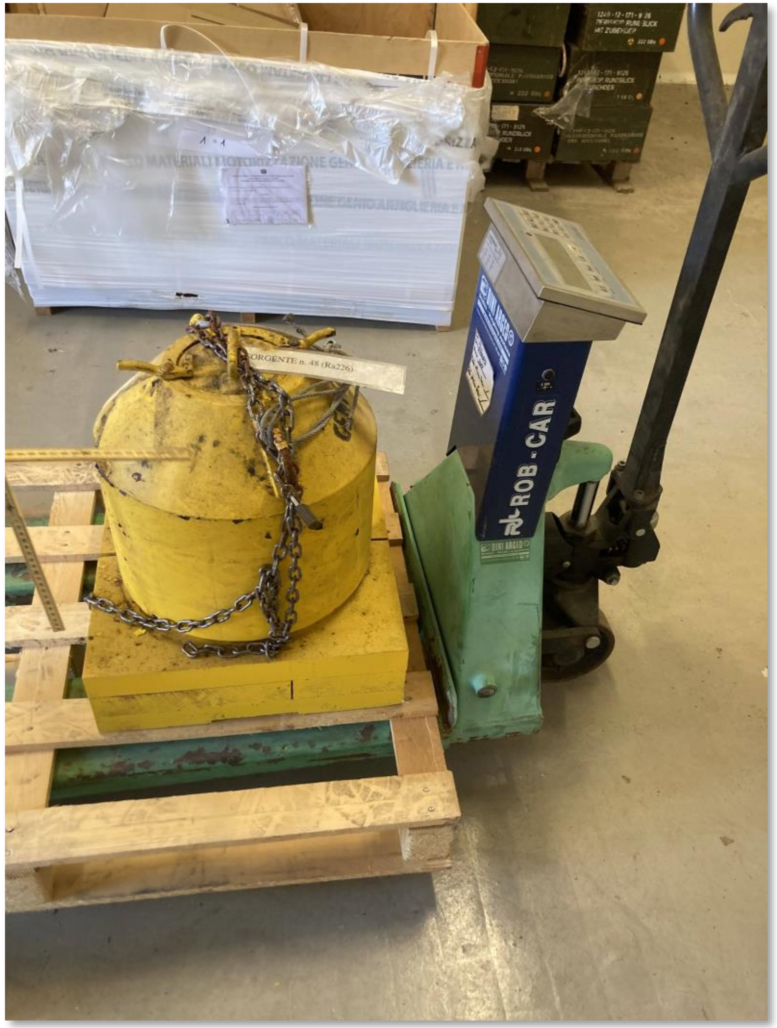
R3 = S3 (E = 2204 keV) / S1 (E = 609 keV)



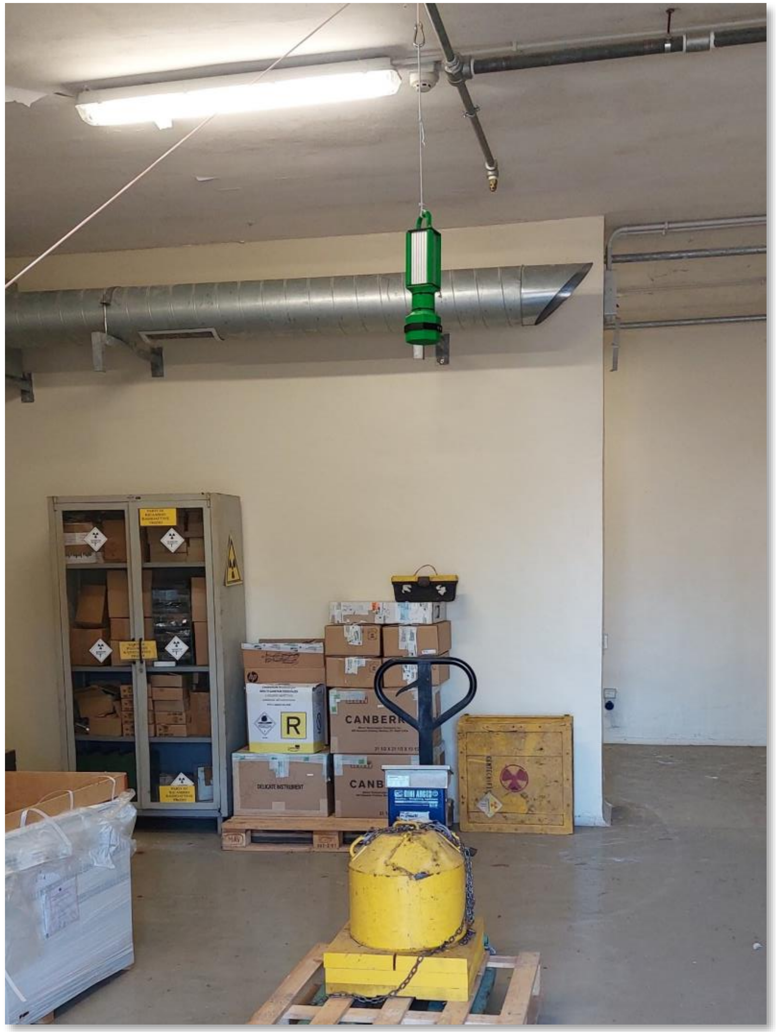
EXPERIMENTAL CONDITIONS



Measurement of the external dimensions of the container



Measurement of the mass of the container

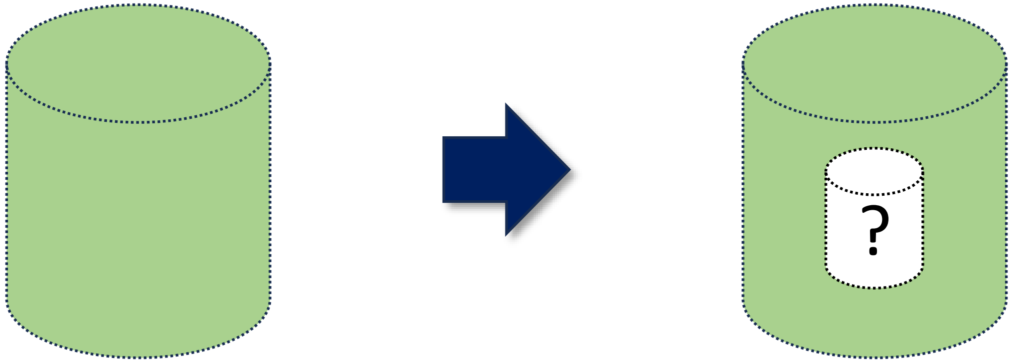


Measurement of the detector-source distance

INDIRECT METHOD FOR ESTIMATING THE THICKNESS

Full cylinder

measured cylinder

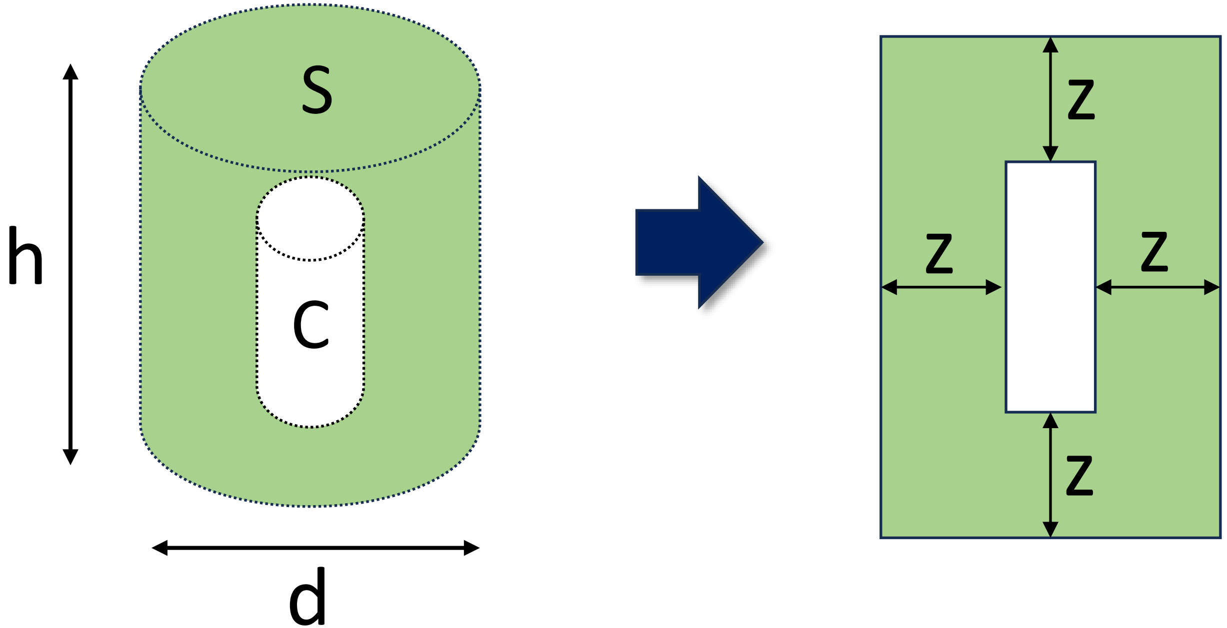


$$V_C = \pi \left(\frac{d - 2z}{2} \right)^2 (h - 2z)$$

- Estimate the volume of air inside
- Estimate the thickness z

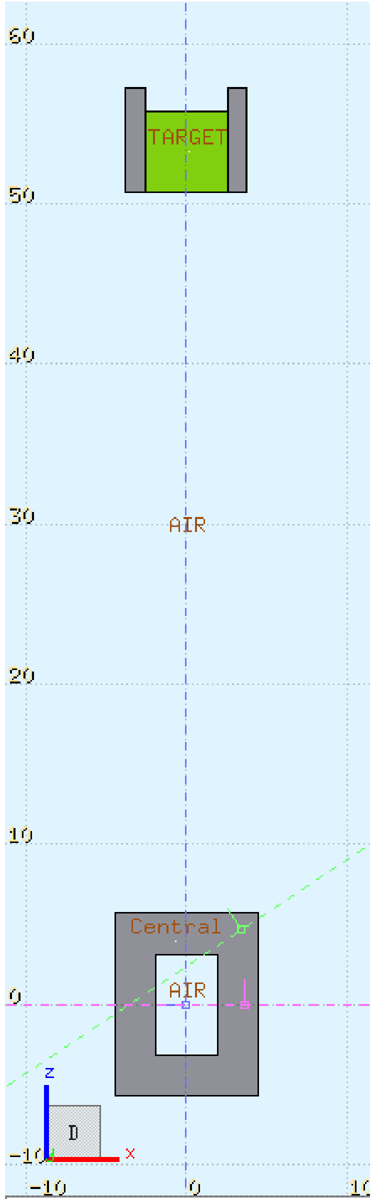
Hypothesis of uniform shielding

Centered air chamber



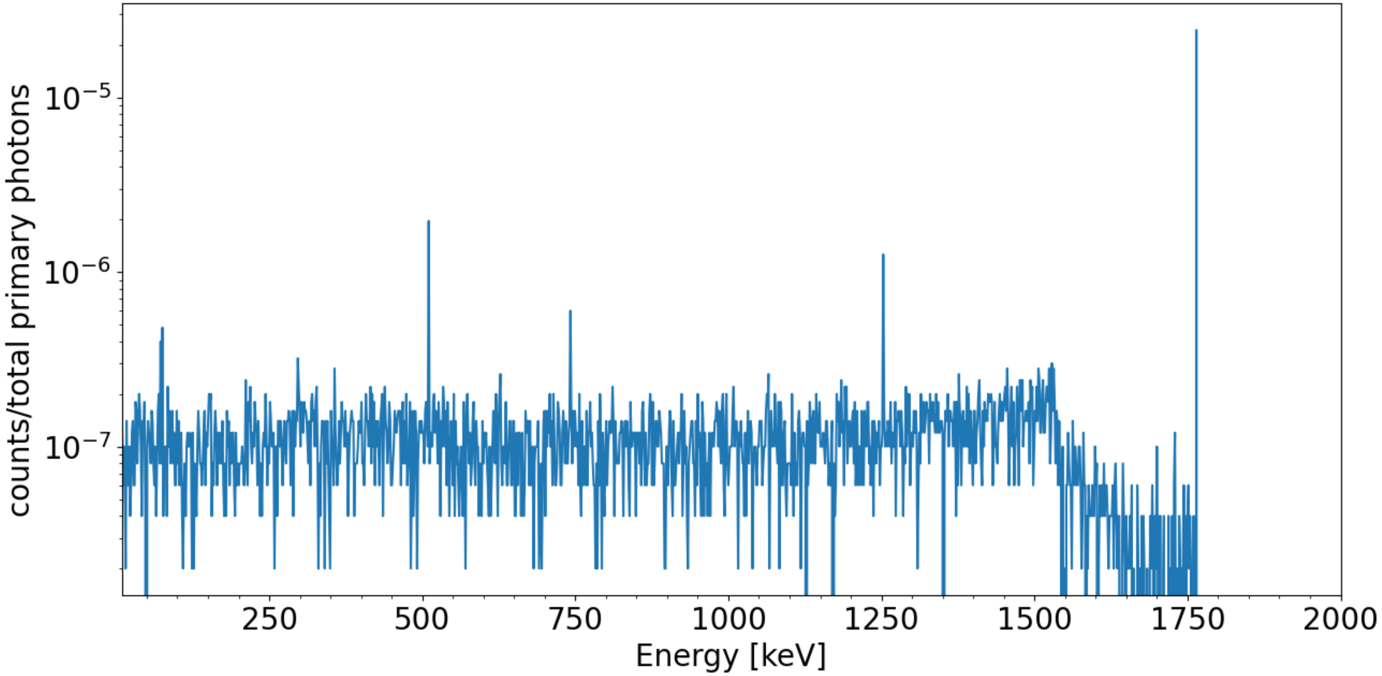
Estimation of z measuring mass and dimensions.

MC SIMULATION ANALYSIS OF S9 (^{226}Ra)



Simulation steps:

- Reconstruction of the Pb cylinder with m = 5 kg and a centered air chamber inside
- Generation of $2 \cdot 10^7$photons with energy **1764 keV**
- The run lasted for 20 minutes.
- This resulted in the following spectrum.



EXPERIMENTAL AND MONTE CARLO SYNERGY

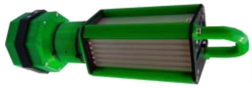
EXPERIMENT

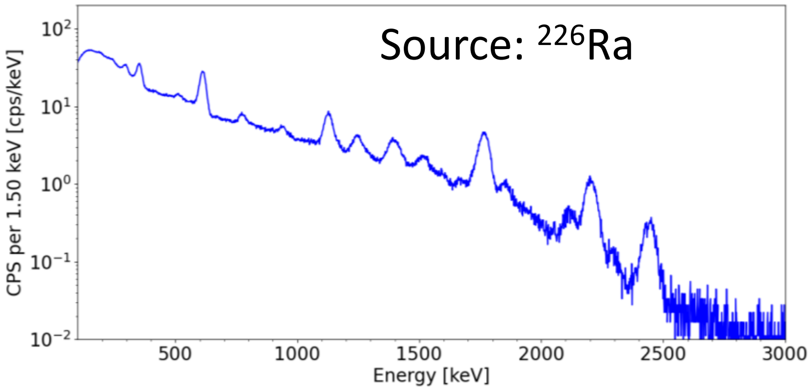
SOURCE

Activity A

$\gamma, E_\gamma, I_\gamma$

DETECTOR





$$N_{net}\,[cps] = A \cdot \varepsilon_{peak}$$

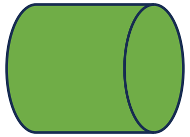
ε_{peak} [cps/Bq] encapsulate information about: geometry, attenuation, E_γ, I_γ

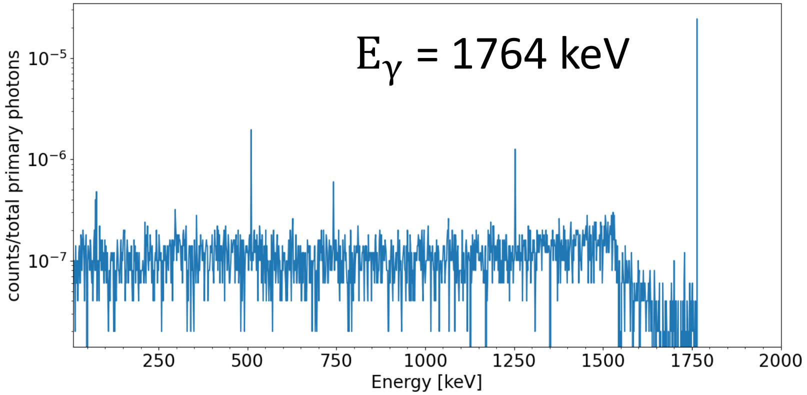
MONTE CARLO

MONOCHROMATIC PHOTON BEAM

N_γ, E_γ

SCORING





FLUKA output:

$$F(E) = \frac{\text{Counts}(E)}{N_\gamma}$$

$$\varepsilon_{peak} = F(E_\gamma) \cdot I_\gamma$$

$$\text{Activity [Bq]} = \frac{N_{Net}\,[cps]}{\varepsilon_{peak}\ [cps/Bq]}$$

ESTIMATION OF THICKNESS AND ACTIVITY WITH IDM

ID	Radionuclide	Shielding material	Thickness with IDM [cm]	Expected Activity [Bq]	Experimental Activity [Bq]
S2	^{226}Ra	Pb	17.0 ± 1.0	$(1.09 \pm 0.16) \cdot 10^{10}$	$(4.98 \pm 1.76) \cdot 10^{9}$
S4	^{60}Co	Depleted Uranium	9.5 ± 1.0	$(8.6 \pm 1.3) \cdot 10^{8}$	$(2.30 \pm 0.80) \cdot 10^{9}$
S5	^{137}Cs	Pb	8.5 ± 1.0	$(7.70 \pm 1.16) \cdot 10^{10}$	$(2.15 \pm 1.23) \cdot 10^{10}$
S6	^{85}Kr	Stainless Steel	0.5 ± 0.2	$(4.4 \pm 0.7) \cdot 10^{6}$	$(4.15 \pm 2.22) \cdot 10^{7}$
S7	^{137}Cs	Depleted Uranium	3.8 ± 1.0	$(3.7 \pm 0.6) \cdot 10^{9}$	$(1.95 \pm 1.05) \cdot 10^{10}$
	^{192}Ir	Depleted Uranium	3.8 ± 1.0	$(2.2 \pm 0.3) \cdot 10^{-3}$	<MDA
S9	^{226}Ra	Pb	2.6 ± 1.0	$(5.3 \pm 0.8) \cdot 10^{7}$	$(2.44 \pm 1.29) \cdot 10^{7}$
S10	^{226}Ra	Pb	5.1 ± 1.0	/	$(1.93 \pm 1.07) \cdot 10^{8}$

- Almost all measurements are within **2 sigma compatibility**.
- Iridium is below the MDA, explaining why it was not detected.
- The measurement furthest from the expected value is S5 (3.2 sigma), a complex situation where the detector was not even axially aligned with the source.
- The best results were obtained for S4 and S7, for which technical sheets were available.
- S10 does not have an expected activity value.

ESTIMATION OF THICKNESS AND ACTIVITY WITH PHM (S9)

ID	Peak 1		Peak 2		$\frac{\boldsymbol{Peak\ 1}}{\boldsymbol{Peak\ 2}}$	Thickness [cm]
	Energy [keV]	Net cps	Energy [keV]	Net cps		
S9	1764	185.0 ± 2.1	1120	189.7 ± 2.2	0.98 ± 0.02	$2.18^{+0.09}_{-0.09}$
	2204	46.1 ± 0.9	1764	185.0 ± 2.1	0.25 ± 0.01	$3.08^{+0.70}_{-0.69}$
	2204	46.1 ± 0.9	1120	189.7 ± 2.2	0.24 ± 0.01	$1.26^{+0.11}_{-0.13}$

- Employed weighted average to minimize error.
- Achieved a **smaller relative uncertainty**, approximately 5%.
- Attained **compatibility within 0.8 sigma** of the expected activity.

	Expected	Indirect Method	Photopeak method
Thickness [cm]	\	2.6 ± 1.0	1.9 ± 0.1
Activity [Bq]	$(5.3 \pm 0.8)\ 10^7$	$(2.4 \pm 1.3)\ 10^7$	$(4.6 \pm 0.2)\ 10^7$

ESTIMATION OF THICKNESS AND ACTIVITY WITH PHM (S10)

ID	Peak 1		Peak 2		$\frac{\boldsymbol{Peak\ 1}}{\boldsymbol{Peak\ 2}}$	Thickness [cm]
	Energy [keV]	Net cps	Energy [keV]	Net cps		
S10	1764	72.9 ± 1.4	1120	51.5 ± 1.3	1.42 ± 0.04	$4.43^{+0.17}_{-0.18}$
	2204	20.2 ± 1.2	1764	72.9 ± 1.4	0.27 ± 0.01	$1.31^{+0.50}_{-0.29}$
	2204	72.9 ± 1.4	1120	51.5 ± 1.3	0.39 ± 0.02	$3.59^{+0.27}_{-0.29}$

- Employed weighted average to minimize error.
- Achieved a **smaller relative uncertainty**, approximately 10%.
- Attained **compatibility within 2 sigma** of the IDM activity.

	Expected	Indirect Method	Photopeak method
Thickness [cm]	\	5.0 ± 1.0	3.5 ± 0.2
Activity [Bq]	\	$(1.9 \pm 1.1)\ 10^{8}$	$(2.8 \pm 0.3)\ 10^{7}$

CONCLUSIONS

How to estimate activity of intense sources with unknown shielding to protect operators from radiological risks?

MC simulations have been developed that have allowed us to reconstruct the efficiency curve of the detector that is one fundamental piece for the analysis of experimental results.

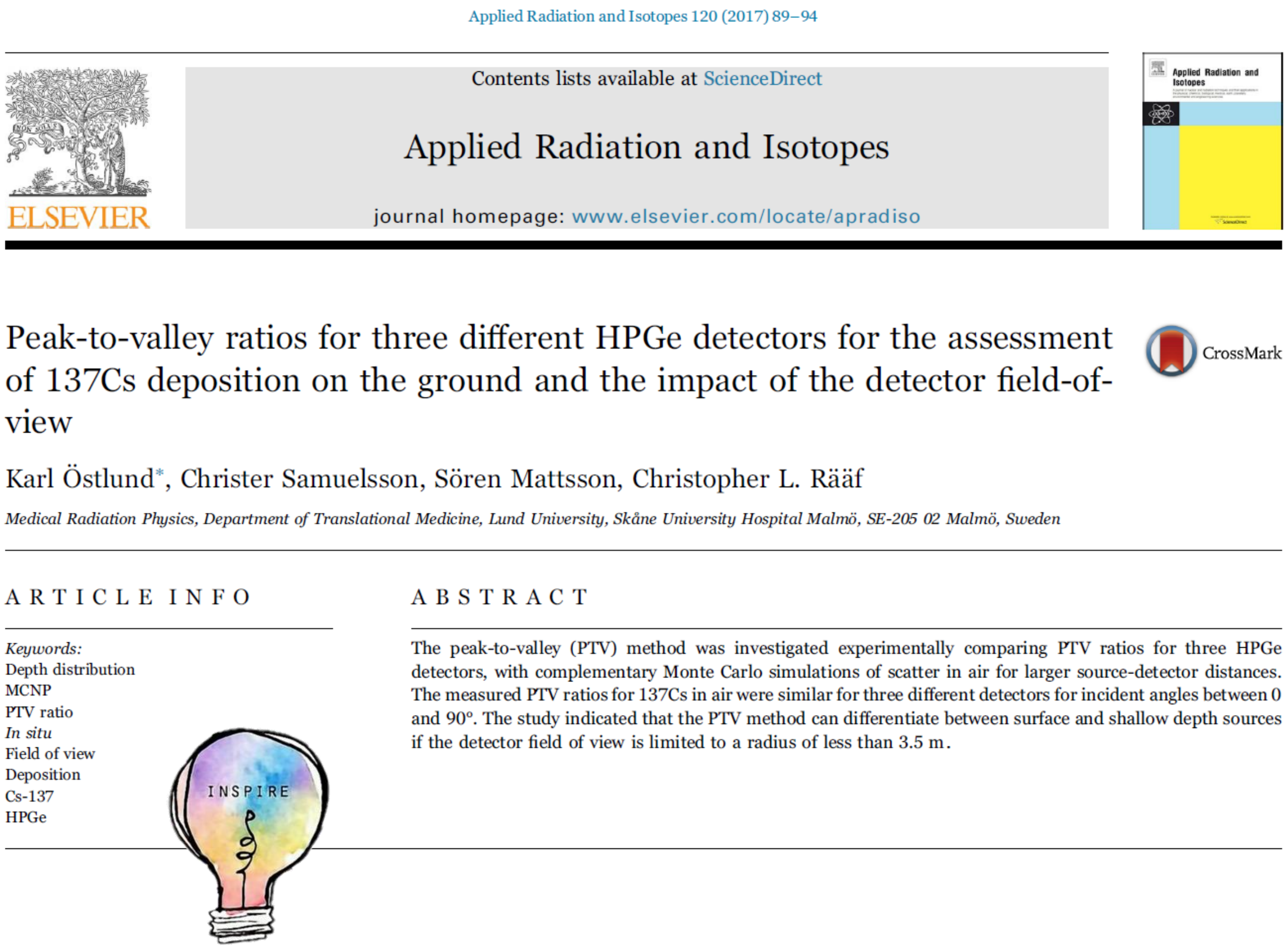
The method allowed to estimate the thickness of an unknown shielding (3.5±0.2 cm) and the activity (28±3 MBq) of an unknown source of ^{226}Ra.

A method has been developed that, considering fixed thickness, detector efficiency and analyzing the relative relationships between the intensity of 3 different photopeaks, has allowed to estimate the intensity (46±2 MBq) of a ^{226}Ra compatible to 1 sigma with the expected value.

Based on indirect measurements of the thickness of the shielding, the activities of 7 high intensity sources were reconstructed at less than 2 sigma compatible with the expected values.

PERSPECTIVES

- PHM cannot be applied in the case of sources emitting only a single gamma line, like ^{137}Cs.
- Advance the study and development of the "**peak-to-valley**" method as an evolution of the photopeak approach.
- Utilize the "peak-to-valley" method to analyze **spectral deformation** due to attenuation, enabling more accurate inference of shielding thickness.



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Peak-to-valley ratios for three different HPGe detectors for the assessment of 137Cs deposition on the ground and the impact of the detector field-of-view

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ABSTRACT

The peak-to-valley (PTV) method was investigated experimentally comparing PTV ratios for three HPGe detectors, with complementary Monte Carlo simulations of scatter in air for larger source-detector distances. The measured PTV ratios for 137Cs in air were similar for three different detectors for incident angles between 0 and 90°. The study indicated that the PTV method can differentiate between surface and shallow depth sources if the detector field of view is limited to a radius of less than 3.5 m.

THANK YOU FOR THE ATTENTION

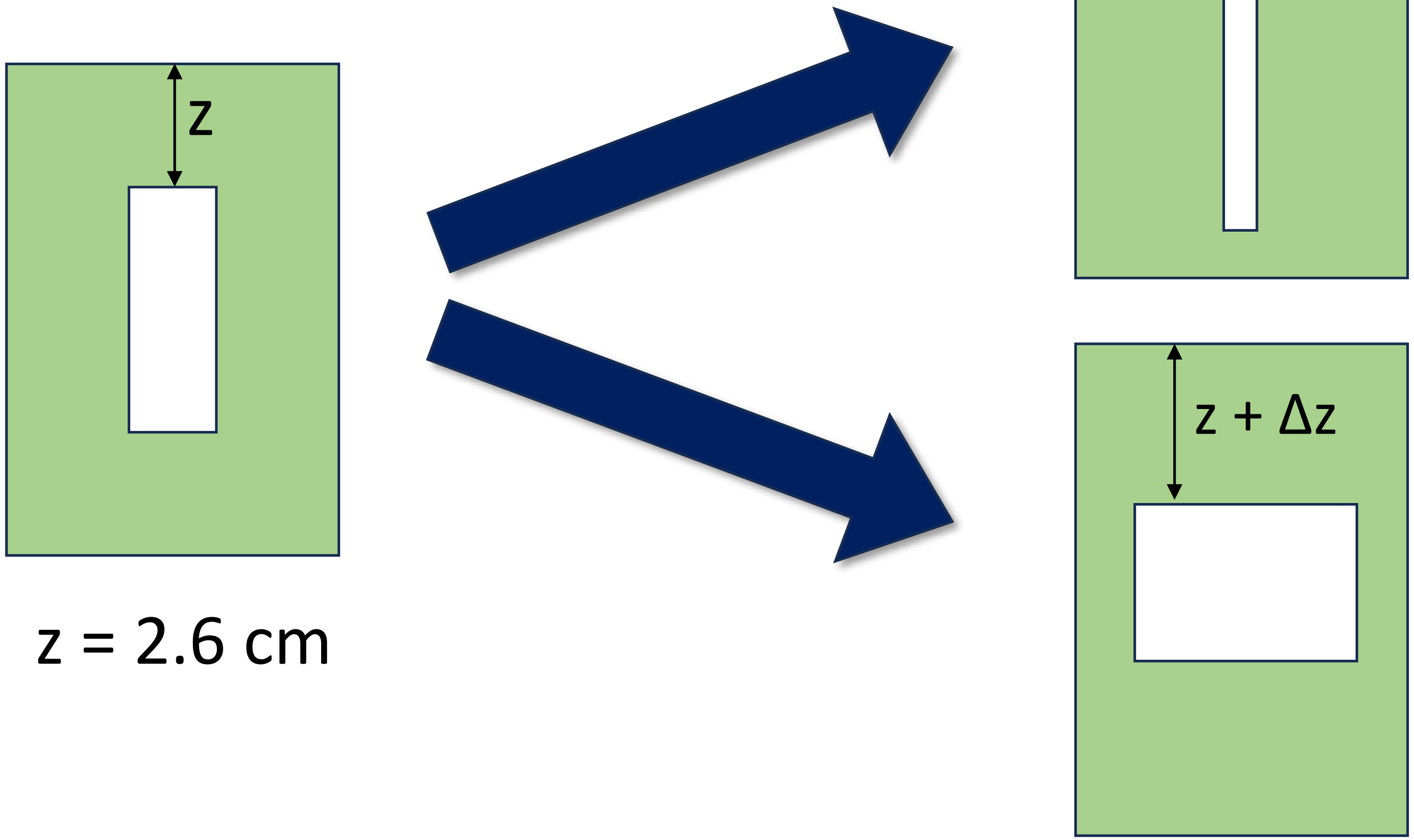
MC VALIDATION

- The Monte Carlo simulation underwent a rapid validation process through a measurement from a ^{137}Cs source, with a certified activity of 128 kBq and an uncertainty of 1%, placed at a 1-meter distance from the detector.
- The activity reconstructed from the net counts at the 662 keV peak and the full-energy peak efficiency simulated with FLUKA, resulted in an estimated activity that is compatible within 1.5 sigma of the certified value.

Net cps	ε_{fep}	Activity certified [Bq]	Activity Reconstructed [Bq]
7.3±0.1	$(5.24\pm0.30)\cdot10^{-5}$	$(1.28\pm0.01)\cdot10^{5}$	$(1.39\pm0.08)\cdot10^{5}$

ESTIMATION OF INTERNAL CHAMBER WITH UNCERTAINTY (S9)

ID	h [cm]	d [cm]	Weight [kg]
S9	11.5 ± 0.1	9.0 ± 0.1	5.0 ± 0.1



z = 2.6 cm

- The uniform shielding hypothesis could be broken if the **shape of the chamber changes** while keeping the chamber volume constant.

- ***conservative approach*** to estimate the correct activity order of magnitude

ΔZ = 1 cm

Z = 2.6 ± 1.0 cm