



Influence of beta spectrum on new primary activity standards

¹B. Sabot, ¹X. Mougeot, ^{1,2}A. Perret



¹Université Paris Saclay, CEA, LIST, Laboratoire National Henri Becquerel (LNE-LNHB), 91192 Palaiseau, France

²Université Claude Bernard Lyon 1, CNRS, ILM UMR 5306, Villeurbanne, France



Context

Metrology:

- As a national metrology institute, the Laboratoire National Henri Becquerel (CEA) is developing primary standard methods for the standardisation of pure beta emitters
- The methods discussed here are based on liquid scintillation counting.
 - Current techniques: Triple to Double Coincidence Ratio (TDCR), which is based on measurements and models
 - The new method: Compton-TDCR, which is based on a full measurement approach.
- For both methods, you need to know which nuclide you are measuring, and then the emission spectra can be used as the basis for calculations
- Here, we will demonstrate the importance of spectra, especially with regard to the new method, which provides lower uncertainties

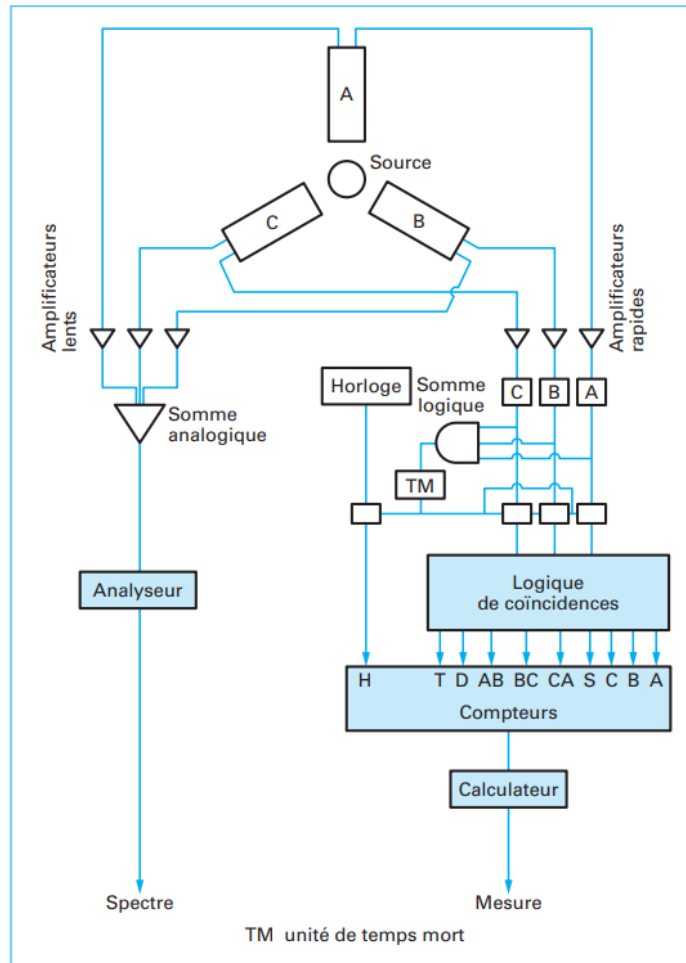




1 ■ Current primary measurement method

Based on liquid scintillation counting

Triple to Double Coincidence Ratio



(P. Cassette, Technique de l'ingénieur, P2552 v2)

Metrology:

- Requires a scintillation counter with 3 photomultipliers (PMTs) 1/3 symmetry: 120° between each PMT
- Measurement geometry: a 20 mL liquid scintillation vial filled with liquid scintillator
- Electronics capable of acquiring the average number of triple (T) and double (D) coincidences, as well as:
 - each individual signal (A, B and C) with adjustable thresholds
 - each double coincidence (AB, BC, BC)
- Detection efficiency calculated from the T/D ratio
- The calculation is based on three fundamental assumptions

Triple to Double Coincidence Ratio

- Assumption 1: for an energy deposit E in the scintillator, the probability of emitting x photons for an average number m follows a Poisson distribution:

$$P(x/m) = \frac{m^x e^{-m}}{x!}$$

- Assumption 2: the probability of detecting a photon is not zero; the detection efficiency is:

$$1 - P(0/m) = 1 - e^{-m}$$

- For a detector with 3 PMTs in 1/3 symmetry and quantum efficiency ν :

Detection efficiency for 1 photo detector	$R_1 = 1 - e^{-\frac{\nu m}{3}}$
Detection efficiency for 2 photo detector in coincidence	$R_2 = \left(1 - e^{-\frac{\nu m}{3}}\right)^2$
Detection efficiency for 3 photo detector in coincidence	$R_T = \left(1 - e^{-\frac{\nu m}{3}}\right)^3$
Detection efficiency for the logical sum of the double	$R_D = 3\left(1 - e^{-\frac{\nu m}{3}}\right)^2 - 2\left(1 - e^{-\frac{\nu m}{3}}\right)^3$

Triple to Double Coincidence Ratio

- Hypothesis 3: The light yield follows Birks' law, which gives the average number of photons emitted per unit of energy E absorbed in the scintillator:

$$m(E) = \int_0^E \frac{\alpha dE}{1 + k_B \frac{dE}{dx}}$$

Where α is the scintillation efficiency (photons·keV⁻¹), k_B is the scintillator parameter (ranging from 0.005 to 0.018 cm·MeV⁻¹), and dE/dx is the linear energy transfer of the ionising radiation

- Thus, considering the normalised $S(E)$ spectrum of energy emitted by the radionuclide

$$RCTD = \frac{\int_{spectre} S(E) (1 - e^{-\frac{\nu \alpha m(E)}{3}})^3}{\int_{spectre} S(E) \left(3 \left(1 - e^{-\frac{\nu \alpha m(E)}{3}} \right)^2 - 2 \left(1 - e^{-\frac{\nu \alpha m(E)}{3}} \right)^3 \right)} \text{ avec } m(E) = \int_0^E \frac{dE}{1 + k_B \frac{dE}{dx}}$$

- Given that, for a large number of events, the ratio of frequencies converges to the ratio of probabilities, the calculation algorithm consists of finding the free parameter, $\nu \alpha$, such that:

$$RCTD = \frac{T}{D}$$

Triple to Double Coincidence Ratio

- The PMs are never identical:

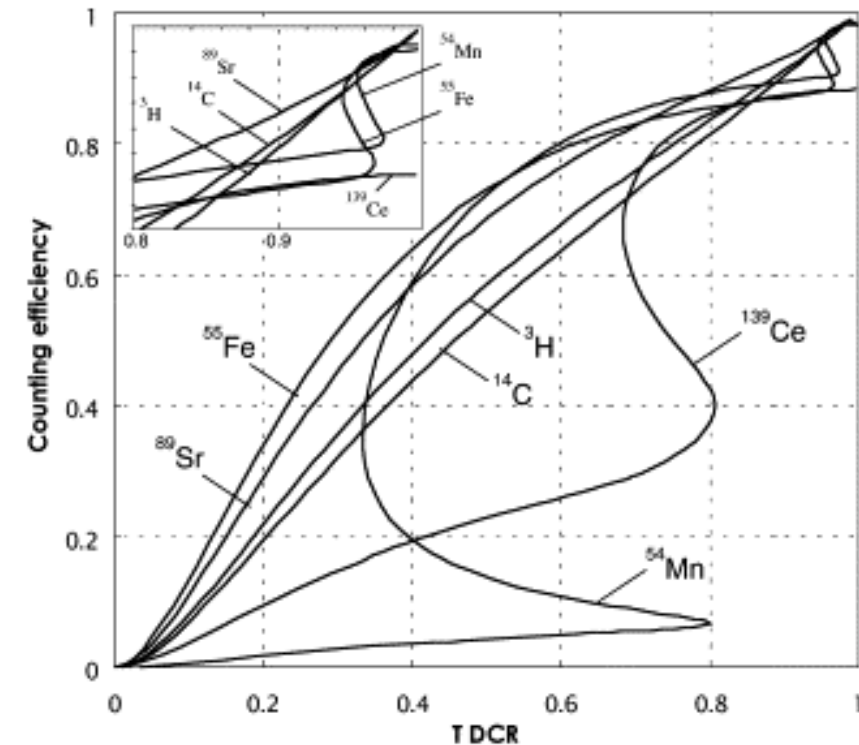
$$\frac{\varepsilon_T}{\varepsilon_{AB}} = \frac{\int_{\text{spectre}} S(E) \left(1 - e^{-\frac{\nu_A \alpha m(E)}{3}}\right) \left(1 - e^{-\frac{\nu_B \alpha m(E)}{3}}\right) \left(1 - e^{-\frac{\nu_C \alpha m(E)}{3}}\right) dE}{\int_{\text{spectre}} S(E) \left(1 - e^{-\frac{\nu_A \alpha m(E)}{3}}\right) \left(1 - e^{-\frac{\nu_B \alpha m(E)}{3}}\right) dE}$$

And similar for $\frac{\varepsilon_T}{\varepsilon_{BC}}$ and $\frac{\varepsilon_T}{\varepsilon_{AC}}$

- The solution to be minimised becomes:

$$\Delta = \left(\frac{\varepsilon_T}{\varepsilon_{AB}} - \frac{R_T}{R_{AB}} \right)^2 + \left(\frac{\varepsilon_T}{\varepsilon_{BC}} - \frac{R_T}{R_{BC}} \right)^2 + \left(\frac{\varepsilon_T}{\varepsilon_{AC}} - \frac{R_T}{R_{AC}} \right)^2$$

- A single solution for electrons
- Up to three solutions for β - γ emitters and electron capture: the solution to resolve the ambiguity is to vary the yield



Changes in yield as a function of TDCR for various radionuclides (R. Broda, 2003, ARI 58)

Triple to Double Coincidence Ratio

- Example of devices:



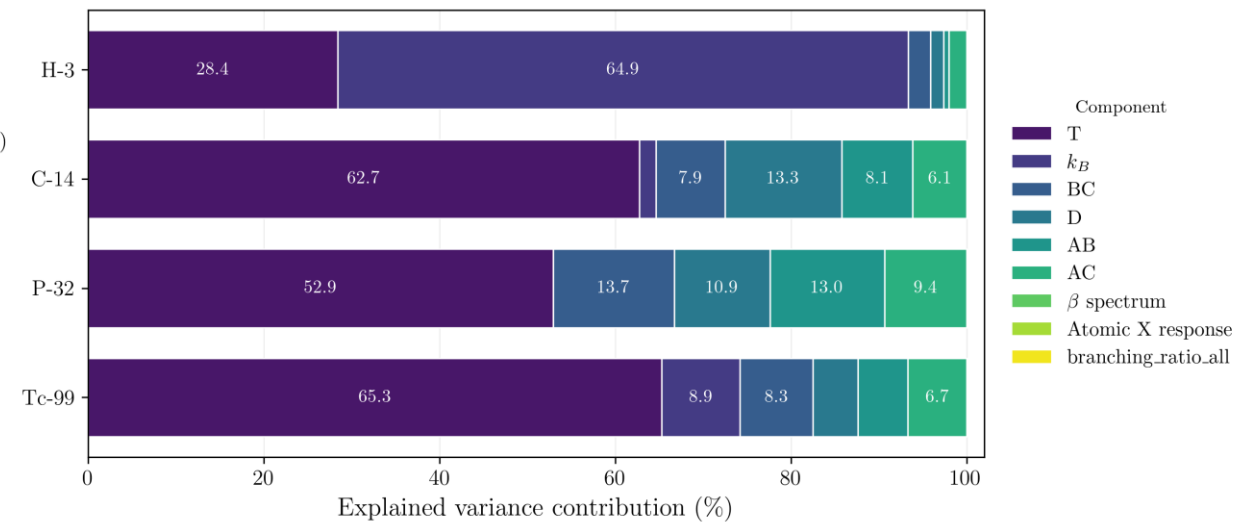
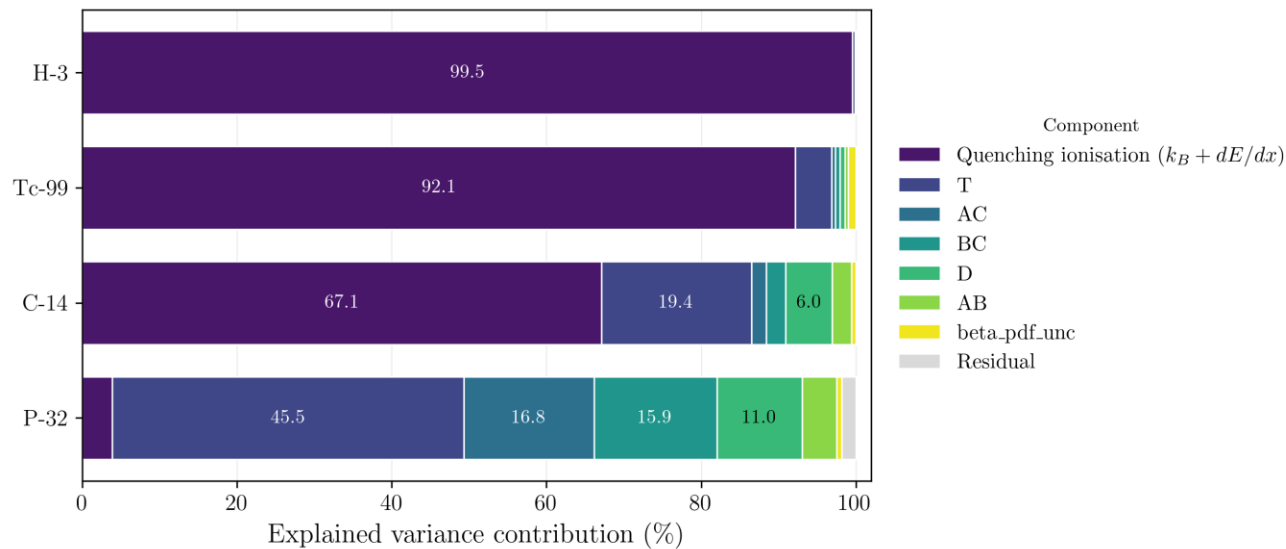
More than 30 years old, still working developed by Robert Vatin, Philippe Cassette, and Jacques Bouchard



Benoît Sabot, Chavdar Dutsov, Philippe Cassette, Krasimir Mitev, Performance of portable TDCR systems developed at LNE-LNHB, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 1034, 2022, 166721

Triple to Double Coincidence Ratio

- Example of application on some radionuclides; pure beta emitter in Ultimagold scintillator in micro-TDCR device, analysis performed with TDCR++
 - NOTE : TDCR++ is a new efficiency computation code we developed with precise model and especially a sensitivity analysis with nuclear data
- Analysis of Variance on computed activity results:



- The main parameter of uncertainty is the model and then spectrum (when counting is very high)

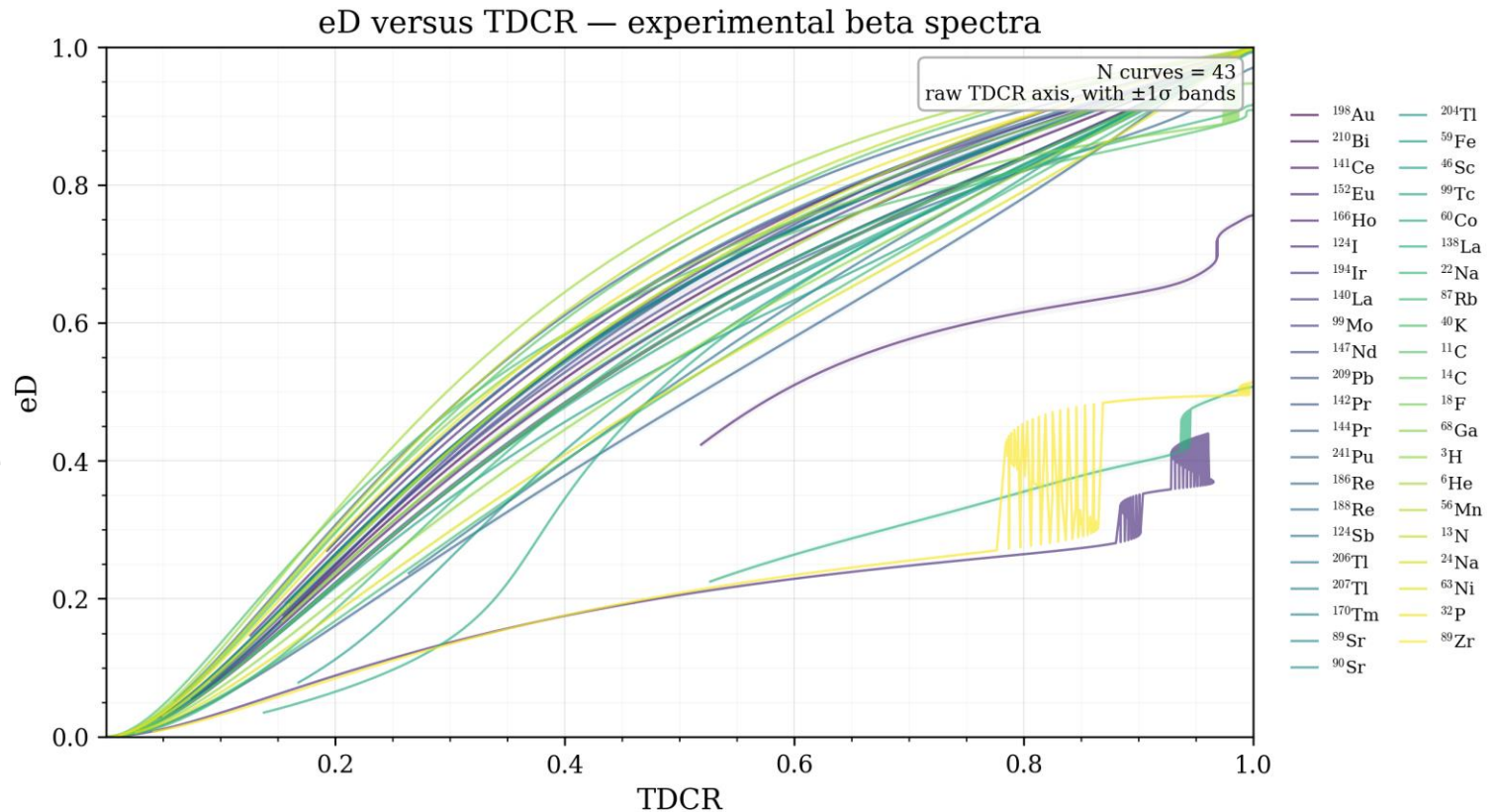


2. Digging a little further into TDCR counting by looking at the impact of beta spectra

PRELIMINARY RESULTS

A systematic study from TDCR and Beta-spectra

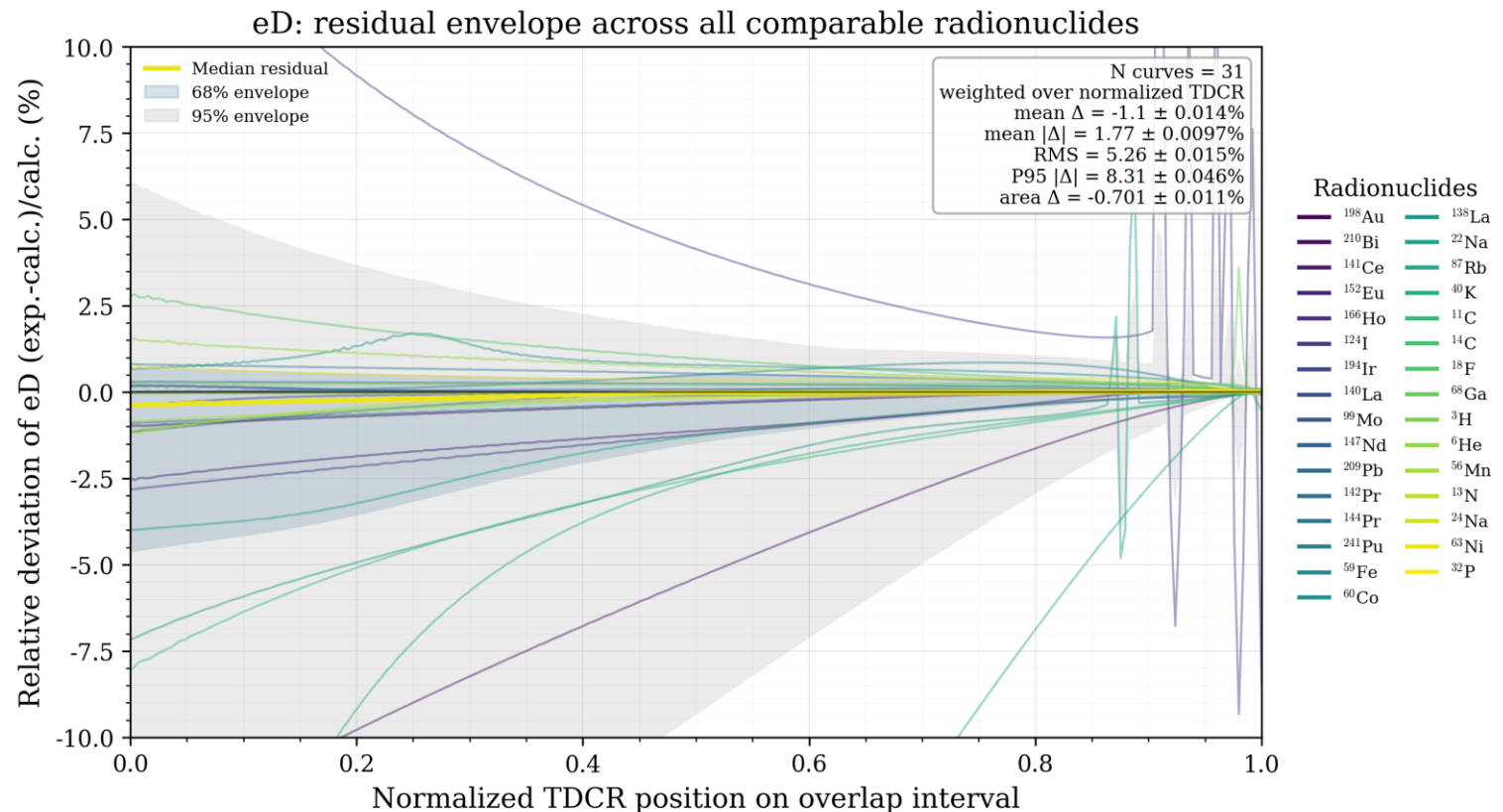
- Our new computation code allows us to carry out new analyses. After discussing this with Xavier, we decided to take a closer look.
- The method is as follows:
 - Take all spectra from BetaShape with computed and experimental data
 - Compute the detection efficiency curve with TDCR++ and compare the results
 - Find indicators, analysis methods and comparison techniques to gather information on how the results are influenced



All computed efficiencies vs TDCR for 43 radionuclides from Betashape, computed with TDCR++

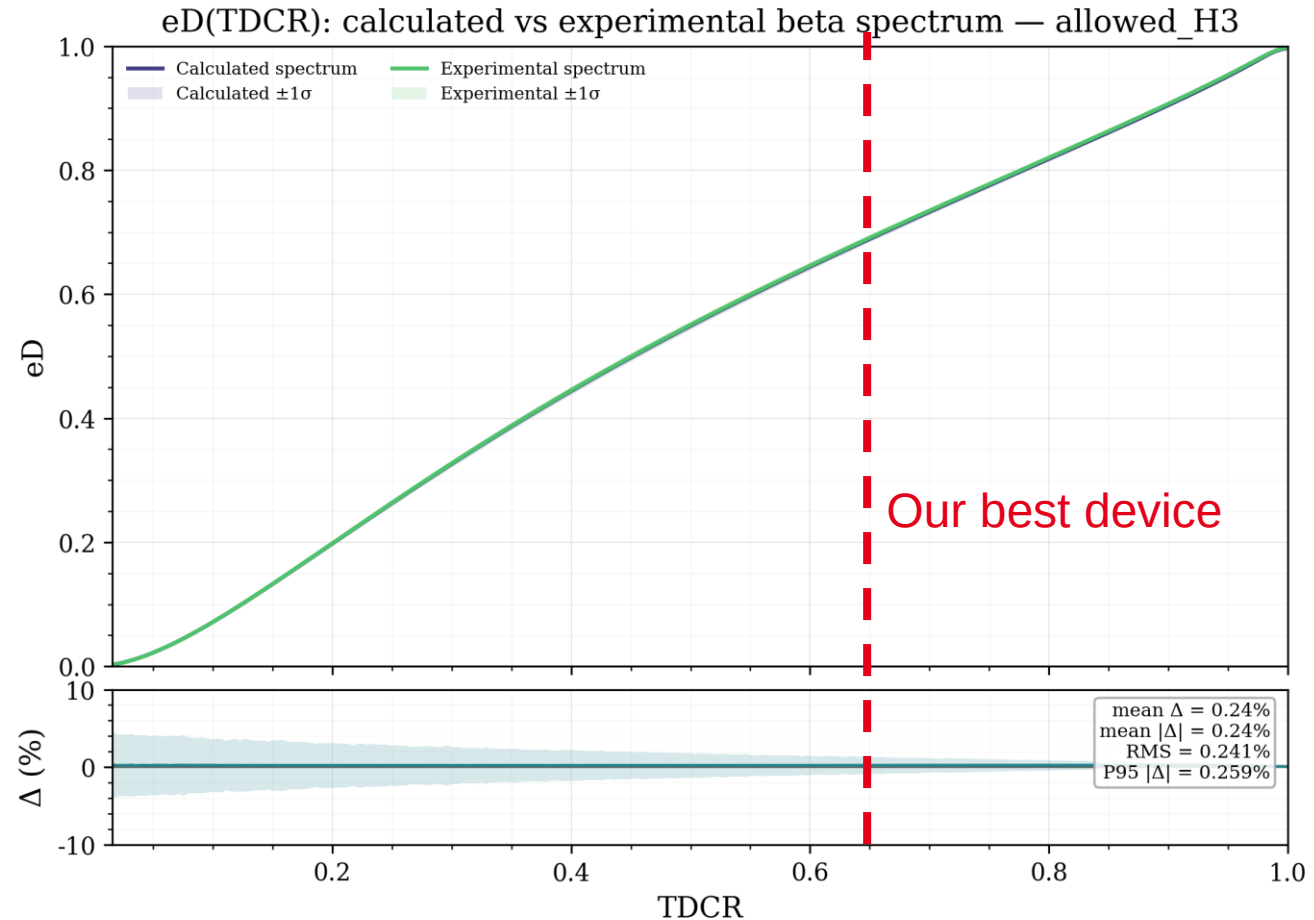
A systematic study from TDCR and Beta-spectra

- The deviation between computed detection efficiency from experimental spectra and calculated spectra from BetaShape 2.4.
- Some strange artefacts are visible: the decay scheme is giving complex computational results
 - NOTE : the full scheme is computed in TDCR++ but we change only the beta spectra
- The global results are hard to interpret but we can analyze one by one



A systematic study from TDCR and Beta-spectra

- Example of ^3H in Ultimagold
 - Very small difference; lower than the current model dependency
- The number of datasets is important, and this study is interesting, so we have decided to publish the work alongside a full dataset
- It provides a kind of warning for our experiments:
 - is the spectrum well known?
 - How sensitive am I to the spectra?
 - Should we investigate further to improve the measurement of these spectra?
- **I view this dataset as a way to identify critical spectra to be measured**



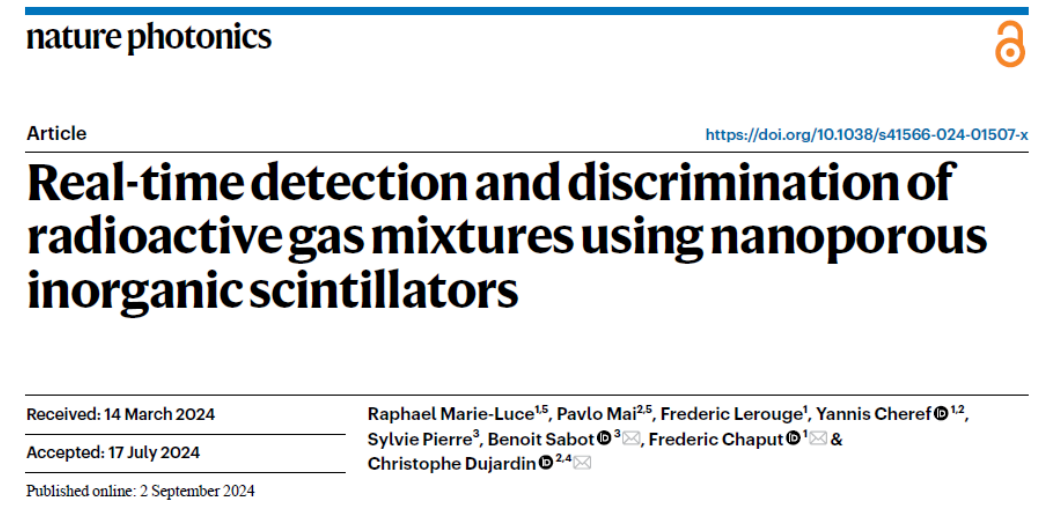
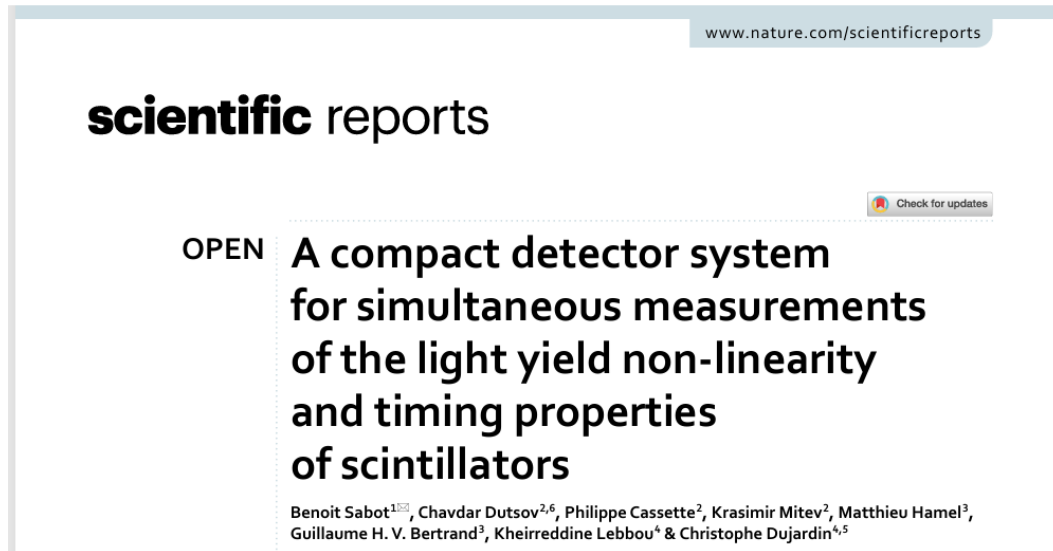


3. And if the model in TDCR disappear?

Here we go ! Compton-TDCR

Compton-TDCR ; more than a new primary method

- We developed this approach to overcome the limitations of the TDCR: the scintillation model and the dependency on liquid scintillators
- We demonstrated its feasibility on many scintillators and with a groundbreaking application

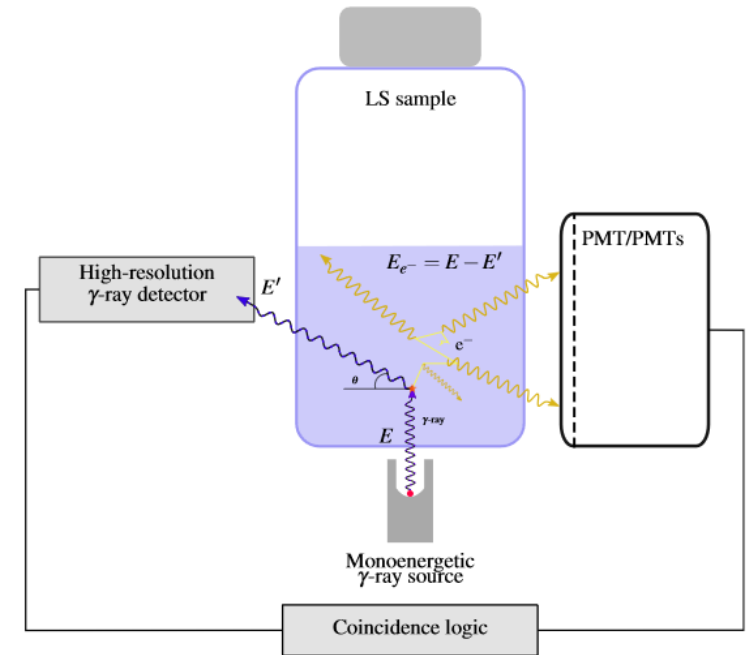


Compton-TDCR

- A radioactive source emits a γ -ray, which interacts with the liquid scintillator mainly through Compton scattering, transferring part of its energy to an electron,
- The Compton electron deposits its energy in the scintillator, producing scintillation light.
- Three photomultiplier tubes (PMTs) simultaneously observe the scintillator. The signals are recorded as double coincidences with gamma (ABG), (ACG), (BCG) and triple coincidences with gamma (ABCG),
- The mean number of photo-electron is calculated as:

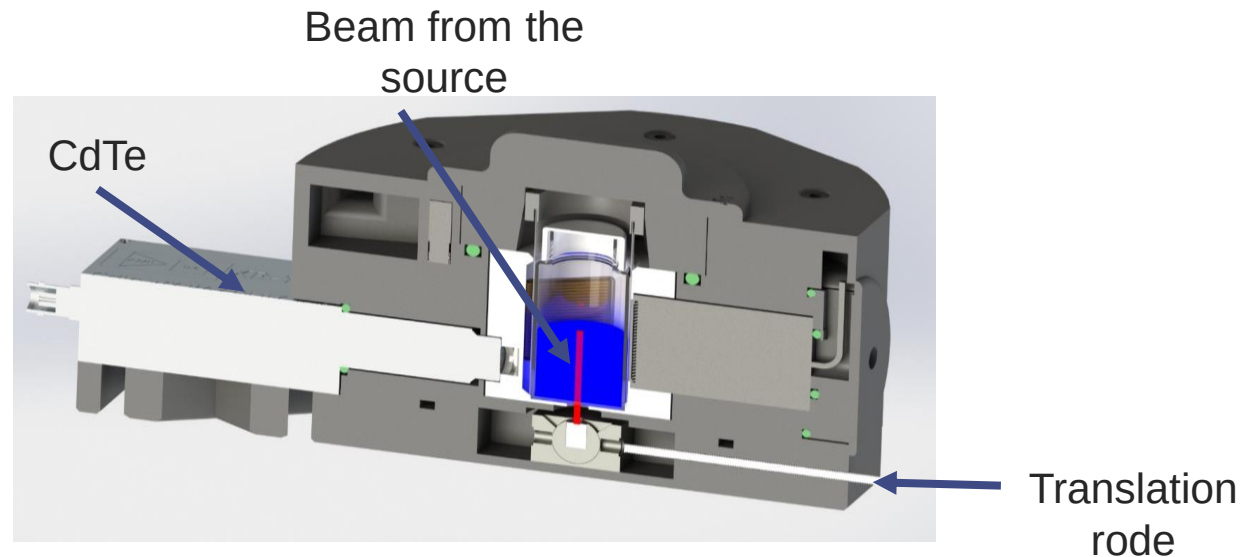
$$\bar{n}_X(E) = -3 \ln \left(1 - \frac{TG(E)}{YZG(E)} \right), \quad X = (A, B, C), \quad YZ = (AB, BC, AC), \quad X \neq Y \text{ or } Z.$$

- By selecting different Compton-electron energies, the scintillator response can be measured as a function of electron energy, allowing its light-yield non-linearity and quenching to be characterized.



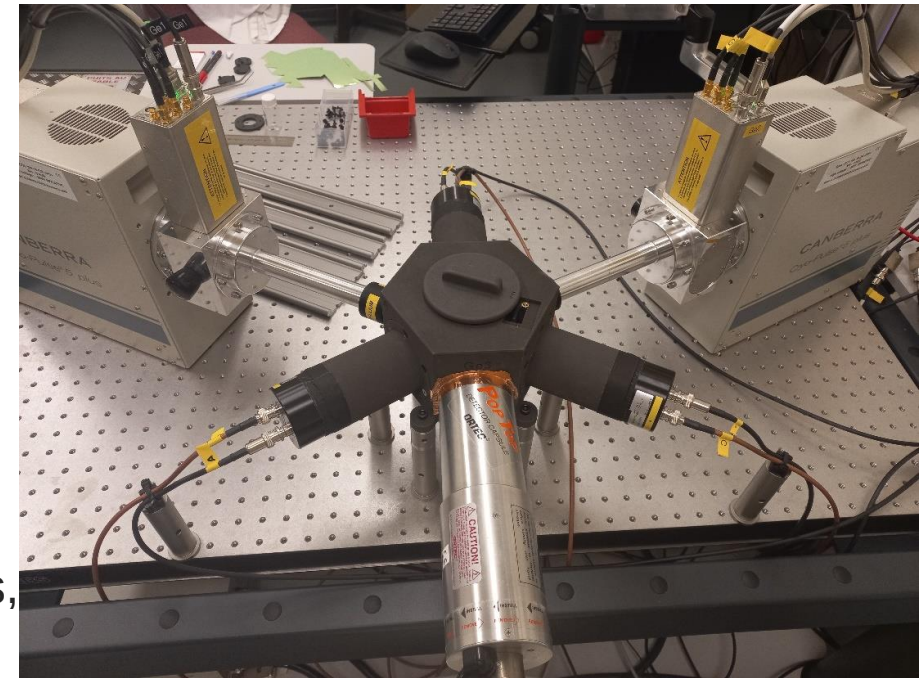
Compton-TDCR, the Little boy

- The Mini C-TDCR is the compact configuration initially developed to demonstrate the feasibility of the Compton-TDCR method
- It combines three compact Hamamatsu PMTs arranged around the scintillation vial with a single CdTe detector (Amptek) used to detect the scattered γ rays
- The system is irradiated with a ^{241}Am source emitting 59.54 keV γ -rays
- Signal acquisition is performed using a 4-channel, 10-bit CAEN DT5751 digitizer operating at 1 GS/s



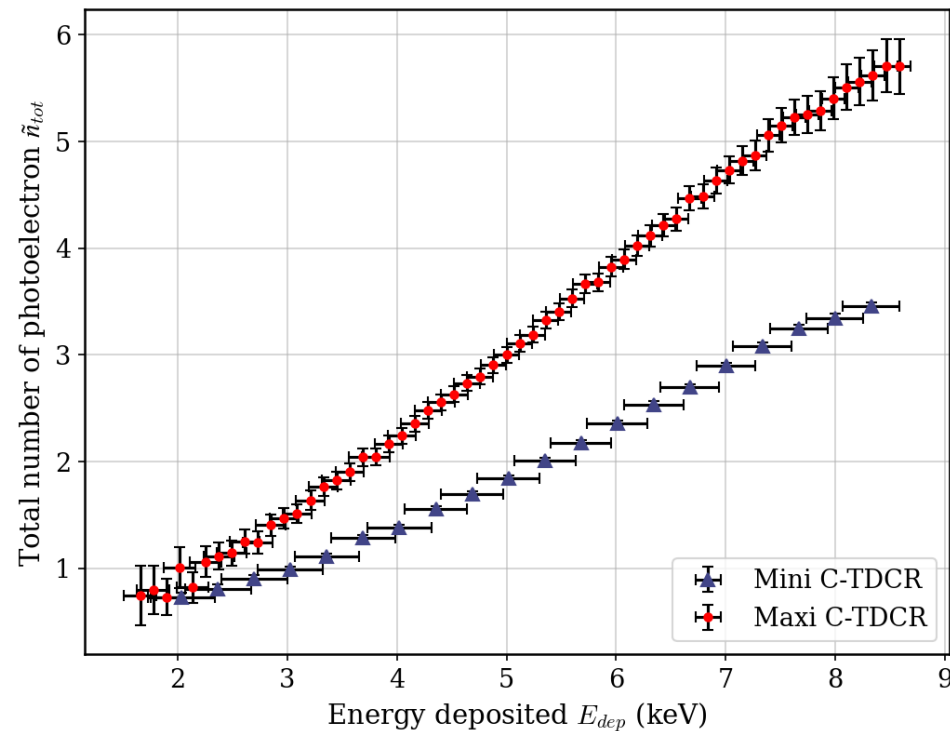
Compton-TDCR, the Fat man

- The mini device was limited by its stability and resolution (360 eV at best), it was portable to allows the use of external beam easily and to path the way to a real instruments
- The Maxi C-TDCR was developed to improve the sensitivity, energy resolution, and flexibility of the initial Mini C-TDCR configuration:
 - Custom Hamamatsu PMTs with 51 mm windows
 - Three HPGe detectors providing improved γ -ray energy resolution (134 eV at best) and a larger angle range for scattered-photon detection
 - 16-channel, 14-bit CAEN DT2751 digitizer at 1 GS/s and
 - adjustable ^{241}Am source geometry with three source positions, allowing to tune scattering angle
 - A very motivated PhD student, Alice Perret, that is intensively working on it !

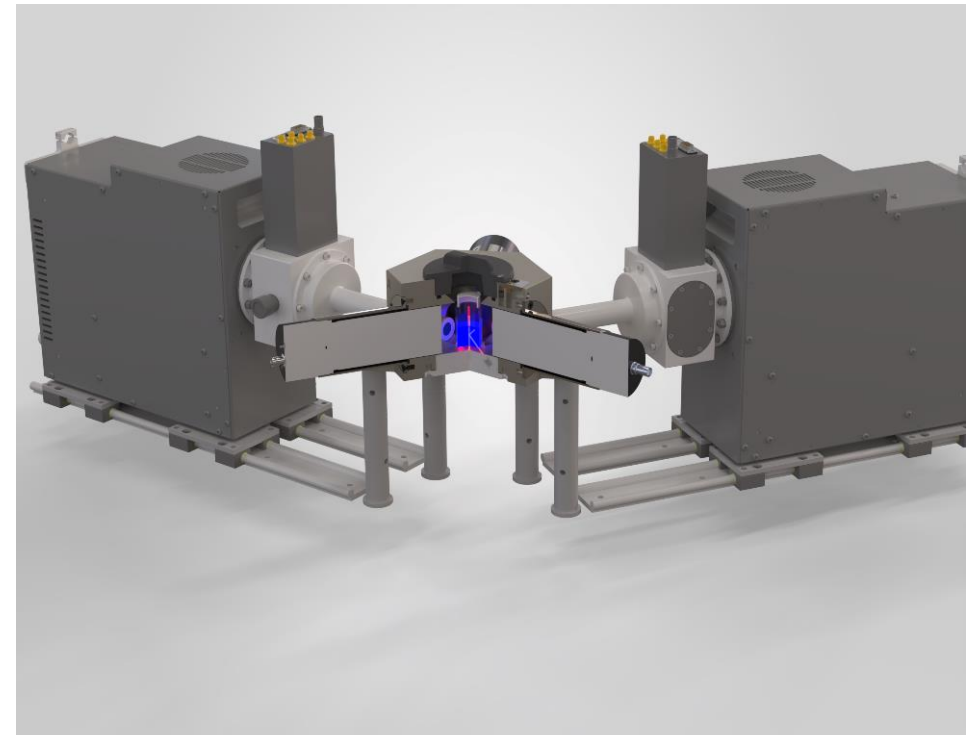


The Maxi device

- Two goals; new primary measurement technique and a device to measure non-linearity curve response of any scintillator material (SCINTI+ database: <http://www.lnhb.fr/home/conferences-publications/scinti-plus/>)

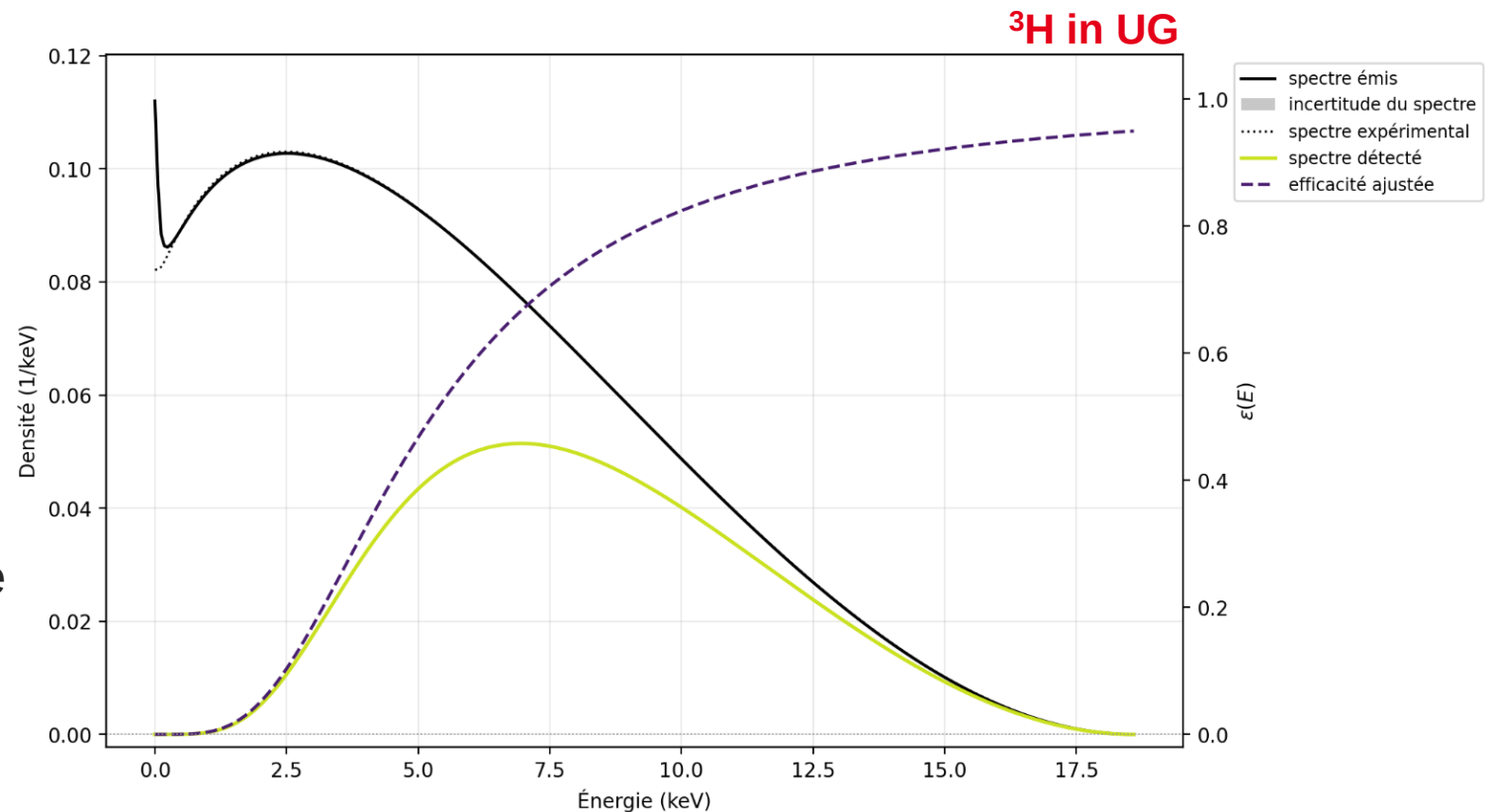


Example of non-linearity curve obtained for Ultimagold scintillator



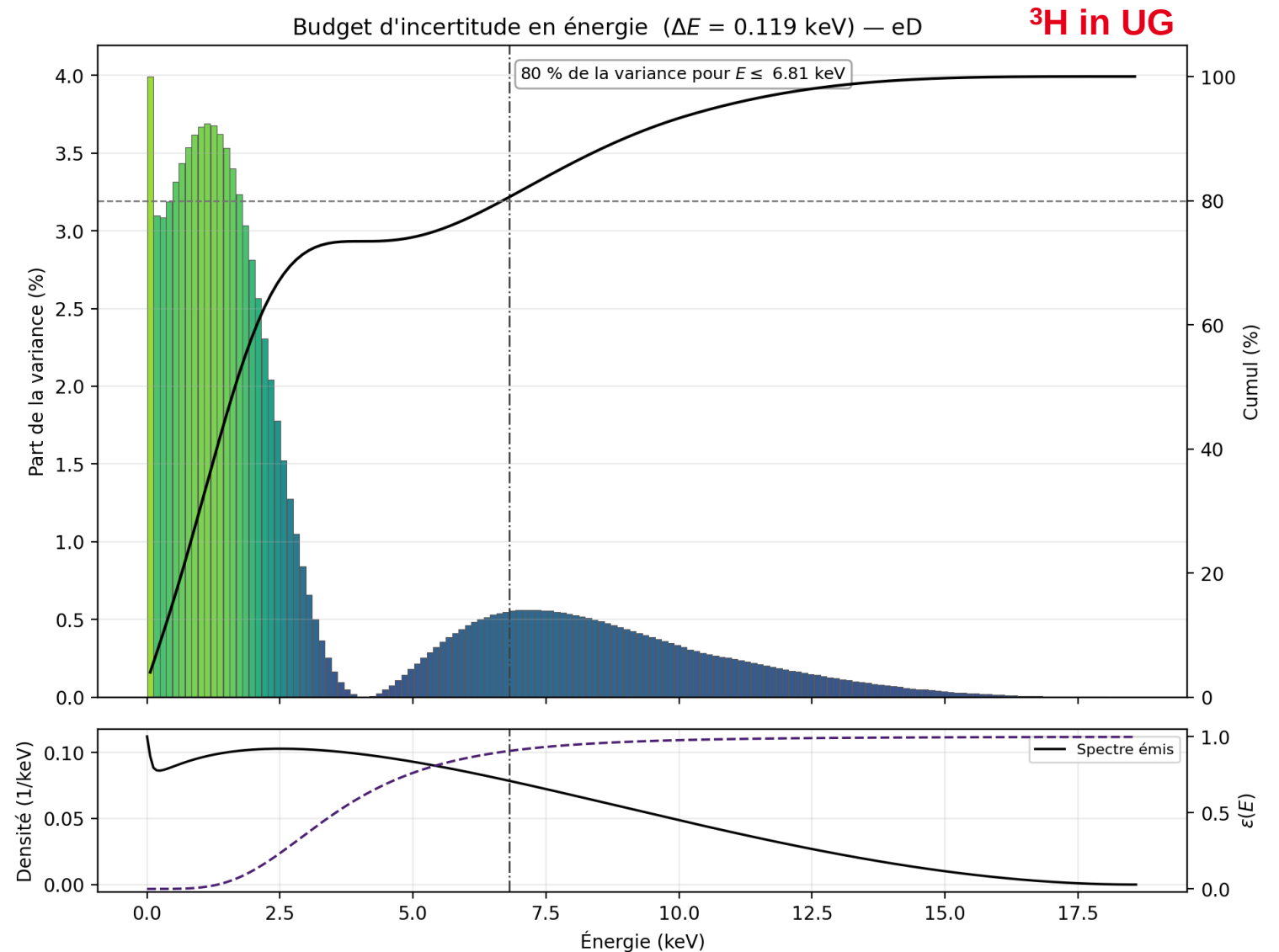
How we use the method ?

- We can derive the mean number of photoelectron detected per PMT for a deposited energy inside the scintillator: if we have a radionuclide with a known spectra inside the scintillator we can then calculate detection efficiency for each PMTs
- Only three data to combine :
 - Response curve (measured, depends on statistic)
 - Beta spectrum
 - Counting rate for radionuclide
- We can achieve below 0.1% relative standard uncertainty on radionuclide such as ^3H , almost one order of magnitude below TDCR !
- But, what about the spectra ?



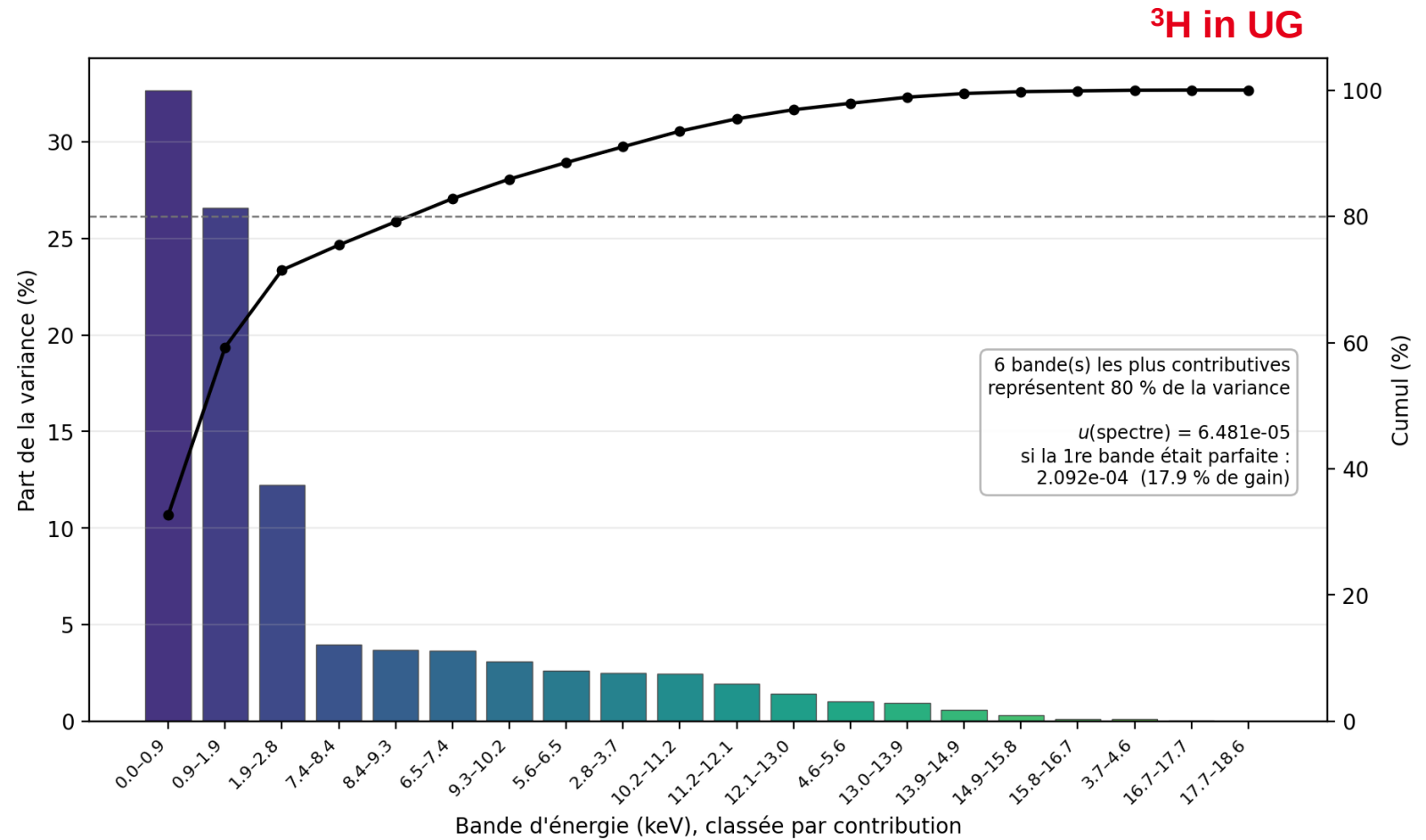
How we are sensitive to spectra ?

- We become again very sensitive, but we can exactly know where and how much !
- We performed sensitivity study with the method over the whole spectra with HPGe resolution



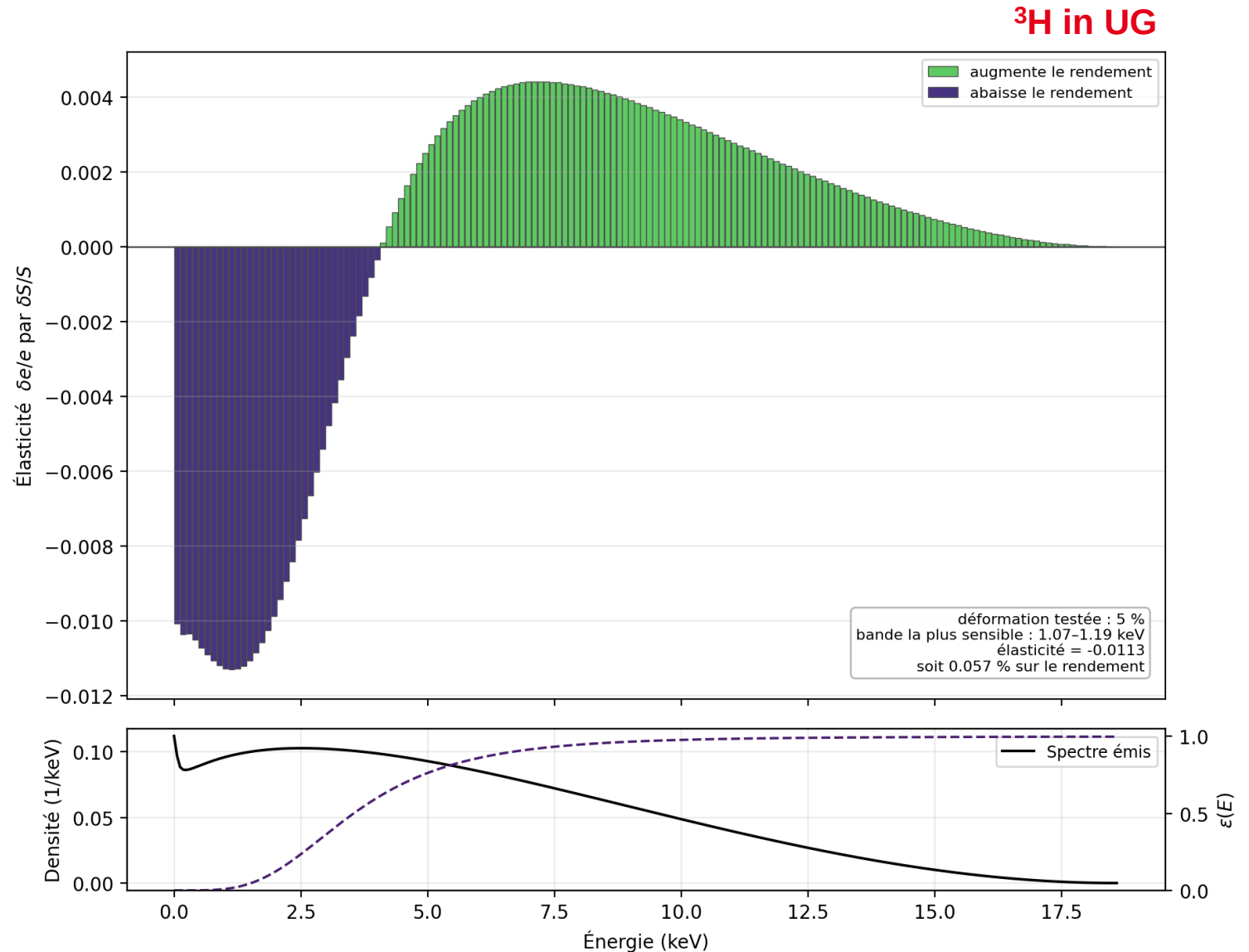
How we use the method ?

- From a broader perspective, we are, of course, aware that the spectrum dependency is at low energies, where the non-linearity of the scintillator is very important.



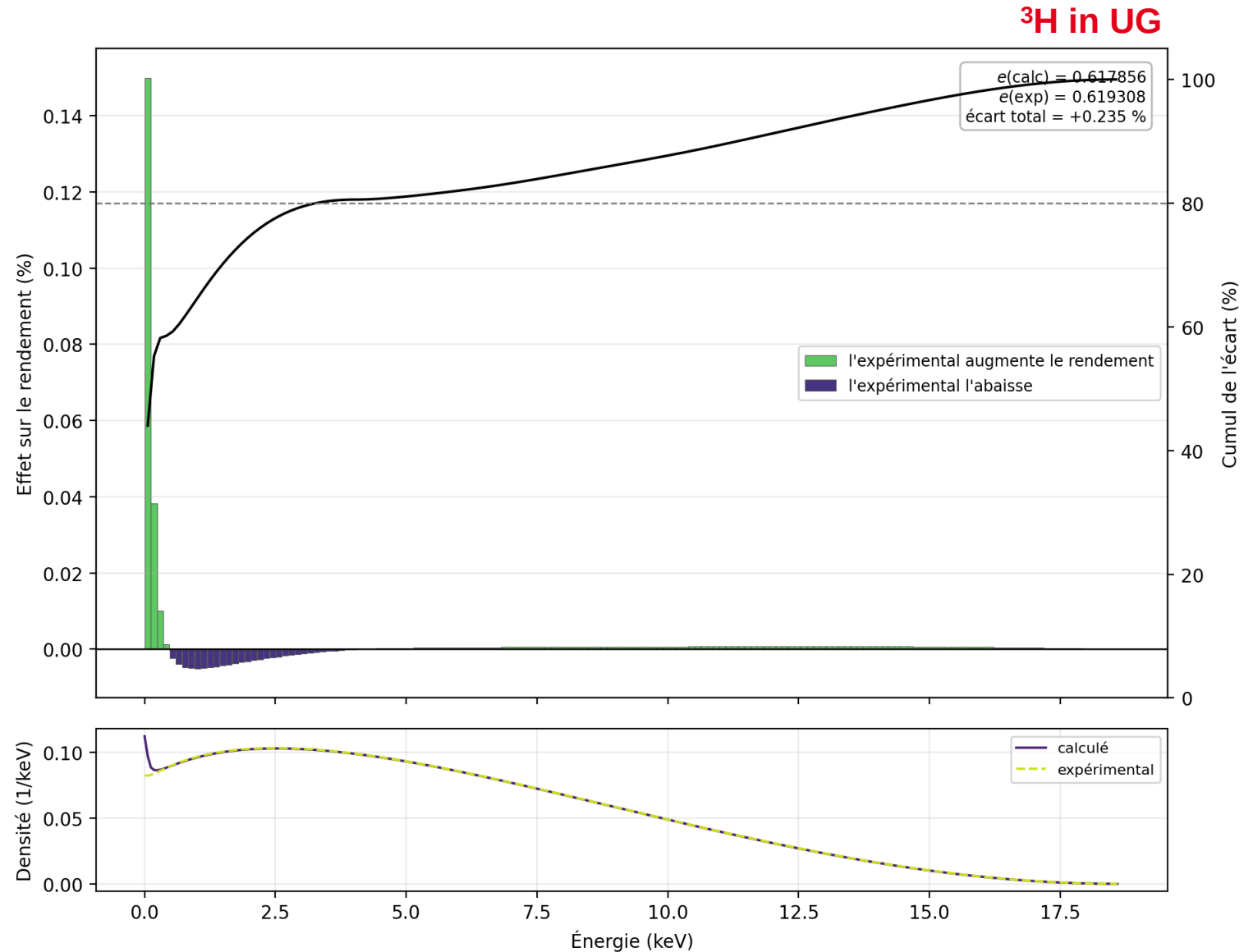
How we use the method ?

- We can also analyse the impact that a change in the spectrum has on the result, particularly as, ultimately, it is the overall spectrum that affects the measurement



How we use the method ?

- And we can already see new problems on the horizon: the spectrum needs to be characterized more precisely!
- Case of ^3H ; between experimental and calculated spectra we have a 0.2% difference below 0,5 keV it is critical. It appears in the computed spectra with the improvement of the model: exchange, screening and overlaps ...

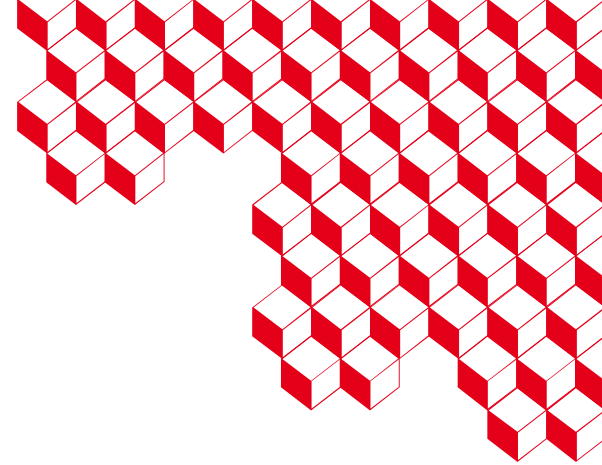




4. To summaries

Conclusion

- We showed you how important spectra are, and how they are becoming even more important for the new primary methods under development.
- The new Compton-TDCR is still in development, even though it is already operational:
 - to increase the energy range, which is currently limited by the gamma beam and angles,
 - to improve the accuracy of the response curve by using multiple GeHPs.
 - We are currently searching for funding and submitting projects; Maybe one day we will have nine GeHPs and a tunable monoenergetic source ! Stay tuned !
- Now it is time to publish all of this work and provide these dataset to the community (ongoing)



Thank you

benoit.sabot@cea.fr

Laboratoire National Henri Becquerel
CEA, Centre de Saclay