Development of a new compact photon/electron detector for radioxenon measurement: The Mobile Analyzer of Radioactive Gases OuTflows (MARGOT)

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CONTEXT: Monitoring atmospheric concentrations of radioxenons is relevant to provide evidence of atmospheric or underground nuclear weapon tests. However, when the design of the International Monitoring System (IMS) network of the Comprehensive Test-Ban Treaty (CTBT) was set up, the impact of industrial releases was not perceived. It is now well-known that industrial radioxenon signature can interfere with that of nuclear tests. Therefore, there is a crucial need to characterize atmospheric distributions of radioxenons from industrial sources and establish radioxenon civilian isotopic ratio signatures for event discrimination purposes.

The CEA has recently developed a detection system that will be integrated into the IMS: the SPALAX-NG [1], working in coincidence mode (HPGe/Si). In its continuous process of R&D, the CEA has start to prospect on a new generation of detection system based on NaI(Tl)/Si coincidence mode. The main goal of this development is to keep the advantage of high resolution coincidence detection system and to tend to a miniaturized design. The robustness and compactness of the MARGOT system is well fitted to be coupled to a mobile unit that extract, purify and concentrate xenon from air in order to perform atmospheric radioxenon measurements anywhere in the world (On-Site Inspection for instance). The first prototype is described here.

1. From Monte Carlo simulations …

Four relevant radio-isotopes: ^{131m}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe (fission products)

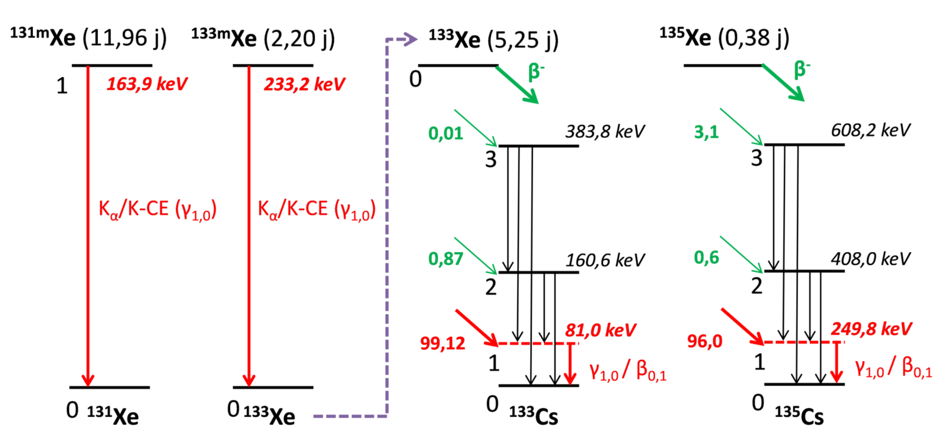


Fig. 1: Simplified level schemes of the four relevant radionuclides [2]. Purple dashed arrow highlights that a disintegration of a ^{133m}Xe nucleus is followed by a ^{133}Xe decay

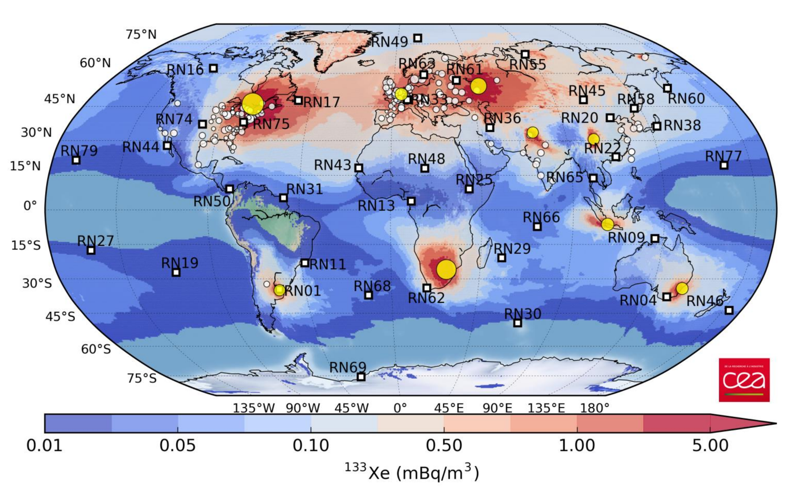


Fig. 2: Annual average concentrations of ^{133}Xe calculated from 2 years of simulated data [3]. Yellow circles: medical isotope production facilities (MIPs); White circle: nuclear power plants (NPPs). The IMS noble gas stations are shown with black squares

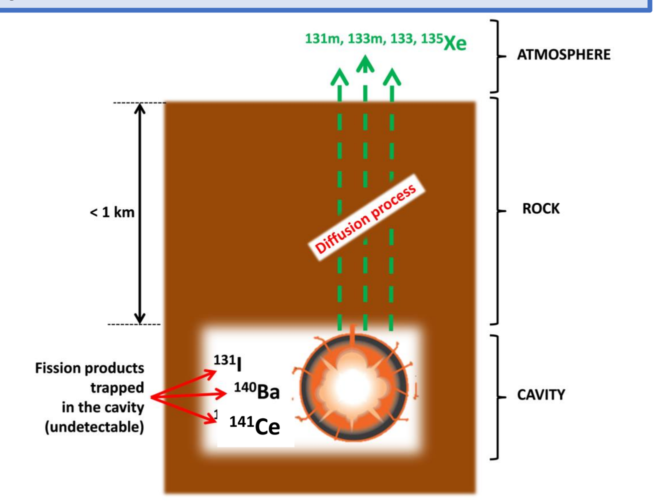


Fig. 3: Simplified scheme of a well confined underground nuclear test

Environmental activities concentrations are in the order of mBq m^{-3}. To perform measurements anywhere in the world, the mobile detection system must have a high detection efficiency with a low detection limit when associated to an efficient gas sampler equipment

REGION	1	2	3	4
Xenon radionuclide	^{135}Xe	^{133}Xe	^{133m}Xe	^{131m}Xe
Coincidence path	$\beta_{0,1}/\gamma_{1,0}$	$\beta_{0,1}/\gamma_{1,0}$	$K\text{-}CE/K\text{-}X_{\alpha+\beta}$	$K\text{-}CE/K\text{-}X_{\alpha+\beta}$
2-D Region Of Interest (width = ± 3 σ) (keV)	[30-915] x [224-278]	[30-346] x [70-92]	[119-139] x [27-41]	[188-210] x [27-41]
Emission probability (%)	90.0	36.7	55.9	54.4

Table 1: Main Region Of Interest (ROI) of each radioxenon

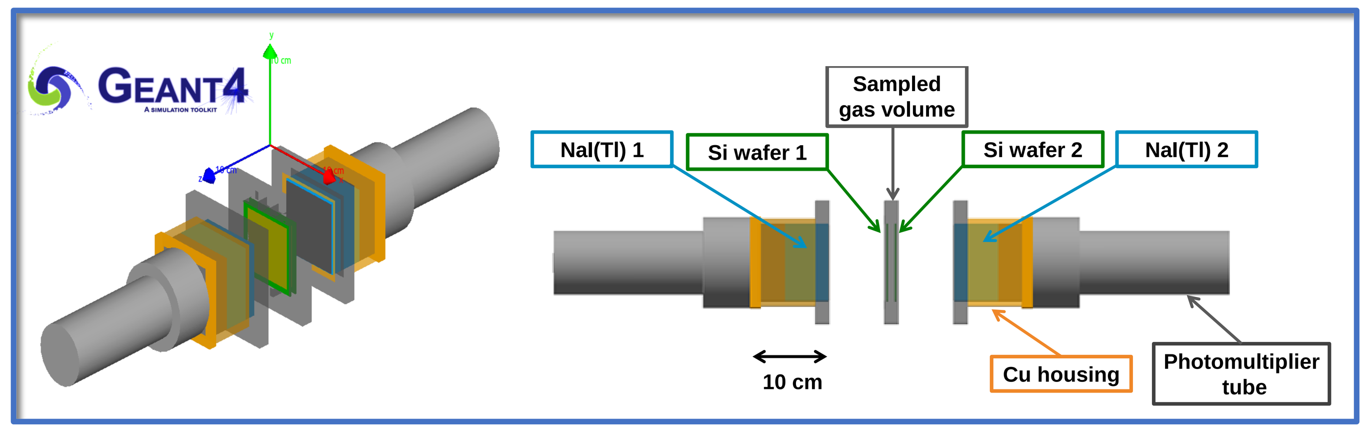


Fig. 4: Geant4 [4] exploded-views of the MARGOT system

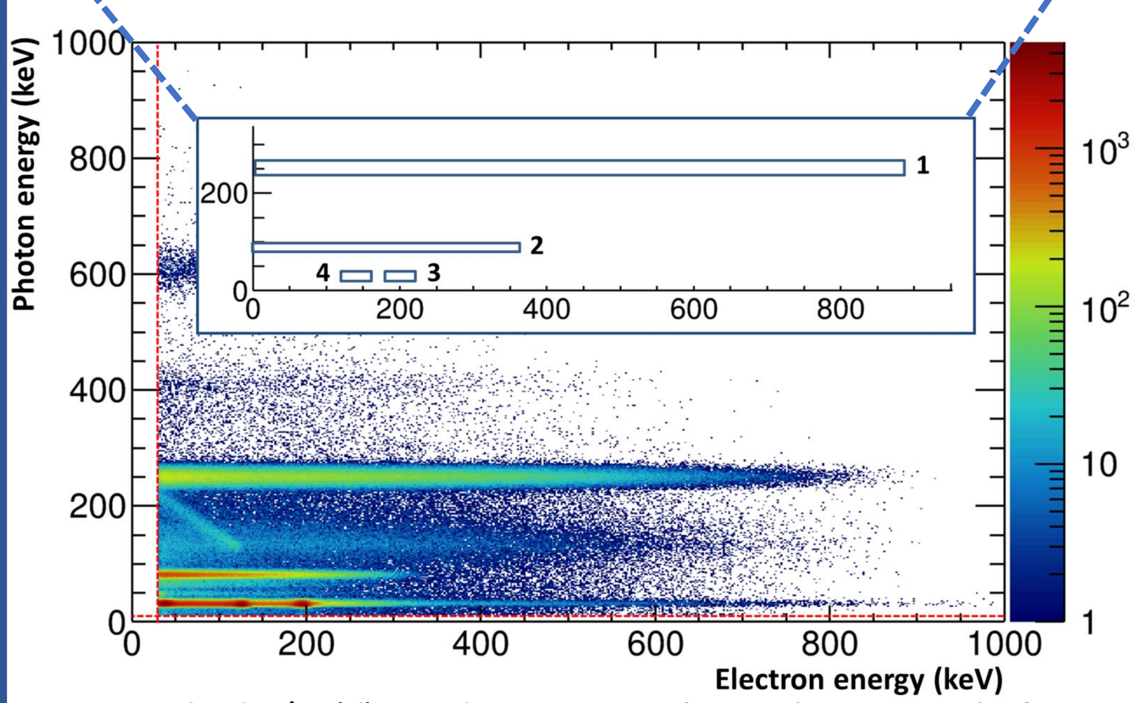


Fig. 5: Simulated Si/NaI(Tl) coincidence spectrum of a sample containing the four relevant xenon radionuclides. Z-axis is given in logarithmic scale. 10^6 disintegrations per radioxenon. Cut-off NaI(Tl) : 10 keV (horizontal red dashed line) Cut-off silicon pixels: 20 keV (vertical red dashed line)

Radioxenon	^{131m}Xe	^{133m}Xe	^{133}Xe	^{135}Xe
Coincidence efficiency (%)	13.2 ± 0.3	12.9 ± 0.3	25.8 ± 0.7	31.9 ± 0.7

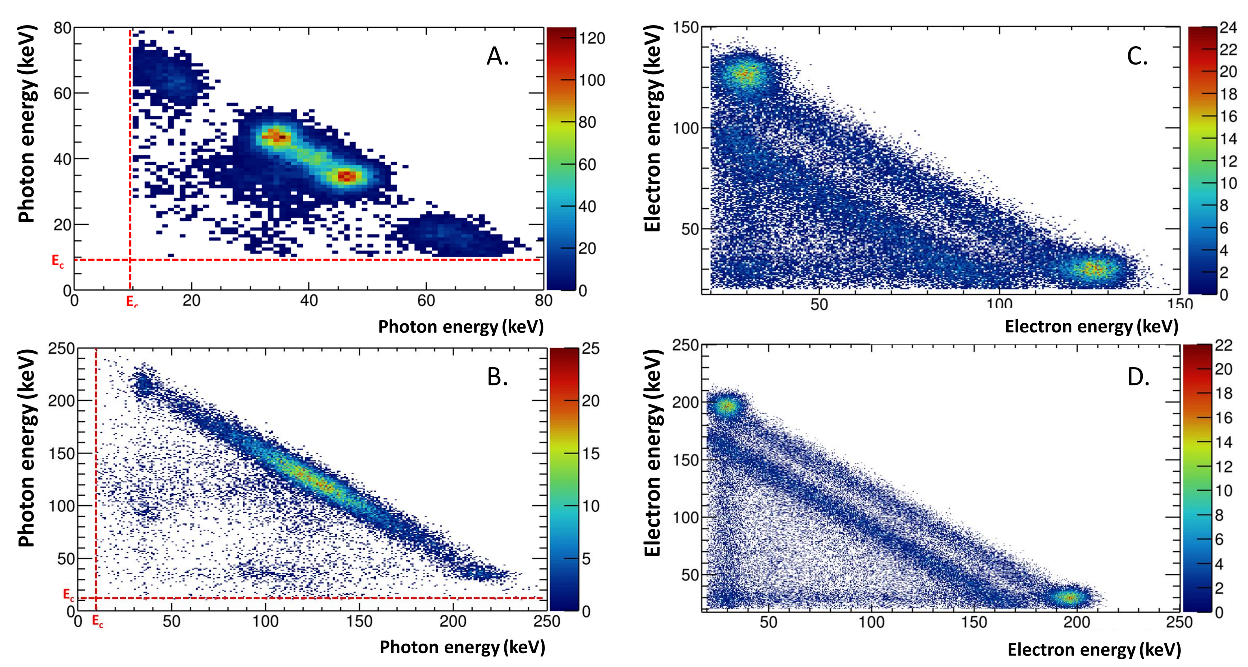
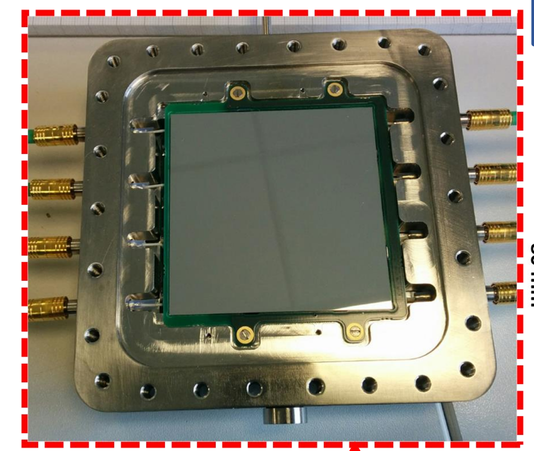
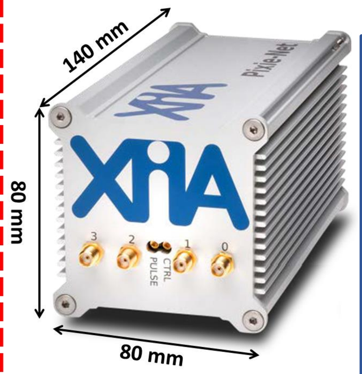


Fig 6: Other coincidences taken into account:

- A: Compton scattering of 81,0 keV photon from ^{133}Xe disintegration simulated by NaI(Tl)/NaI(Tl) coincidence mode
- B: Compton scattering of 249,6 keV photon from ^{135}Xe disintegration measured by NaI(Tl)/NaI(Tl) coincidence mode
- C: Reconstructed energy of 129,0 keV conversion electron from ^{131m}Xe disintegration simulated by Si/Si coincidence mode
- D: Reconstructed energy of 198,7 keV conversion electron from ^{133m}Xe disintegration measured by Si/Si coincidence mode

2. … To Laboratory tests





Pixie-NET digital spectrometer

- ✓ 2 x Large surface (60*60 mm^2), Passivated Implanted Planar Silicon (PIPS™) detectors (Mirion technologies)
- ✓ Each silicon wafer has been pixellized (4 independent pixels) in order to reduce leakage current
- ✓ Very large sample volume: 54.3 ± 1.2 cm^3
- ✓ Gas pressure up to 200 kPa
- ✓ 2 x Low Background NaI(Tl) detectors (Scionix Holland)
- ✓ 3 x Pixie-NET digital electronics (XIA LLC)
- ✓ 1 Switch MOXA EDS 405-A PTP for time synchronization between modules
- ✓ Network time synchronization of the readout electronics using IEEE 1588 Precision Time Protocol [6]
- ✓ No shielding, light and compact system
- ✓ Works at ambient temperature

Calibration of the detectors

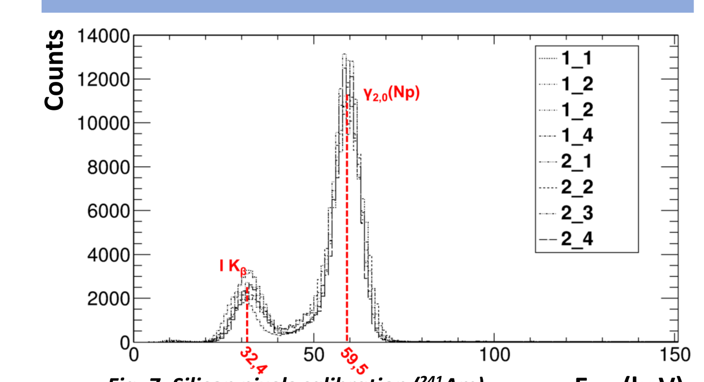


***Fig. 7:** Silicon pixels calibration (^{241}Am)*

Pixel ID	1_1	1_2	1_3	1_4	2_1	2_2	2_3	2_4
Leakage current (nA)	12.2	6.1	5.1	6.1	2.1	2.3	2.7	1.7
Cut-off energy (keV)	25	25	25	25	20	20	20	20

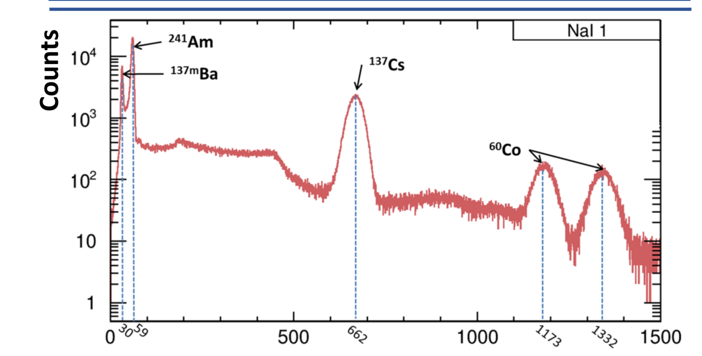


***Fig. 8:** NaI(Tl) response function (multi-γ)*

3. Spikes of radioxenons

Spike prepared by Seibersdorf Laboratories, the sample consisted in a mix of the four radionuclides in a SPALAX bottle (300 cm^3), at a pressure of 120 kPa (carrier gas: nitrogen)

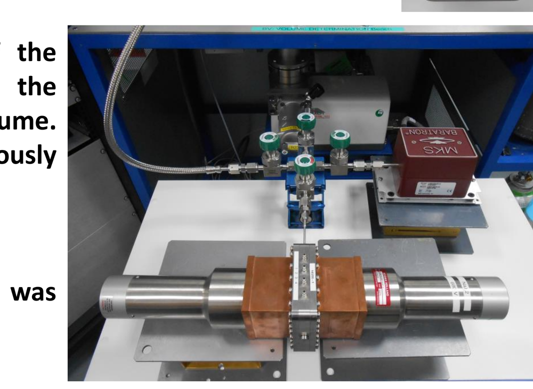


Fig 9: Transfer of the archive bottle to the detector inner volume. The cell was previously vacuumed (P= 3.0 10^{-4} kPa).

After transfer, P_{gas} was 9.38 10^1 kPa. (at T = 293.7 K)

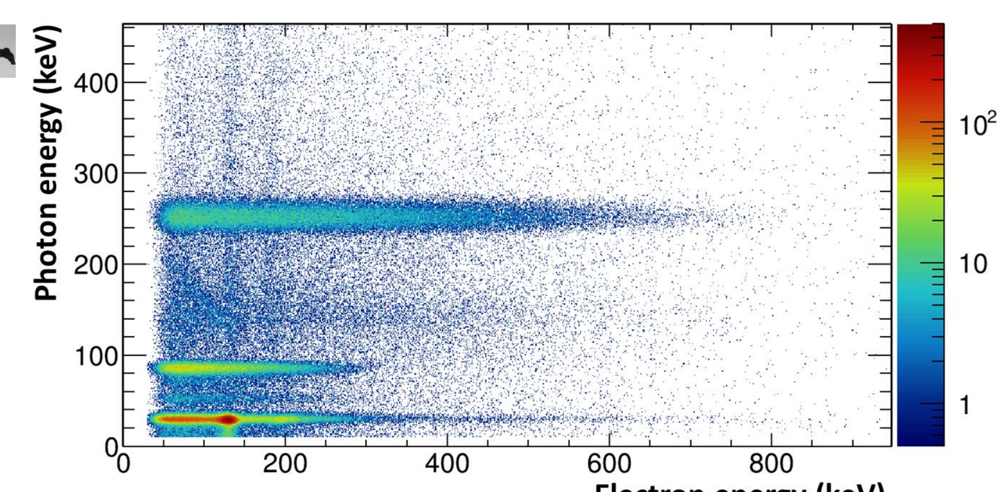


***Fig. 10:** Si/NaI(Tl) coincidence spectrum. Acquisition time = 43,200 s*

***Table 2:** Activities of each radioxenon referenced to June 4, 2019 at 12:00 UTC.* [7]

Xenon radionuclide	^{131m}Xe	^{133m}Xe	^{133}Xe	^{135}Xe
Certified Activity (Bq)	120 ± 5.3	5.7 ± 0.3	115 ± 6	117± 6
Activity at T_0 (Bq)	117.4 ± 5.4	5.3 ± 0.3	109.4 ± 6.1	60.6 ± 6.1
Decay correction	0.986	0.925	0.968	0.657

Coincidence efficiency (%)	^{131m}Xe	^{133m}Xe	^{133}Xe	^{135}Xe
Monte Carlo Simulation	13.2 ± 0.3	12.9 ± 0.3	25.8 ± 0.7	31.9 ± 0.7
Experimental	12.0 ± 1.7	11.8 ± 1.6	23.2 ± 2.9	34.9 ± 4.8

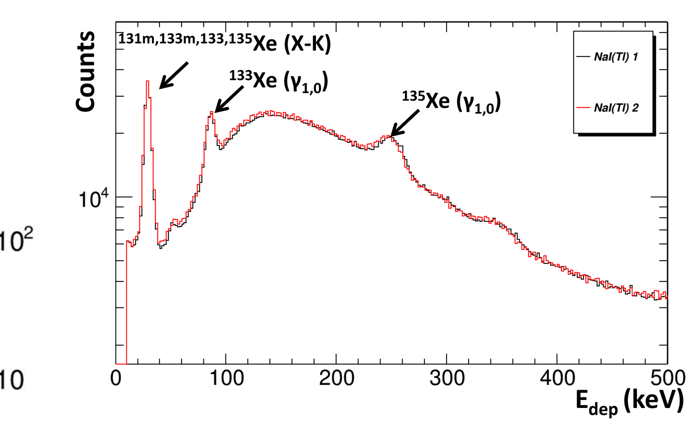


***Fig. 11:** Response function of the two NaI(Tl) detectors*

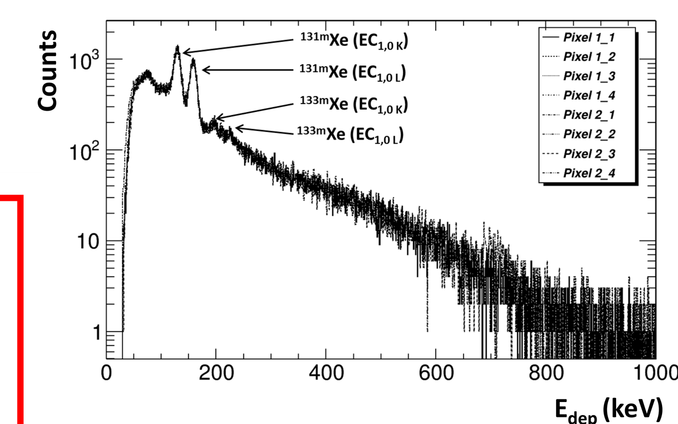


***Fig. 12:** Response function of the 8 silicon detectors*

References

[1] G. Le Petit *et al.,* "SPALAX-NG: a sensitive and selective noble gas system for nuclear explosion monitoring", *in Applied Radiation and Isotopes,* 2015.
[2] M.M. Bé, V. Chisté *et al.,* "Table of Radionuclides vol.8", *Decay Data Evaluation Project Working Group (DDEP-WG),* 2016.
[3] P. Achim, S. Generoso *et al.*, "Characterization of Xe-133 global atmospheric background: Implications for the International Monitoring System of the Comprehensive Test-Ban Treaty " in *Journal of Geophysical Research: Atmospheres*, 2016.
[4] S. Agostinelli *et al.;* "GEANT4—a simulation toolkit" in *Nuclear instruments and methods in physics research section A: Accelerators, Spectrometers, Detectors and Associated Equipment,* 2003.
[5] V. Thomas, O. Delaune *et al.*, ''Introducing the MARGOT prototype: an ultra-compact and mobile gas detection system for nuclear explosion monitoring'' (*submitted to Applied Radiation and Isotopes*), 2019.
[6] W. Hennig, V. Thomas *et al.*, ''Network Time Synchronization of the Readout Electronics for a New Radioactive Gas Detection System'' in *IEEE Transactions on Nuclear Science,* 2018.
[7] V. Thomas, O. Delaune *et al.* ''Introduction to a new radioactive gases detector: the Mobile Analyzer of Radioactive Gases OuTflows" (*submitted to Applied Radiation and Isotopes*), 2019.