DE LA RECHERCHE À L'INDUSTRIE

Bayesian Approach for Multigamma Radionuclide Quantification Applied on Weakly Attenuating Nuclear Waste Drums

Workshop MASCOT-NUM, 21-22 November 2022

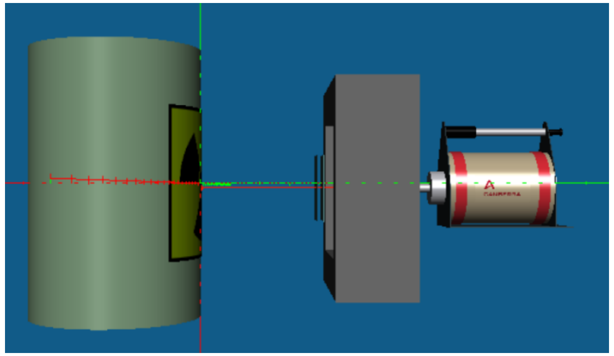
Aloïs CLEMENT

I’m sorry, but I’m a nuclear physicist.

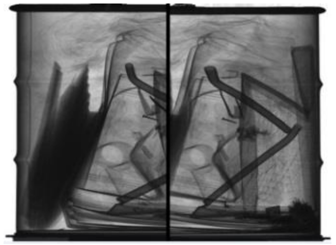
Some context

- *In situ* nuclear measurement of radioactive items

- Gamma spectrometry : HPGe 30% (Canberra)

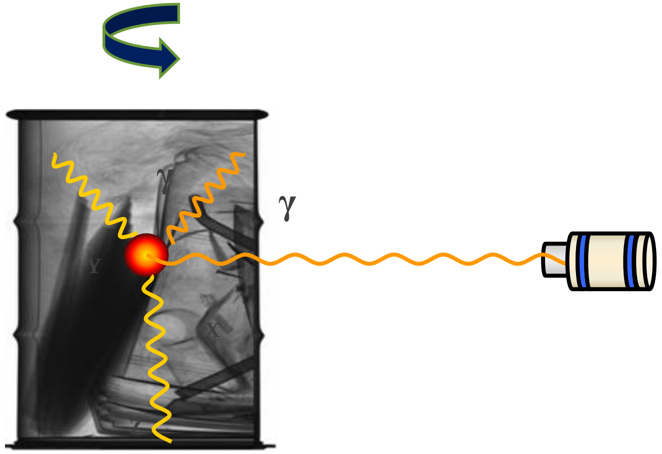


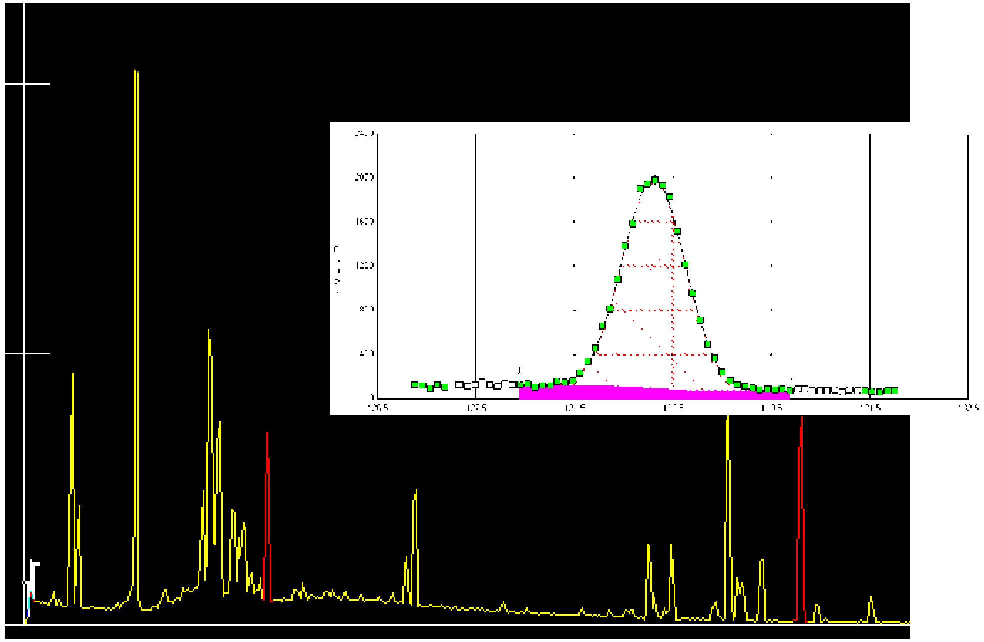
- Software : Génie 2000, ISOCS
- Waste drums : 100 et 200 L



- Actinides : U, Pu, Am, Np

- Aims : identify and quantify actinides





$$A = \frac{S(E)}{\epsilon(E, env)\ t\ I_\gamma(E)}$$

$$m = \frac{A}{A_m}$$

Multi-gamma radionuclide :

$$\mathbf{E} = (E_1, \dots, E_N)$$

$$A = \frac{\mathbf{S(E)}}{\epsilon(\mathbf{E}, env)\ t\ \mathbf{I_\gamma(E)}}$$

Let suppose :

$$\mathbf{Y^{obs}} = \frac{\mathbf{S(E)}}{t\ \mathbf{I_\gamma(E)} A_m}$$

$$\mathbf{Y^{obs}} = m\boldsymbol{\epsilon}(\mathbf{E}, env)$$

Inverse problem to solve

$$\mathbf{Y}^{\mathbf{obs}} = m\epsilon(\mathbf{E}, env) + \boldsymbol{\xi}^{\mathbf{obs}} \qquad \boldsymbol{\xi}^{\mathbf{obs}} \sim N(\mathbf{0}, \boldsymbol{\Sigma}^{\mathbf{obs}})$$

Likelihood function **Priors**

Bayes theorem : $\pi(m, env|\mathbf{Y}^{\mathbf{obs}}) = \frac{\pi(\mathbf{Y}^{\mathbf{obs}}|m, env)\pi(m|env)\pi(env)}{\int \pi(\mathbf{Y}^{\mathbf{obs}}|m, env)\pi(m|env)\pi(env)}$

Posterior

$$\pi(m, env|\mathbf{Y}^{\mathbf{obs}}) \propto \pi(\mathbf{Y}^{\mathbf{obs}}|m, env)\pi(m|env)\pi(env)$$

What I want

What I'll estimate

What I think I know

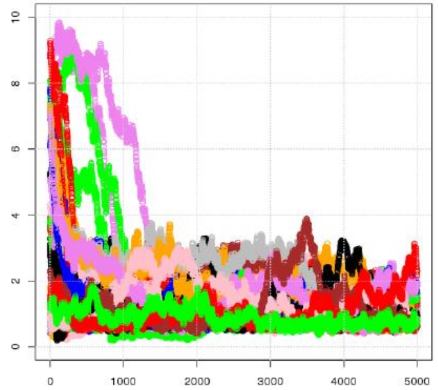
What about the « env » environment variable ?

Monte Carlo Markov Chains

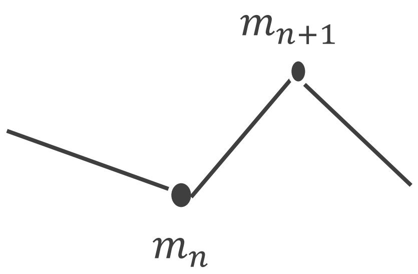
$$\pi(m,\mathbf{X}|\mathbf{Y^{obs}}) \propto \pi(\mathbf{Y^{obs}}|m,\mathbf{X})\,\pi(m|\mathbf{X})\,\pi(\mathbf{X})$$

Observation likelihood **Priors**

- Independent Y_i^{obs} : $\pi(\mathbf{Y^{obs}}|m,\mathbf{X}) = \prod_{i=1}^{N} \pi(Y_i^{obs}|m,\mathbf{X}) = \prod_{i=1}^{N} \frac{1}{\sqrt{2\pi\sigma_i}} e^{-\frac{1}{2}\frac{\left(Y_i^{obs}-m\epsilon(\mathbf{X})\right)^2}{\sigma_i^2}}$
- Independent (m, X_i) : $\pi(m|\mathbf{X})\pi(\mathbf{X}) = \pi(m)\pi(\mathbf{X}) = \pi(m)\prod_{i=1}^{D}\pi(X_i)$



MCMC : Metropolis-Hastings algorithm



$$m_{n+1} \leftarrow m_n + A.\delta, \qquad \delta \sim N(0, k_m)$$

$$\rho = \min\left(\frac{\pi(m_{n+1},\mathbf{X_{n+1}}|\mathbf{Y^{obs}})}{\pi(m_n,\mathbf{X_n}|\mathbf{Y^{obs}})}, 1\right)$$

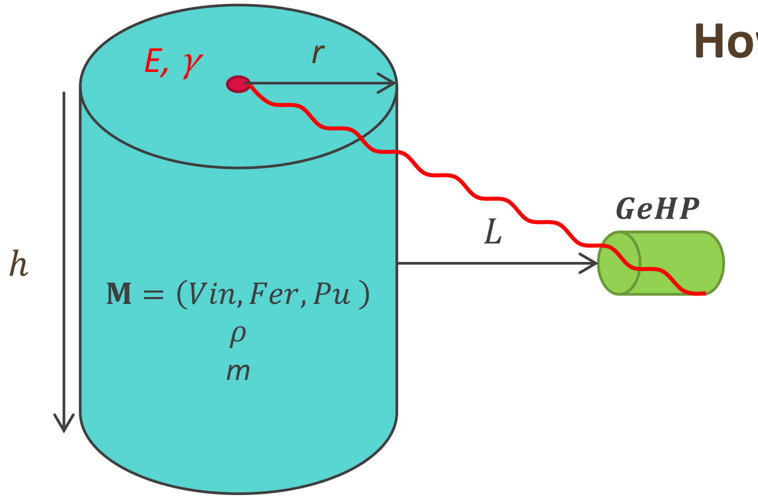
- High number of $\epsilon(\mathbf{X})$ estimations ($\approx 10^5$) by the simulation code MCNP6.2
- Time-consuming

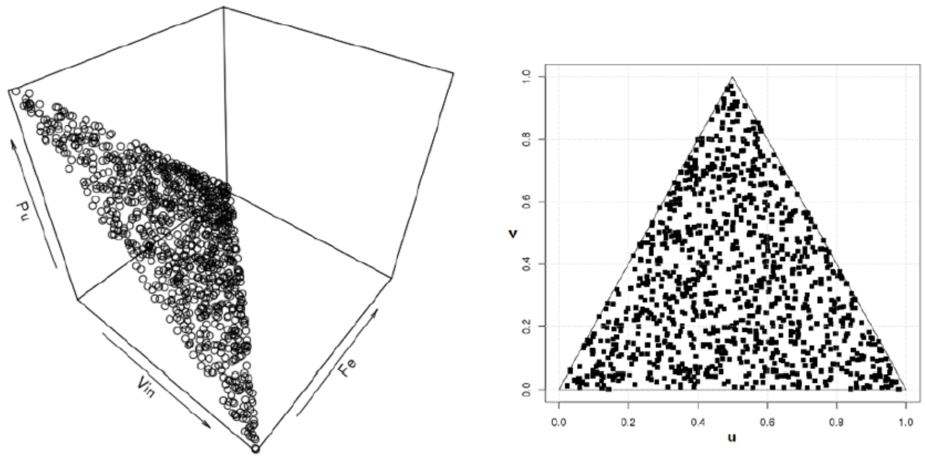
Surrogate model f of the measurement efficiency

$$\epsilon = f(\mathbf{X})$$

Kriging model

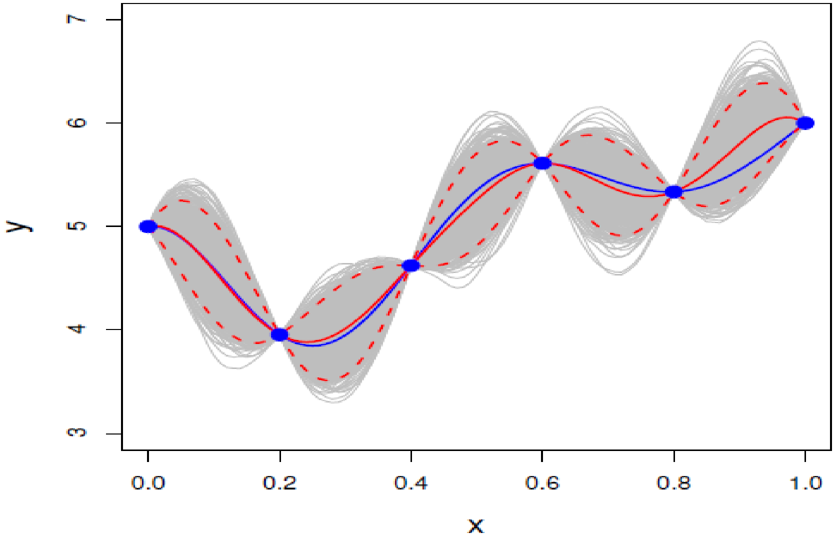
How could I estimate « *env* » and $\epsilon(\mathbf{E}, env)$?



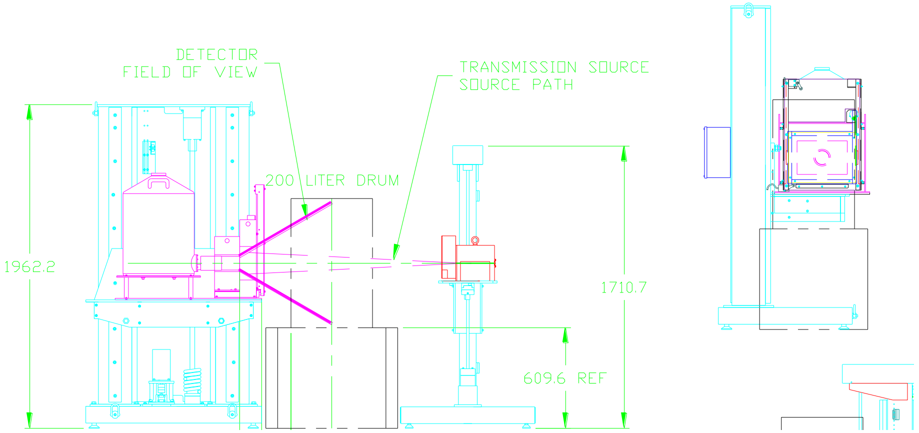


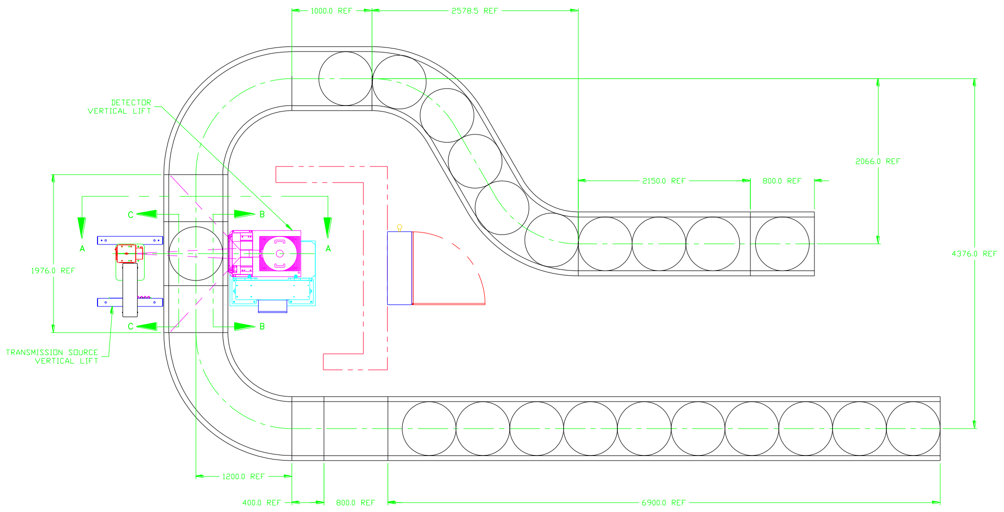
- Input: **X** = { E, r, h, **M**, L, ρ } et m (Card(**X**) = 8)
- DoE: Latin Hypercube Sampling (n = 500)
- Surrogate model: Kriging :
  - Universal Kriging (UK) : $f(X) = \mu(X) + Z(X)$
  - Deterministic : $\mu(X) = \sum_{j=1}^{p} \beta_j f_j(X)$
  - Stochastic : $Z(X)$: Matérn 5/2

- $\epsilon(\mathbf{E}, env) \approx \boldsymbol{f}(\mathbf{X}) = \boldsymbol{f}(\mathbf{E}, r, h, \rho, L, \mathbf{M})$



MADAGASCAR : Automatic Nuclear Waste Drum Measurement Device





Experimental validation with MADAGASCAR

MADAGASCAR : Validity range

- Bulk density < 0.4
- Detection limit : 1 MBq
- Maximal activity: 200 GBq
- 100L and 200L drums
- Measurement distance : ≈ 60 cm
- Global uncertainties: ± 42%

MADAGASCAR : Database

- 242 nuclear drums (100L)
- 6 plutonium standards : 0.4 to 60 grams
- Fixed measurement duration

- Fixed measurement distance : ≈ 60 cm
- Global uncertainties: ± 42%

Validation steps :

0. Convergence analysis
1. Linearity :

Assay on plutonium standards

2. Reproducibility

Repetitions on 10 items (drums)

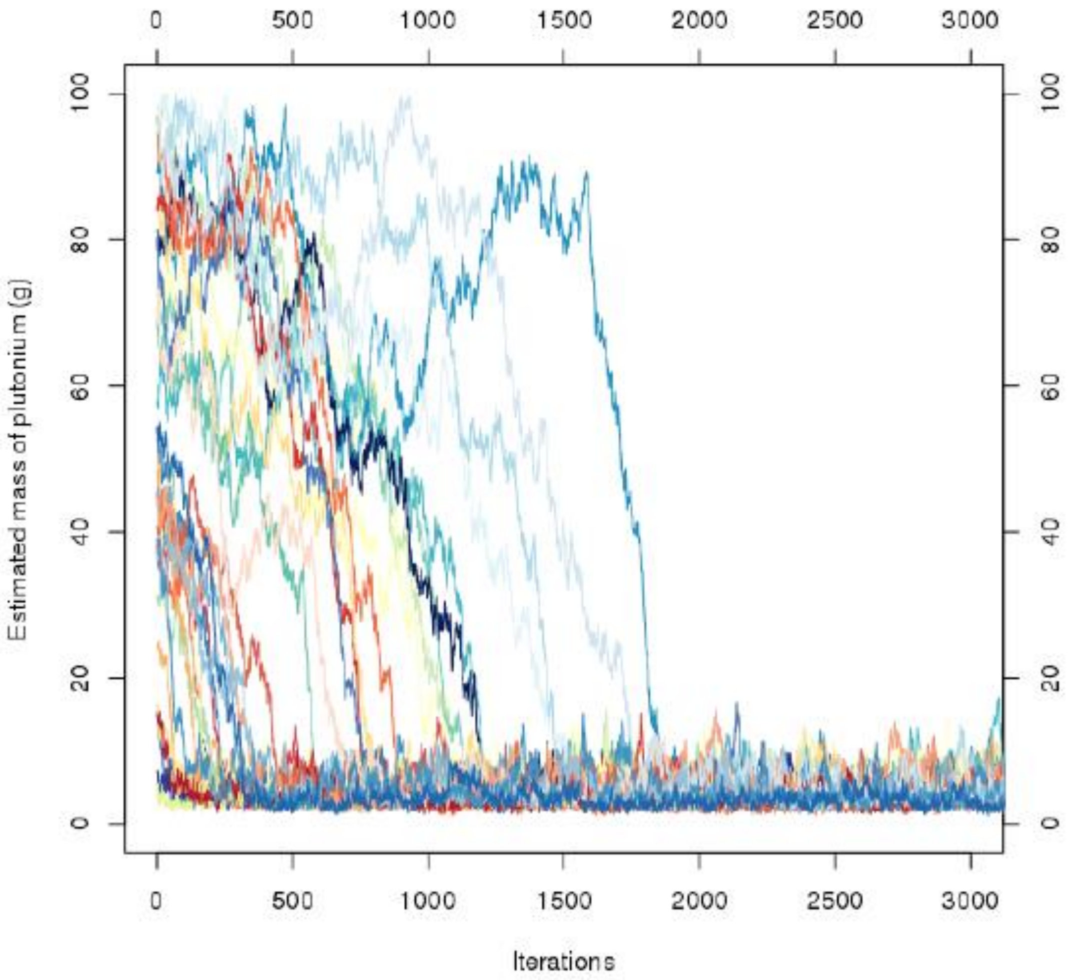
3. Comparison

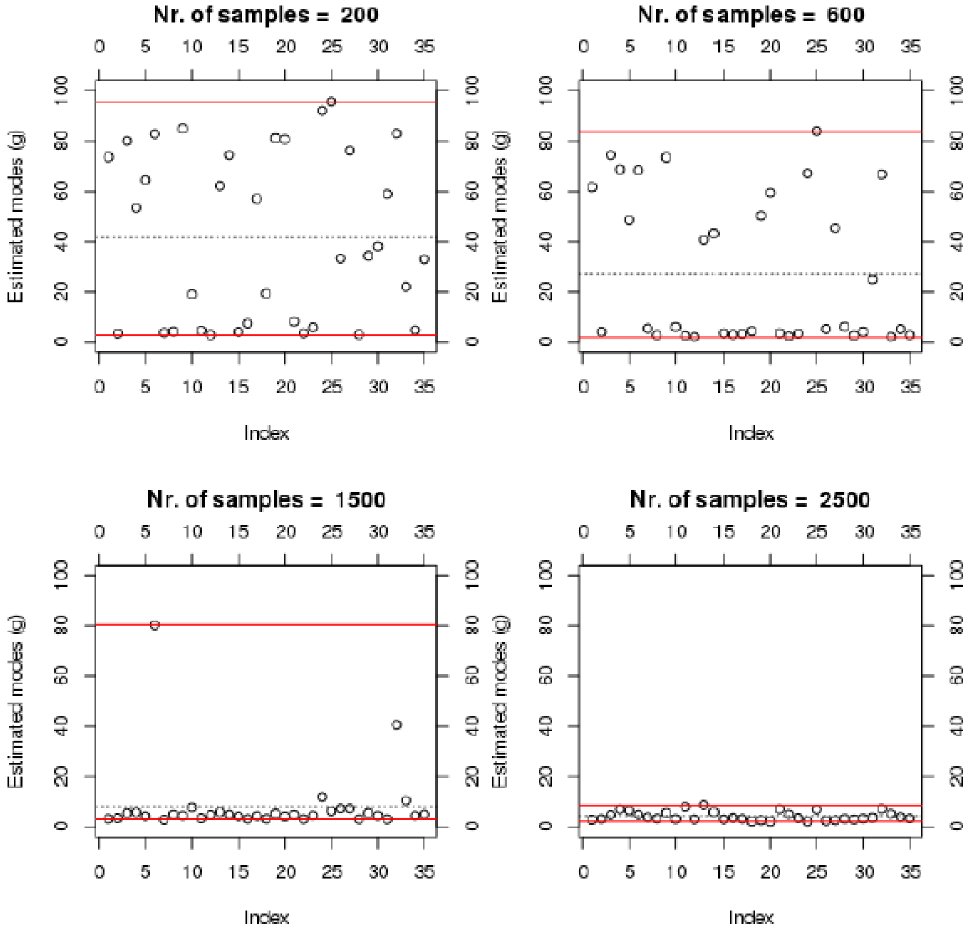
Comparison of results between MADAGASCAR and the Bayesian approach

Hardware :

- 72 cores (35 Markov chains)

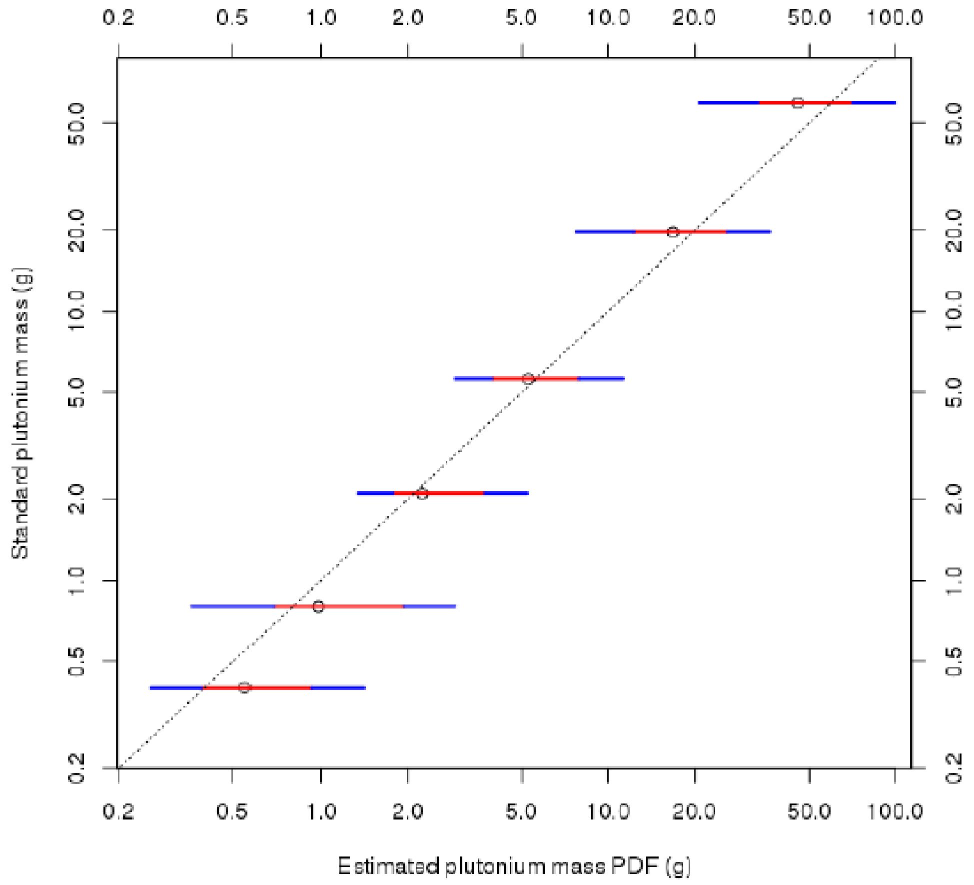
Convergence analysis





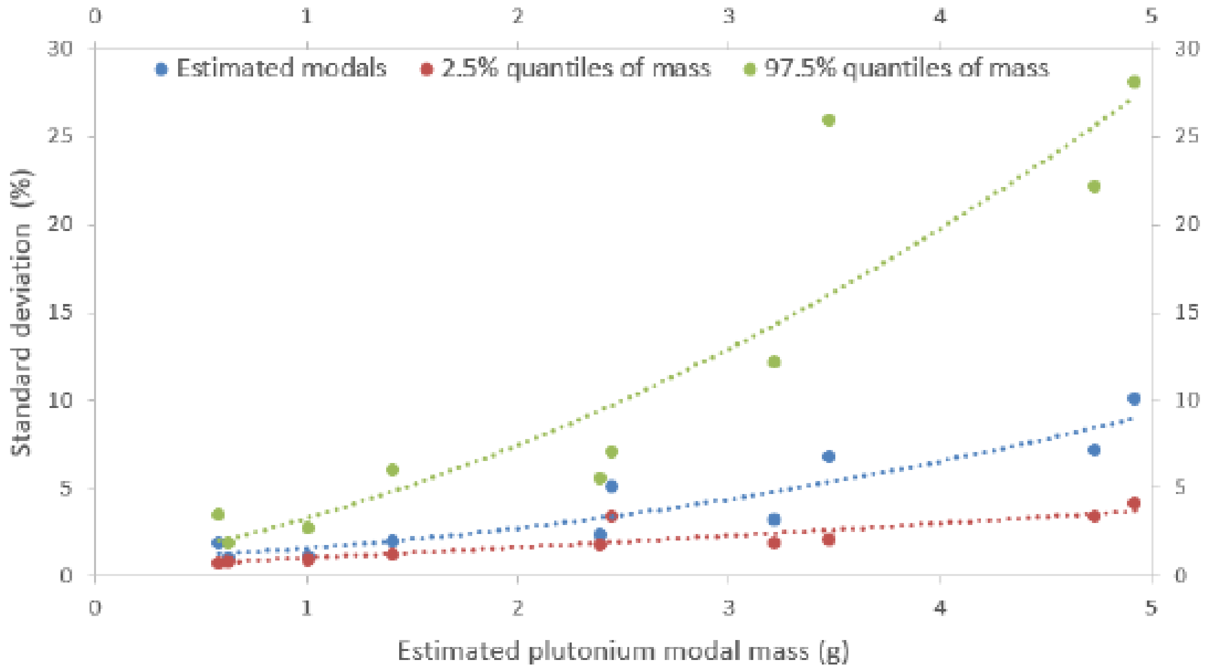
6 plutonium standards: 0.4 to 60 grams

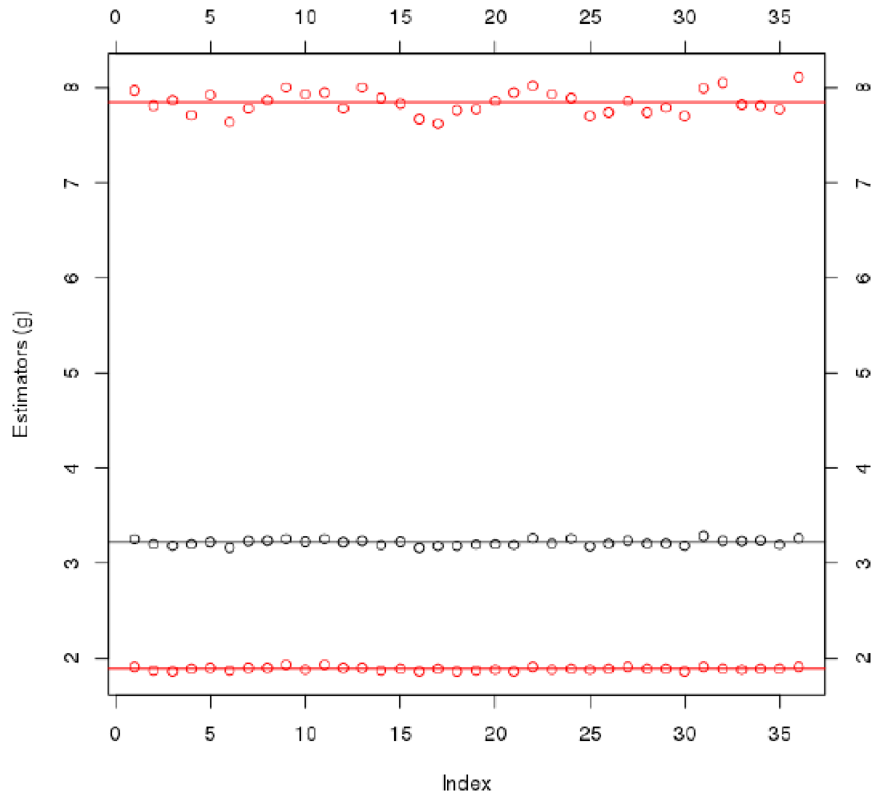
Item	M_{Pu}(g)	M_{Q1}(g)	M_{Q2}(g)	M_{Mod}(g)	M_{Q3}(g)	M_{Q4}(g)
1.	0.400	0.26	0.39	0.55	0.93	1.42
2.	0.800	0.36	0.69	0.98	1.95	2.94
3.	2.104	1.35	1.79	2.26	3.72	5.22
4.	5.619	2.93	3.94	5.27	7.91	11.3
5.	19.731	7.71	12.2	16.9	26.0	36.4
6.	59.541	20.8	33.3	45.6	71.0	99.8



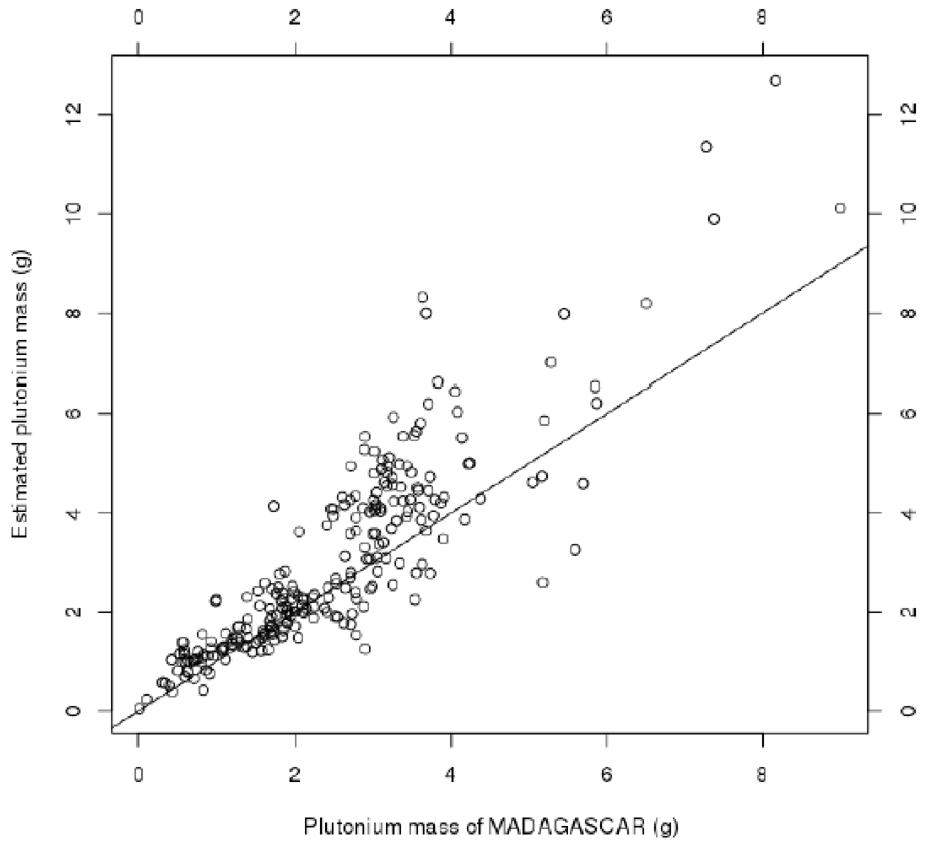
< 30 repetitions on 10 items (drums)

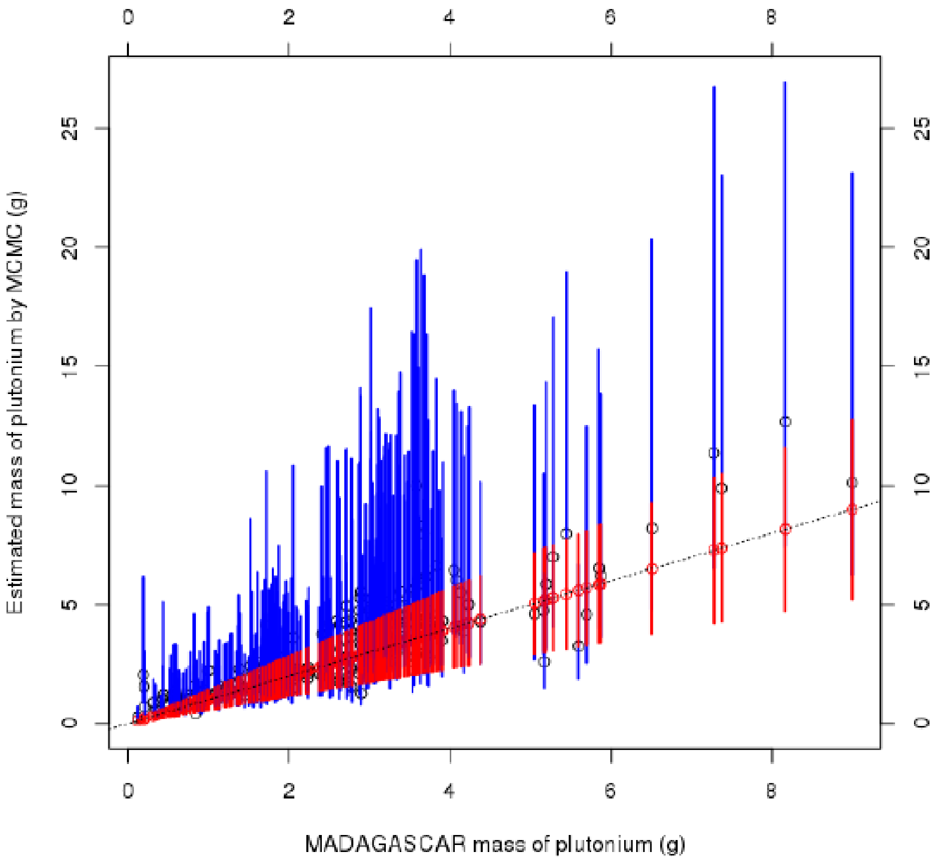
Calculation duration: 15 min





Experimental comparison





91% of estimated masses ∈ CI95% of MADAGASCAR

92,3% of estimated mass PDF overlap 50% of the MADAGASCAR CI95% (±42%)

Conclusion

- Validation : good results on plutonium standards
- Comparison : good results on simple cases : low bulk density (< 0,4 $g.cm^3$)
- Useful priors for physicists : bulk density, compositions, Gaussian, Uniform, etc.
- Easy to code (Python) and possibilities to use specific libraries
- Breaking point with classical approach : scalars vs PDF
- Available to other RN (Am, FP, activated)

- Multigamma RN only
- Increasing uncertainties

Thanks for your attention