Meta-analysis of key comparison data in radionuclide metrology

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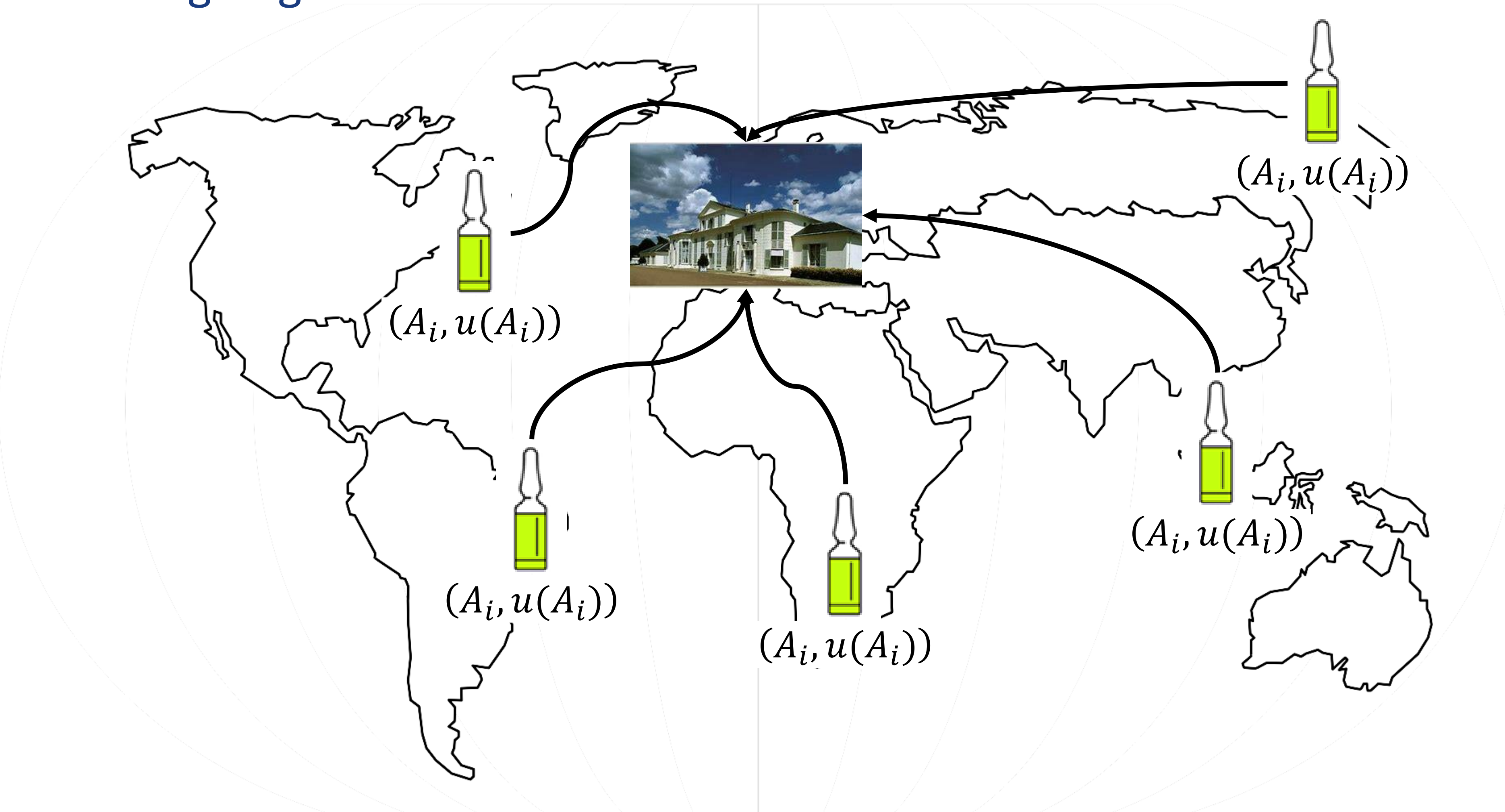
The International System of Reference for radionuclide metrology

- An ongoing centralised measurement service since 1976

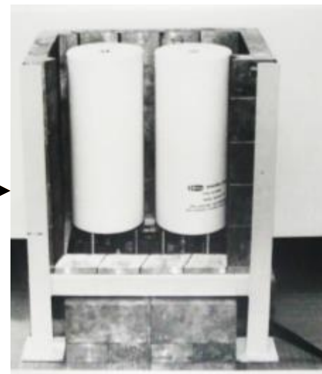


The International System of Reference for radionuclide metrology (2)

- A transfer instrument based on ionization chamber measurement

$^{226}\mathrm{Ra}_j, j = \{1,2,3,4,5\}$

$$F_j = \frac{I_{Ra,5}}{I_{Ra,j}}$$



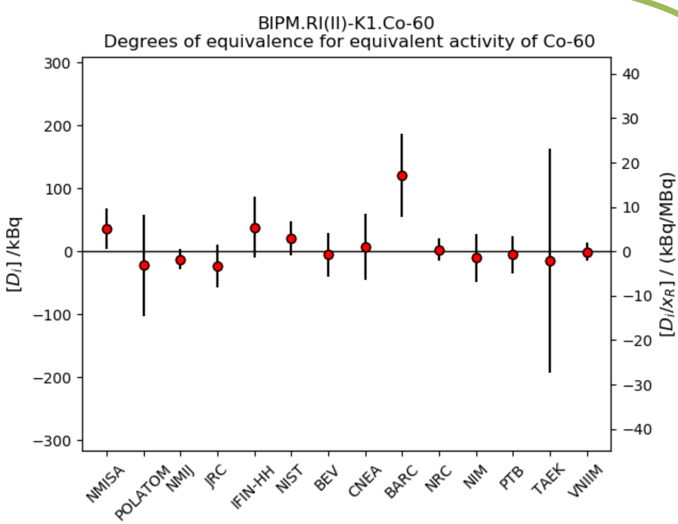
$(I_{\mathrm{Ra},j}, u(I_{\mathrm{Ra},j}))$

$(I_i, u(I_i))$

$(A_i, u(A_i))$

Equivalent Activity

$$A_{\mathrm{e}i} = A_i \frac{I_{Ra,j}}{I_i} F_j$$



$$KCRV = PMM(A_{\mathrm{e}i})$$

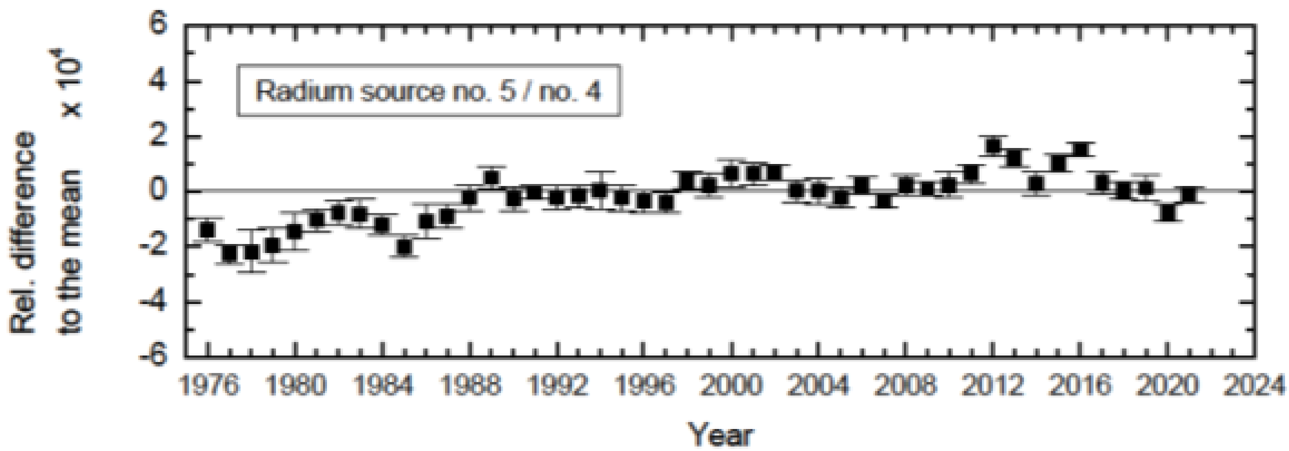
$$D_i = A_{\mathrm{e}i} - KCRV$$

Key assumptions (1)

Equivalent Activity

$$A_{ei} = A_i \frac{I_{Ra}}{I_i} F_j$$

- Equivalent activities are assumed to be robust over the long term because:
  - The approach (using a ratio) is intrinsically independent to experimental fluctuations of the efficiency of the ionisation chamber



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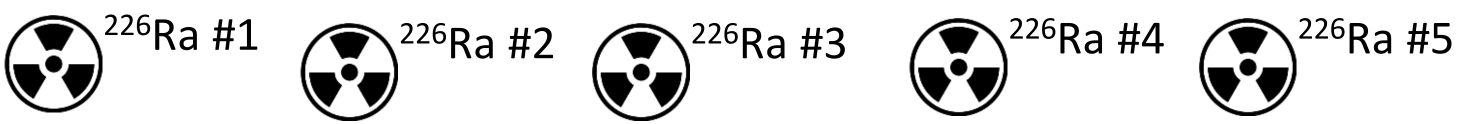
Key assumptions (2)

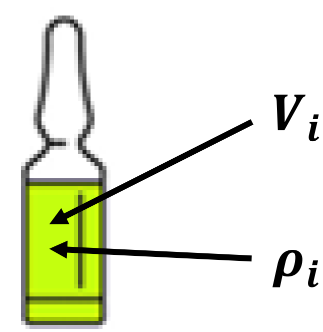
Equivalent Activity

$$A_{ei} = A_i \frac{I_{Ra}}{I_i} F_j$$

- Other hypothesis are made:
  - The homogeneity of batch of glass ampoules with respect to wall thickness
    - Mitigation: only a unique batch stored by the BIPM is used
  - The robustness of the SIR against the choice among the different radium sources
    - Monitoring: the current ratio between reference sources is monitored



  - The robustness of the SIR against the properties of the solution to be measured, notably the filling height and the density.
    - Mitigation:
      - A range for the mass is specified to participant - 3.6 ± 0.2 g
      - No action with density



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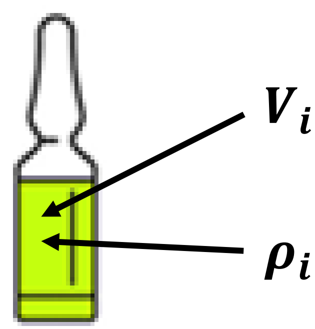
Assessment of the robustness of the SIR

- The database of the SIR has been updated in a machine readable format permitting to easily implement statistical analysis of the data

[R. Coulon *et al.*, *Meas. Sci. Tech.* **33** 024003, 2022]

- Knowing the data accumulated since 1976,
  - Is the SIR robust against the choice among the five radium reference sources?

  - Is the SIR robust against encountered changes in solution density and volume?



Data preconditioning

- The SIR provide one KCRV per radionuclide. To aggregate the data in a common figure of merit, relative degrees of equivalence are used

$$\dot{D}_i^{(rj)} = \frac{D_i^{(rj)}}{KCRV^{(r)}}$$

- The standard uncertainty of the degrees of equivalence is the combination of the uncertainty form the laboratory $u\left(A_i^{(r)}\right)$ and the uncertainty from the SIR $u_{SIR}\left(A_{\mathrm{e}i}^{(rj)}\right)$,

- The uncertainty considered in this study is only the SIR component

$$u\left(\dot{D}_i^{(rj)}\right) = \frac{u_{SIR}\left(A_{\mathrm{e}i}^{(rj)}\right)}{KCRV^{(r)}}$$

Data resampling

- To propagate uncertainties through Monte-Carlo method, data set are resampled in a Normal distribution

$$\delta_i^{(rjz)} \sim \mathcal{N}\left(\dot{D}_i^{(rj)}, u^2\left(u\left(\dot{D}_i^{(rj)}\right)\right)\right)$$

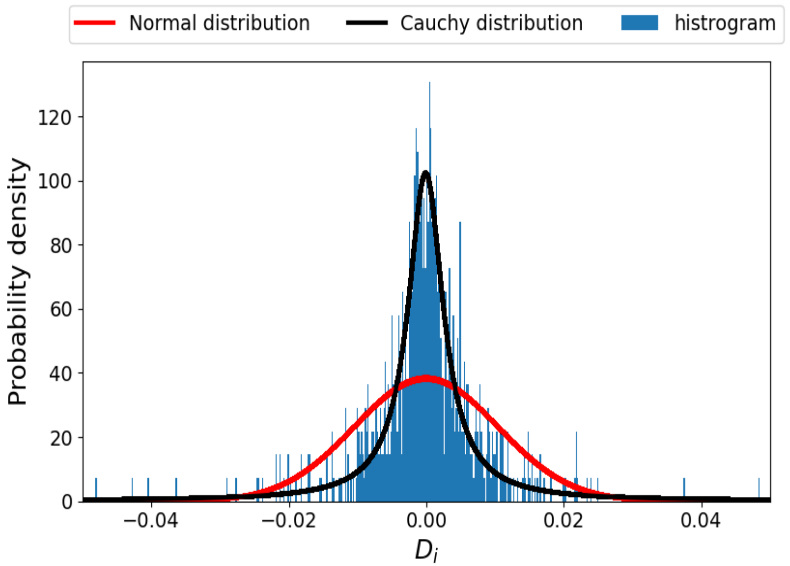
Where,

- *r* is the radionuclide
- *i* is the laboratory
- *j* is radium source number
- *z* is the data set (MC trial number)

Model of the data (1)

- To find the model that best fit the data
  - Parameters maximizing the likelihood of the parametric distributions are estimated
  - Using these parameters, a Kolmogorov-Smirnov (KS) test is applied for each distribution assumptions

Model	MLE p_1	MLE p_2	KS test p-value
Normal	$-3(1)\ 10^{-4}$	$1.19(8)\ 10^{-2}$	**$0(3)\ 10^{-13}$**
Cauchy	$-1.3(6)\ 10^{-4}$	$3.54(4)\ 10^{-3}$	**0.31(6)**



Aggregated degrees of equivalence

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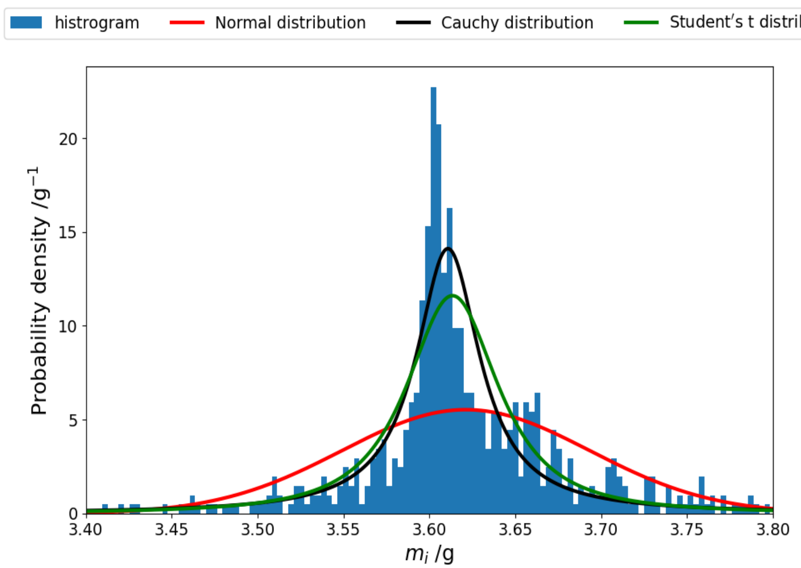
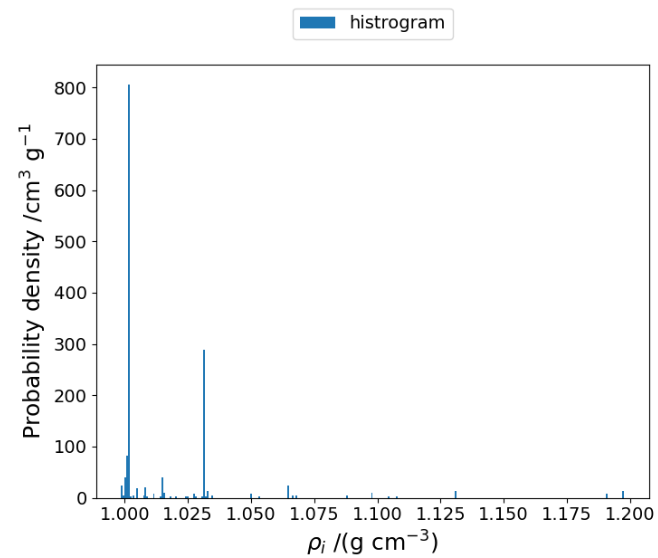
Model of the data (5)

- Normal hypothesis can also be rejected for the mass and density parameters





- Marginal distributions of the SIR output, mass and density of the solution are not Normal
- As a consequence, non parametric tests are implemented

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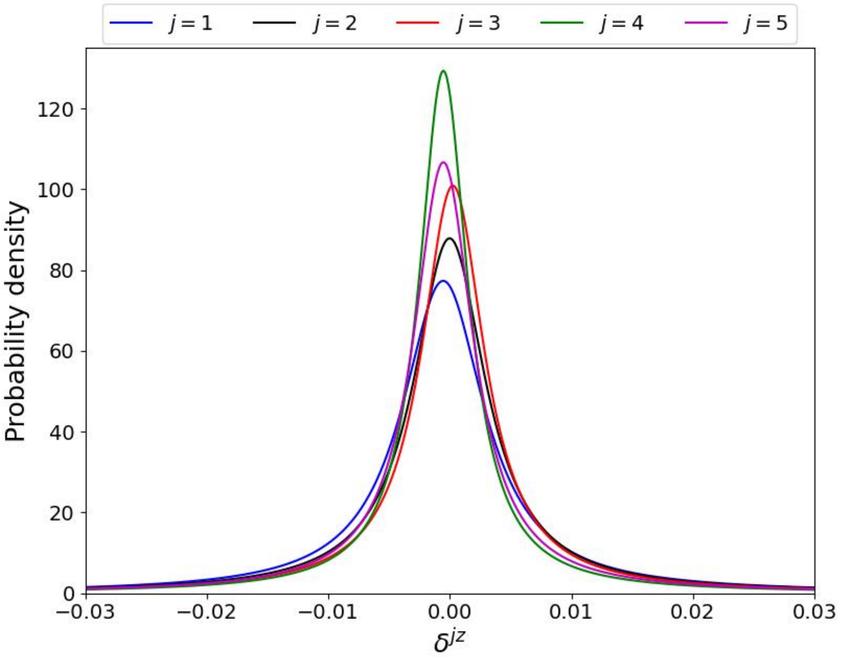
Correlation with the choice of the reference source (1)

- To test the absence of correlation with the choice among the 5 reference sources $j = \{1, 2, 3, 4, 5\}$, the Kruskal-Wallis and Mood's median tests were implemented.
  - $\boldsymbol{p}$**−values** $= \mathbf{0.61(15)}$ **and** $\mathbf{0.68(19)} > \mathbf{0.05}$
  - There is no evidence at 95 % confidence that the SIR measurement delivers different expectation values as a function of the reference source used.

Cauchy-fitted Distributions of relative degrees of equivalence for the subgroups defined by the five ^{226}Ra sources



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Correlation with the properties of submitted solutions (1)

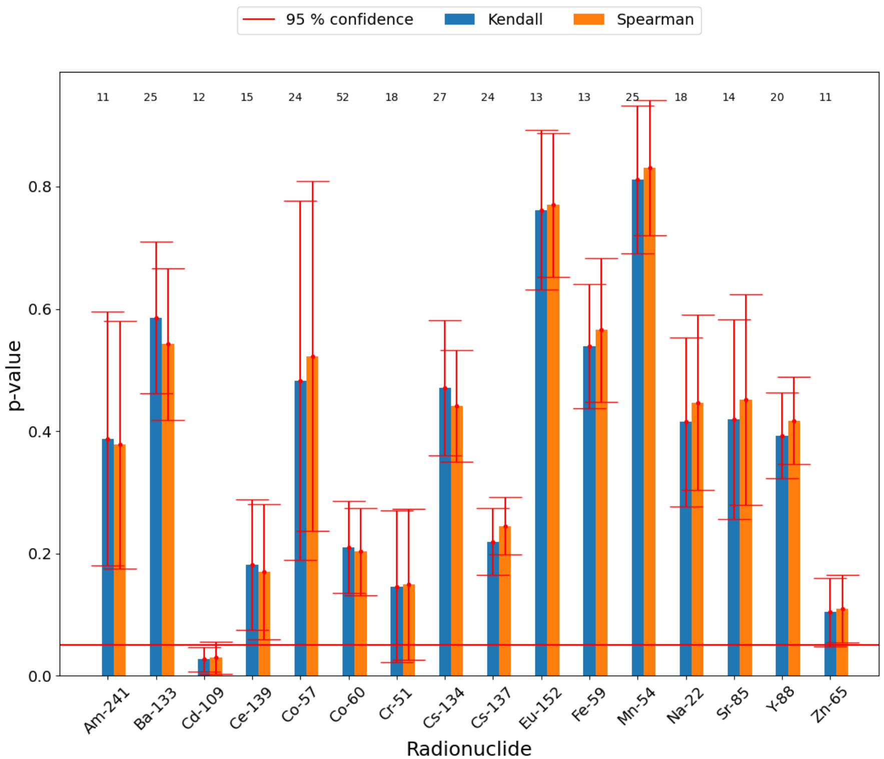
- Kendall tau $\boldsymbol{\tau}$ and Spearman's rank r correlation coefficients are calculated
  - $\boldsymbol{p}$**−value of** $\boldsymbol{\tau_{\delta,m}}$ **and** $\boldsymbol{r_{\delta,m} = 0.12(7) > 0.05}$
  - The null hypothesis that "**there is no correlation between the SIR measurement and the mass of submitted solution**" cannot be rejected with a confidence of 95%.
  - This corroborate an initial experimental analysis stating that restraining the volume of solution in the range 3.6 ± 0.2 g should make negligeable the impact of the source height on SIR measurements.

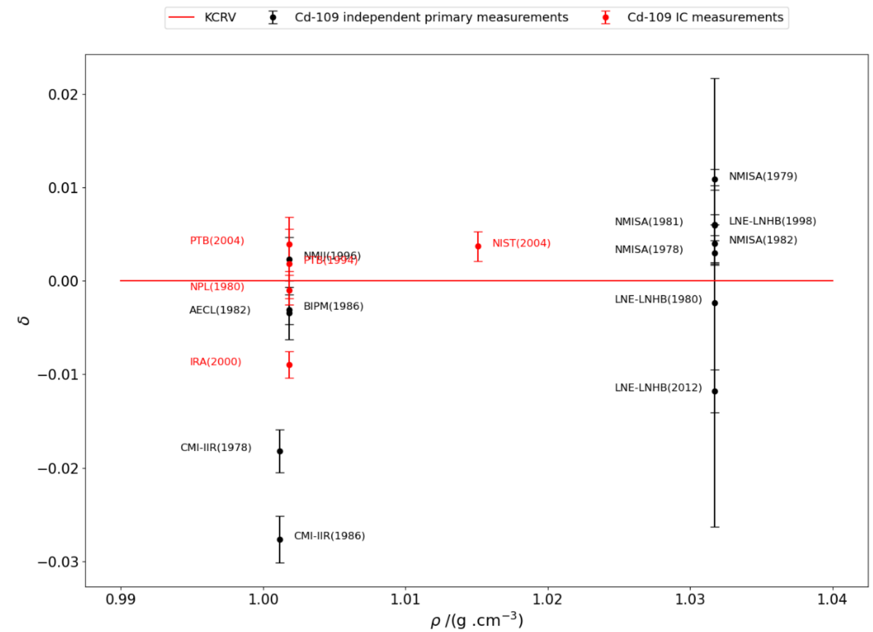
[A. Rytz, *Environment International* **1** 15, 1978]

  - $\boldsymbol{p}$**−value of** $\boldsymbol{\tau_{\delta,\rho} = 0.078(8)}$ **and** $\boldsymbol{p}$**−value of** $\boldsymbol{r_{\delta,\rho} = 0.10(1) > 0.05}$
  - The null hypothesis that "**there is no correlation between the SIR measurement and the density of submitted solution**" cannot be rejected with a confidence of 95%.
  - The impact of the density is strongly radionuclide-dependent because correlated with the energy of x rays and γ rays
  - Some experimental studies have underlined the significant impact of the solution density with ionization chamber measurement when the emitted radiation are with low energy – notably for ^{241}Am (Michotte, for the SIR) and ^{57}Co (Cessna, for other types of ICs)

[J.T. Cessna et al., *Applied Radiation and Isotopes* ***109*** *402–4*, 2016]
[C. Michotte et al., *Metrologia* **54** 06010, 2017]

Correlation with the density of submitted solutions





- A significant correlation between the SIR measurement and solution density has been revealed for the ^{109}Cd (88 keV γ-rays and x-rays below 26 keV)

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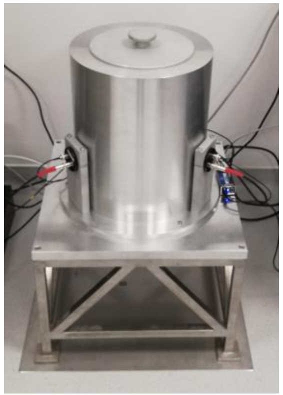
Conclusion and perspectives

- A meta-analysis was conducted using the new key comparison database
  - Kolmogorov-Smirnov tests concluded
    - The model of the measurement data is not normally distributed but rather Cauchy distributed
  - Non-parametric null hypothesis tests concluded
    - The absence of correlation between the SIR measurement and the reference ^{226}Ra source used
    - The absence of correlation between the SIR measurement and the mass of submitted standard solution
    - The absence of correlation between the SIR measurement and the density of submitted standard solution **except for low energy ones emitting only low energy x/γ-rays (e.g. ^{109}Cd)**

- The **extension of the SIR** based on liquid scintillation counting will improve the robustness of BIPM services **for radionuclides where the SIR cannot measure or is proven not to being robust**.



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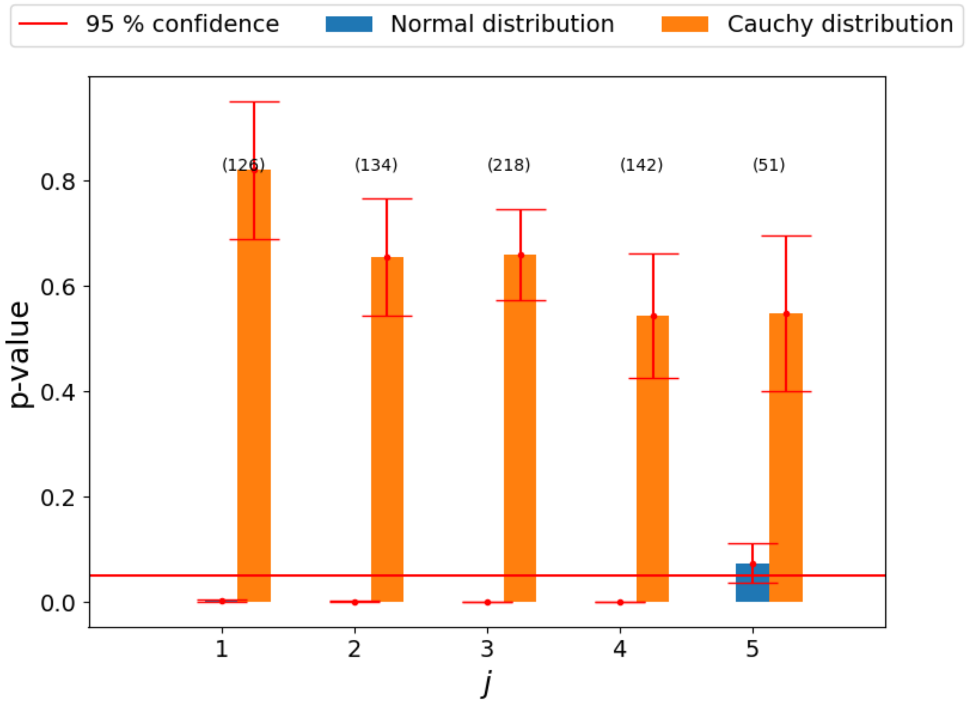
Back up

Model of the data (2)

- When looking at the data grouped per radium sources



The Normal hypothesis can be rejected with a confidence level of 95% for all the radium sources excerpt the number 5

The Cauchy distribution better fit the data

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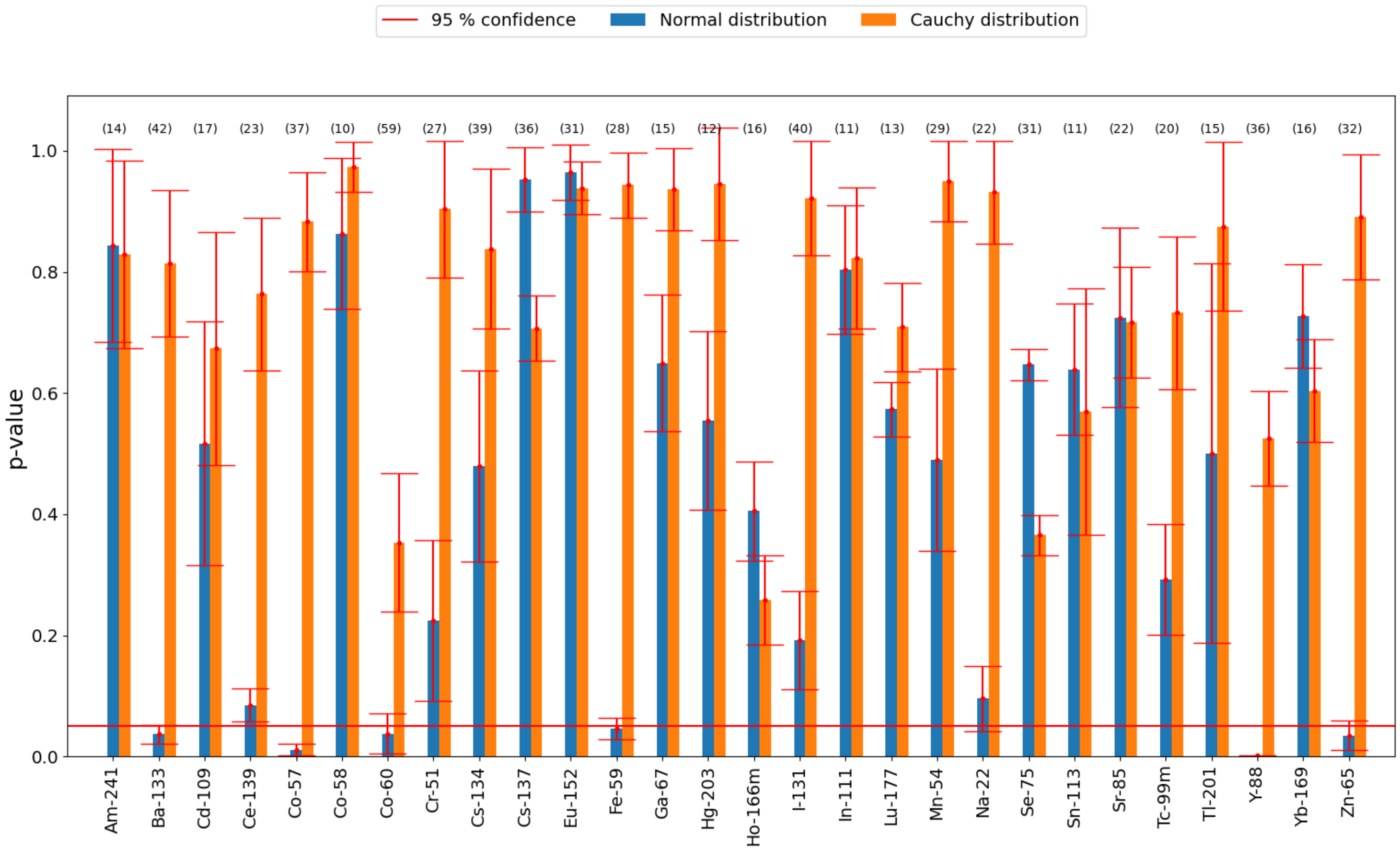
Model of the data (3) - $p(D_i, r)$

- When looking at the data grouped per radionuclides



The Normal hypothesis can be rejected for ^{133}Ba, ^{57}Co, ^{60}Co, ^{59}Fe, ^{88}Y and ^{65}Zn

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Model of the data (4)

- The deviation for the Normal hypothesis can come from
  - The aggregation of SIR measurements (independent measurements - outliers)
  - The SIR measurement model

Normal r.v.

Normal r.v.

$$? \longrightarrow \boldsymbol{A_{\mathbf{e}i} = A_i \frac{I_{Ra}}{I_i}}$$

Normal r.v.

  - We tend to observe a compliance with the normal hypothesis when:
    - $\boldsymbol{u(I_i) \ll u(A_i)}$
  - Indeed, the ratio of normally distributed r.v. is Cauchy distributed
  - So, the normality of the SIR output value cannot be systematically assumed