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EURAMET.RI(II)-K2.Ac-225

Activity of radionuclide Ac-225

KEY COMPARISON

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Final report of the EURAMET.RI(II)-K2.Ac-225 comparison of the activity measurements of a solution of ^{225}Ac in equilibrium with its progeny

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Abstract

A key comparison of the measurement of the activity per unit mass of a solution of ^{225}Ac in equilibrium with its progeny, was organized in 2024. Flame sealed ampoules filled with master solution of ^{225}Ac were prepared and distributed by NPL, based on the provision of ^{225}Ra by the CERN-MEDICIS platform in the framework of the PRISMAP project. Seven laboratories participated, and all the submitted final values were obtained using primary measurements based on liquid scintillation (i.e., TDCR and CIEMAT/NIST methods). A dedicated ampoule was also submitted to the International Reference System (SIR) at BIPM for a future link to the BIPM.RI(II)-K1.Ac-225 comparison using the activity measurement provided by LNE-LNHB. This report describes the main aspects of the comparison, including the values submitted by the participants, used to calculate the Comparison Reference Value (CRV) and provisional degrees of equivalence for each laboratory. This study was supported by the project 22HLT03 AlphaMet, which has received funding from the European Partnership on Metrology (EPM), co-financed from the European Union's Horizon Europe Research and Innovation Programme and by the participating States.

1. Introduction

Targeted Alpha-particle therapy (TAT) is a growing area of research and development as a cancer treatment modality, with presently only $^{223}RaCl_2$ having regulatory approval. Due to the short penetration range and high linear energy transfer of alpha-particles, which can result in a higher cytotoxic dose to a tumour cell over a shorter range than beta-particles, TAT is showing promising efficacies and increased survival in early phase clinical trials. The AlphaMet project (Metrology for Emerging Targeted Alpha Therapies), started in 2023 for a duration of three years, is a European research project funded by the EURAMET R&I programme that brings together experts from National Metrology Institutes (NMIs), hospitals, academia and industry to support end-to-end traceability before wide routine adoption of TAT. In particular, the AlphaMet project aims at establishing the metrological traceability of emerging alpha-emitting radiopharmaceuticals such as ^{211}At, ^{212}Pb/^{212}Bi and ^{225}Ac in order to accelerate their development and safe use for clinic application.
The present comparison was carried out to compare the activity per unit mass of a master solution of ^{225}Ac in equilibrium with its progeny. This exercise was declared as an EURAMET.RI(II)-K2.Ac-225 key

comparison between seven radionuclide laboratories involved in the AlphaMet project. The list of the participating laboratories is provided in Table 1. The comparison is registered under the number 1682 in the EURAMET database. A dedicated ampoule was also prepared from the same master solution of ^{225}Ac and submitted to the BIPM for a future link to the BIPM.RI(II)-K1.Ac-225 comparison (SIR). This EURAMET.RI(II)-K2.Ac-225 was piloted by LNE-LNHB and NPL.

Table 1. Radionuclide laboratories involved in the EURAMET.RI(II)-K2.Ac-225 comparison.

NMI/DI	Full name	Country	Contact person
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	Spain	M. Roteta Ibarra
CMI	Český metrologický institut	Czech Republic	J. Sochorová V. Dutka
ENEA/INMRI	Instituto Nazionale di Metrologia delle Radiazioni Ionizzanti	Italy	M. Capogni
CHUV/IRA	Institut de Radiophysique	Switzerland	F. Juget
CEA/LNE-LNHB	Laboratoire national Henri Becquerel	France	C. Bobin M.-N. Amiot
NPL	National Physical Laboratory	United Kingdom	S. M. Collins
POLATOM/NCBJ	Narodowe Centrum Badań Jądrowych Ośrodek Radioizotopów POLATOM	Poland	J. Marganiec-Gałązka

2. Measurand

The measurand for this comparison is the activity per unit mass of ^{225}Ac, in equilibrium with the decay progeny. The measurement results of the participating laboratories are reported at the following reference date and time:

4 October 2024 at 12:00 UTC

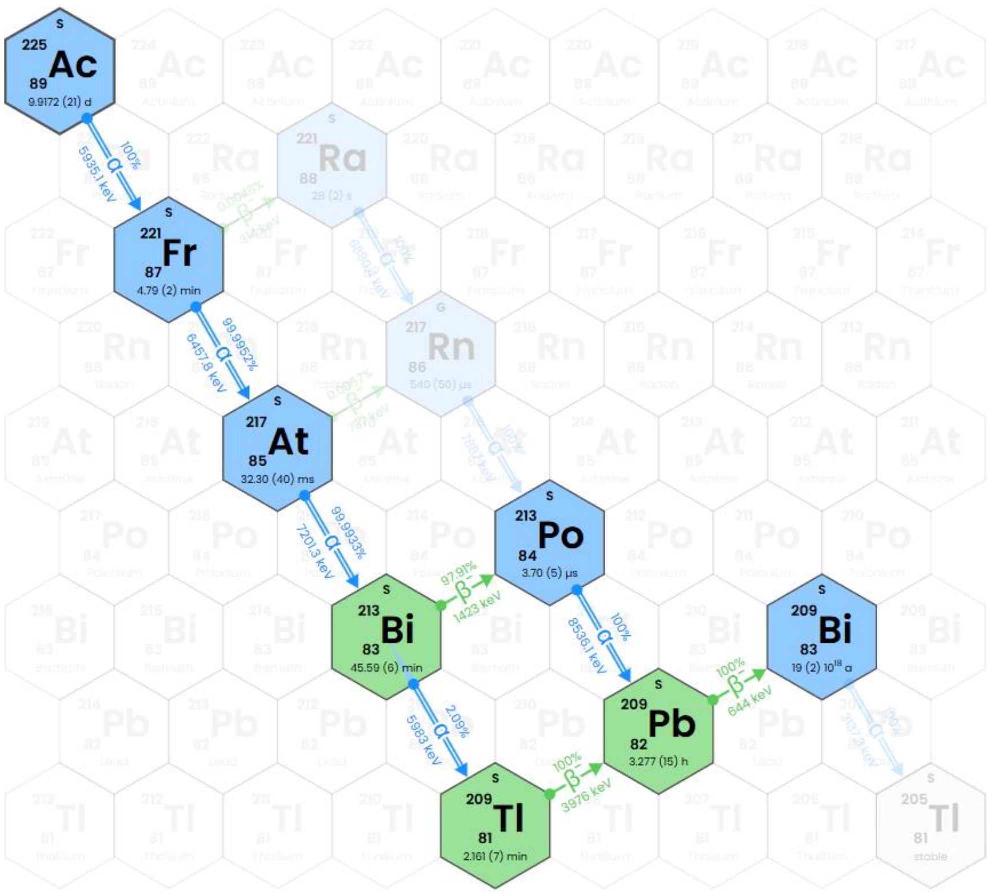

Figure 1. Decay chain of ^{225}Ac (http://www.lnhb.fr/Laraweb/).

Recommended nuclear data:	Decay Data Evaluation Project for ^{225}Ac [1] and its progeny and BetaShape calculations [2] ^{225}Ac half-life: $T_{1/2}$ = 9.9172(21) d ^{221}Fr half-life: $T_{1/2}$ = 4.801(5) min ^{217}At half-life: $T_{1/2}$ = 32.3(4) 10^{-3} s ^{213}Bi half-life: $T_{1/2}$ = 45.59(6) min ^{213}Po half-life: $T_{1/2}$ = 3.70(5) 10^{-6} s ^{209}Tl half-life: $T_{1/2}$ = 2.161(7) min ^{209}Pb half-life: $T_{1/2}$ = 3.277(15) h

It was part of the exercise for each participant to identify and measure the activity of any possible impurity.

3. Preparation

For this comparison, ^{225}Ra as a salt was received at NPL in September 2024 from CERN-MEDICIS in the framework of the PRISMAP project (PRISMAP – The European medical radionuclides programme, H2020 grant agreement No 101008571). The chemical separation to extract the ^{225}Ac, remove the decay progeny, and the preparation of the ampoules with ^{225}Ac solution were carried out at NPL before shipment to the participating laboratories. The chemical composition of the ^{225}Ac solution was in the form of actinium chloride ($^{225}Ac(III)Cl_3$) in 0.1 M hydrochloric acid. The flame sealed ampoules were prepared with about 5 g aliquots of solution. The information related to each ampoule is given in Table 2.

Table 2. Information related to each ampoule of ^{225}Ac sent to the participating laboratories.

Ampoule identification	Participating laboratory	Active mass (g)
A240631	LNHB	4.979669
A240632	IRA	5.000167
A240633	CIEMAT	5.022977
A240634	ENEA	4.974693
A240635	CMI	5.006983
A240636	POLATOM	4.995221
A240637	NPL	4.997250

The homogeneity check was carried out before shipment by γ-spectrometry measurements using the energy peak at 440 keV related to the decay of ^{213}Bi, when in equilibrium with the ^{225}Ac. The results of the homogeneity check are displayed in Fig 2.

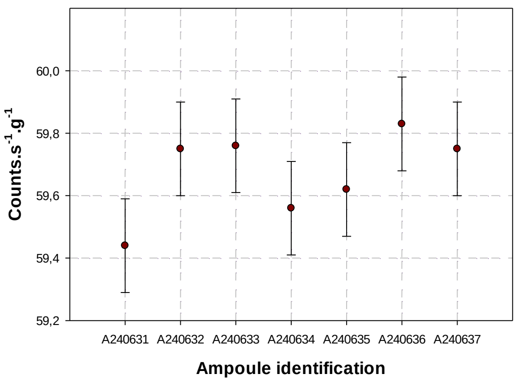

Figure 2. homogeneity check carried out before shipment by γ-spectrometry measurements at NPL.

A BIPM ampoule containing 3.59538(36) g of ^{225}Ac solution having the same chemical characteristics as described above, was shipped to the BIPM by NPL for a future link to the BIPM.RI(II)-K1.Ac-225 comparison (SIR) using the measurement result provided by LNE-LNHB.

4. Results and measurement methods

The results submitted by the participating laboratories and the measurement methods used, are listed in Table 3. All the adopted values correspond to primary measurements based on liquid scintillation counting techniques. The TDCR method was applied by all the participants. The final result submitted by CHUV/IRA is given by the arithmetic mean of the TDCR and CIEMAT/NIST values. Other methods were also implemented. Measurements were carried out at LNE-LNHB and CHUV/IRA using $4\pi\gamma$ integral counting based on a well-type NaI(Tl) detector. The $4\pi\beta$–γ coincidence method was applied by two laboratories: CIEMAT and CHUV/IRA. CIEMAT submitted a result based on absolute $2\pi\alpha$ counting with a grid ionization chamber. A value obtained by γ-spectrometry was given by CHUV/IRA. All the results are given in Table 3 and displayed in Fig. 3. The uncertainty budgets and information about measurements are available in the Appendix.

Table 3. Measurement results submitted by the participating laboratories.

NMI / DI	Acronym of the measurement method	Activity per unit of mass (kBq g^{-1})	Value adopted by the participants (kBq g^{-1})	Relative standard uncertainty (x 100)
CIEMAT	4P-LS-MX-00-00-TD	225.6(10)	225.6	0.4
	4P-PPC-AP-NA-GR-CO	226.37(270)		
	2P-IC-AP-00-00-00*	225.9(11)		
CMI	4P-LS-MX-00-00-TD	224.97(90)	224.97	0.38
ENEA	4P-LS-MX-00-00-TD	227.53(161)	227.53	0.71
IRA	4P-LS-MX-00-00-TD	223.363(991)	223.30**	0.44
	4P-LS-MX-00-00-CN	223.229(867)		
	4P-NA-MX-00-00-HE	225.56(128)		
	4P-PS-PO-4P-NA-GR-CO	227.85(355)		
	UA-Ge(HP)-GR-00-00	223.95(315)		
LNHB	4P-LS-MX-00-00-TD (Hidex 300 SL)	225.73(81)	225.41	0.36
	4P-LS-MX-00-00-TD (RCTD1)	225.41(81)		
	4P-NA-MX-00-00-HE	226.5(15)		
NPL	4P-LS-MX-00-00-TD	224.44(57)	224.44	0.25
POLATOM	4P-LS-MX-00-00-TD	225.0(8)	225.0	0.35

* Measurement based on absolute 2π-α counting with a grid ionization chamber.
** The final result of IRA is obtained by the arithmetic mean of the TDCR and CIEMAT/NIST values.

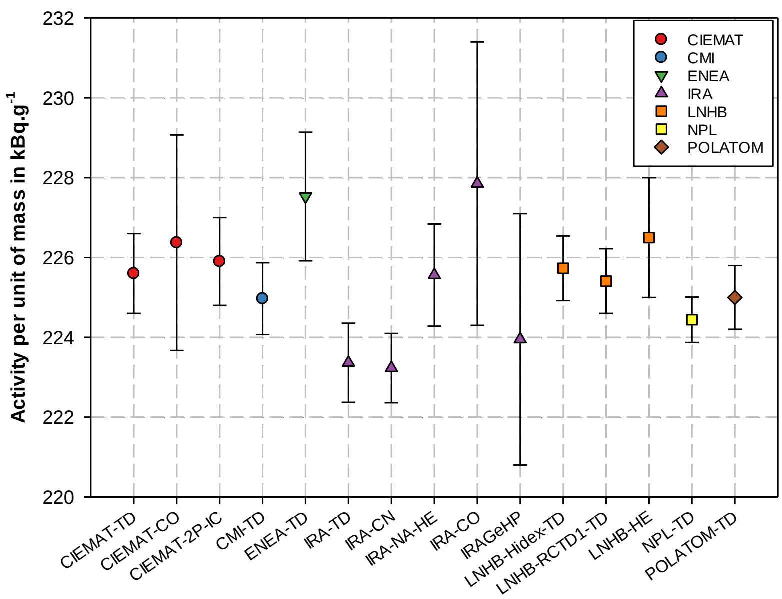

Figure 3. Measurement results provided by each participating laboratory.

Impurities were checked by each laboratory using γ-spectrometry. The presence of ^{225}Ra was reported by NPL and POLATOM. The measured impurity to ^{225}Ac activity ratios were equal to 5.47(20) 10^{-4} and 1.85(18) 10^{-4}, respectively. In both cases, the contribution of the contaminant was considered as negligible and no correction was applied.
Regarding the adopted values based on liquid scintillation, the Birge ratio is close to 1 (1.14) indicating that the data set is consistent and the standard uncertainties are suitable.

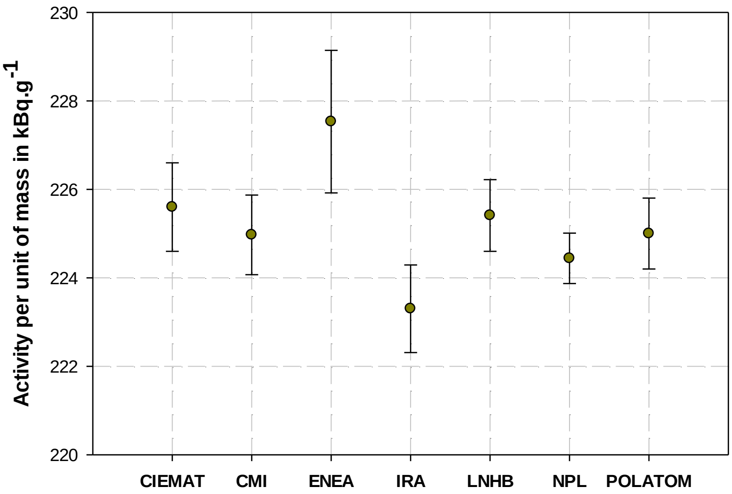

Figure 4. Adopted values of the participants.

5. Comparison reference value and provisional degrees of equivalence

The comparison reference value (CRV) was estimated using the power-moderated mean [3] applied to all the results adopted by each laboratory given in Table 3 and displayed in Fig. 4. The calculation was implemented using a value of α equal to 2-3/N, where N is the number of results, corresponding to informative but imperfect uncertainties [3]. The resulting CRV is equal to 224.94(39) kBq.g^{-1}. Comparisons with arithmetic and weighted means are given in Table 4. The variation between the estimators remains lower than their associated uncertainties.

Table 4. Comparison between the values given by the PMM, the arithmetic mean and weighted mean.

	C_A (kBq.g^{-1})	u (kBq.g^{-1})
PMM value	224.94	0.39
Arithmetic mean	225.18	0.49
Weighted mean	224.87	0.32

Table 5. Provisional degrees of equivalence and related expanded uncertainties (k = 2) for each laboratory i.

Laboratory	D_i (kBq.g^{-1})	U_i (kBq.g^{-1})
CIEMAT	0.66	2.03
CMI	0.03	1.84
ENEA	2.59	3.22
IRA	-1.64	2.00
LNHB	0.47	1.68
NPL	-0.5	1.26
POLATOM	0.06	1.66

The provisional degrees of equivalence D_i for each participating laboratory, i, with the CRV are obtained as the difference of the activities per unit of mass, a_i, given in Table 3 with respect to the CRV

$$D_i = a_i - \text{CRV}$$

and the expanded uncertainty (k = 2) of the difference, U_i, is calculated as described in [3]. The results are given in Table 5 and displayed in Fig. 5.

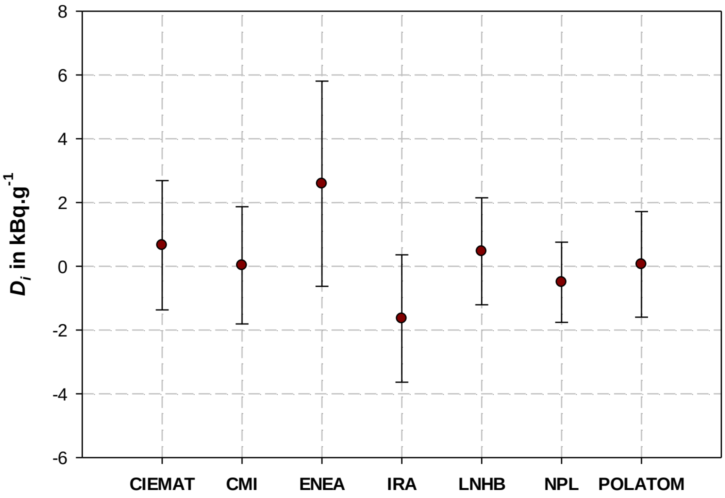

Figure 5. Provisional degrees of equivalence (DoE) with the CRV. The uncertainty bars represent the expanded uncertainty (k = 2) of the DoE.

6. Conclusion

The EURAMET.RI(II)-K2.Ac-225 comparison was carried out in the framework of the European AlphaMet project in September 2024. Seven radionuclide laboratories participated and all the adopted values correspond to primary measurements based on liquid scintillation counting, mainly obtained using the TDCR method. The Birge ratio calculated from these results is close to 1 indicating that the data set is consistent and the standard uncertainties are adequate, with no outliers identified. The CRV, estimated using the power-moderated mean, is equal to 224.94(39) kBq.g^{-1}. Provisional degrees of equivalence for each participating laboratory calculated with this CRV, are also proposed. In the future, a link to the BIPM.RI(II)-K1.Ac-225 comparison will be established using the activity measurement provided by LNE-LNHB

7. Reference

[1] Mougeot X., Huang X. and Wang B., http://www.lnhb.fr/nuclides/Ac-225_tables.pdf
[2] Mougeot X., 2015. Erratum: reliability of usual assumptions in the calculation of β and ν spectra. Phys. Rev. C92, 059902.
[3] Pommé S. and Keightley J., 2015. Determination of a reference value and its uncertainty through a power-moderated mean. Metrologia 52, S200-S212.

8. Acknowledgements

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Appendix

Table A1. LS counters and measurement parameters for TDCR (4P-LS-MX-00-00-TD) and CIEMAT/NIST (4P-LS-MX-00-00-CN) methods

NMI/DI	LS counter Method	Age (y)	PMT	Resolving Time (ns)	Dead time type Duration (μs)	Efficiency variation	Acquisition	Scintillator	Type of LS vial	Typical TDCR value	Maximum detection efficiency	Number of sources	Typical count rate (s^{-1})
CIEMAT	In-house TDCR	9	ET 9807b	40	Extending 40 Live time	/	MAC3	UG	low potassium borosilicate glass	0.9948	5.0067	/	3100
CMI	In-house TDCR	8	Hamamatsu R331-05	40	Extending 40 Live time	Grey filters	In-house electronics	UG	low potassium borosilicate glass Polyethylene	0.992	5.012	6	2000
ENEA	Hidex 300SL TDCR	15	ET 9102	40, 60, 80,100	Non-extending 40 Live time	/	Hidex Metrology	UG	low potassium borosilicate glass	0.992	/	3	16000
IRA	In-house TDCR	5	Hamamatsu R331-05	80	Extending 28	CCl_4	MAC3	UG UG AB	low potassium borosilicate glass HD Polyethylene	0.997	5.05	12 Glass/UGAB 11 HDPE/UG	6000
	TriCarb 2700 CIEMAT/NIST	18	Hamamatsu	/	/	CCl_4	/	UG UGAB	low potassium borosilicate glass	N.A.	5.17	5 UG 5 UGAB	2000
LNHB	In-house TDCR	> 30	BURLE 8850	40	Extending 50 Live Time	Grey filters	MAC3	UG AB	low potassium borosilicate glass	0.9924	5.008	8	2500
	Hidex 300SL TDCR	1	ET 9102	40	Non-extending 40 Live time	/	Hidex Metrology	UG AB	low potassium borosilicate glass	0.9945	5.016	8	2000
NPL	In-house TDCR	10	Hamamatsu R6095	40	Extending 50	Nitrobutane	In-house electronics	Optiscint MP	HD Polyethylene	0.99	~5	8	2400
POLATOM	In-house TDCR	9	ET 9214B	90	Extending 90 Live time	/	In-house TDCRG	UG UG AB	low potassium borosilicate glass	0.99	5.02	12 / UG 12 / UGAB	7585

Table A2. Counters and measurement parameters for 4πγ NaI(Tl) integral counting

NMI/DI	NaI(Tl) counter	Discrimination level	Dead time	Dead time duration (μs)	Dead time measurement	Background rate (s^{-1})	Typical Source rate (s^{-1})	Maximum detection efficiency of ^{225}Ac
IRA	Well type 127 mm diameter 127 mm height	22.6 keV	Non extending	15	Two oscillator technique	27	4000	0.557 Geant4 simulation
LNHB	Well type 152 mm diameter 127 mm height	10 keV	Extending	25	Live time technique MTR2 module	15	2600	0.571 Geant4 simulation

Table A3. Counters and measurement parameters for 4πβ-γ coincidence counting

NMI/DI	β-channel	γ-channel	Type of source	Digital coincidence system	Dead time	Resolving time	Extrapolation	Detection efficiency correction
CIEMAT	Pressurized proportional counter Argon (90%) Methane (10%) 3 10^5 Pa	NaI(Tl)	VYNS solid source	Yes	Non extending	3.7 μs	No extrapolation	/
IRA	Plastic scintillators optically coupled to a photomultiplier tube (Hamamatsu R1924A)	Well type 4"x4" NaI(Tl)	Solid plastic scintillator	No	Non extending Two oscillator technique	0.975 μs	Electronic discrimination Linear function	5.047(20)

Table A4. Uncertainty budgets for the results submitted by CIEMAT (4P-LS-MX-00-00-TD and 4P-PPC-AP-NA-GR-CO).

CIEMAT - 4P-LS-MX-00-00-TD			CIEMAT - 4P-PPC-AP-NA-GR-CO		
Quantity	***u*(*a*)/*a* in %**	**Comments**	**Quantity**	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.14		Counting statistics	0.6	
Weighing	0.08	From calibration certificate	Weighing	0.06	
Background		Included in counting statistics	Background	0.5	
Dead/live time	0.05	Live time clock	Dead/live time	0.7	
Resolving time	0.05		Resolving time	0.5	
Decay data	0.19		Decay data	0.3	
Extra-/inter-polation of efficiency curve	0.14	Effect of TDCR uncertainty on detection efficiency	Extra-/inter-polation of efficiency curve	0.12	
Value of *kB* parameter	0.015	Efficiency variation depending on the value established for kB parameter			
Scintillation composition	0.12	From data obtained with 2 scintillation cocktails			
Nuclear data	0.012	Effect of beta spectra on detection efficiency calculations (beta spectra obtained from EFFY9 and BetaShape codes)			
Dilution factor	0.004				
Calculation of TDCR Parameter	0.13	Effect of counting statistics on TDCR calculation			
PMT asymmetry	0.1				

Table A5. Uncertainty budgets for the results submitted by CMI (4P-LS-MX-00-00-TD) and ENEA (4P-LS-MX-00-00-TD).

CMI - 4P-LS-MX-00-00-TD			ENEA - 4P-LS-MX-00-00-TD		
Quantity	***u*(*a*)/*a* in %**	**Comments**	**Quantity**	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.1		Counting statistics	0.3	
Weighing	0.15		Weighing	0.3	
Background	0.05		Background	0.01	
Dead/live time	0.1		Dead/live time	0.4	
Decay data	0.3	Sum of model and decay data uncert.	Resolving time	0.15	
Decay correction	0.01		Decay data	0.2	
Impurities	0.1		Impurities		Not detected
Adsorption	0.05		Decay correction	0.1	
			Extra-/inter-polation of efficiency curve	0.3	Interpolation of the efficiency curve computed for Double Coincidences as function of TDCR parameter as reported in the paper Broda et al, ARI 201 (2023) 110987. This uncertainty component also considers the contribution to the total combined uncertainty due to the model and code used.

Table A6. Uncertainty budgets for the results submitted by IRA (4P-LS-MX-00-00-TD and 4P-LS-MX-00-00-CN).

IRA - 4P-LS-MX-00-00-TD			IRA - 4P-LS-MX-00-00-CN		
Quantity	**$u(a)/a$ in %**	**Comments**	**Quantity**	**$u(a)/a$ in %**	**Comments**
Counting statistics	0.17	The counting statistics contribution was estimated using one thousand sets of correlated random Gaussian net coincidence counting rates generated for each of the counting points of a defocusing measurement. These sets of Monte Carlo simulated rates were computed in such a way as to reproduce the corresponding experimental moments and covariance matrices. These rates were then fed into the code that calculates the efficiencies and activities. The ESTAR stopping powers were used and kB was fixed at 0.075 mm/MeV. The standard deviation of the distribution of the activities thus generated was assumed to be the propagation of the rates' pdfs on the activity.	Counting statistics	0.1	Statistical standard deviation of the mean count rates observed during repeated counting of sources
Weighing	0.17	Δm/m for the lightest source of the sources used	Weighing	0.17	Δm/m for the lightest source of the sources used
Background	0.01	One thousand sets of correlated random Gaussian background coincidence counting rates were generated for each counting point. The simulated rates have mean values, standard deviations of the means and covariance matrices in agreement with the measured background rates. Corresponding net counting rates were generated by subtracting the Monte Carlo background rates from the measured gross counting rates. The 10^3 defocusing data sets were then fed into the code that computes the efficiencies and activities. The standard deviation of the distribution of the activities thus obtained was taken as the propagation of the background uncertainty on the activity.	Background	0.01	ΔB/Rmin where ΔB the maximum dispersion of the background rate, while Rmin is the smallest Tricarb countrate measured
Decay scheme	0.028	Propagation of uncertainties on Bi-213, Tl-209, and Pb-209 beta branch intensities and maximum energies on the efficiencies	Dead/live time	0.15	
Beta spectra	0.041	Relative differences between using Geant4 default spectra and BetaShape spectra for Bi213, Tl209 and Pb209	Beta spectra	0.04	
Decay correction	0.042	Propagation of the half-life uncertainty on decay correction factor	Decay data	0.03	Propagation of uncertainties on Bi-213, Tl-209, and Pb-209 beta branch intensities and maximum energies on the efficiencies
kB parameter and Q(*E*)	0.2	Separate sets of 2×10^3 uniform deviates of kB lying between 0.0075 and 0.0120 cm/MeV were randomly generated, assuming a rectangular distribution, and then coupled with five ionisation quenching functions to compute altogether 10^4 activities. Four of these ionisation quenching functions are obtained from the ESTAR stopping powers (Berger, 1993), and those of (Gümüs et al., 2010) and (Tan et al., 2012), whereas the fifth is that of (Grau Malonda et al., 1999). The relative standard deviation of the 10^4 activities thus generated is about 0.2 %.	Quenching	0.1	Propagation of the tracer activity and tSIE uncertainties on total efficiency
Dilution factor	0.06		Tracer		Included above
Sample repeatability	0.31	Relative standard deviation of activities of the 23 UG-made sources measured with three efficiency variation methods	Decay correction	0.047	Propagation of the half-life uncertainty on decay correction factor
			kB parameter and Q(*E*)	0.2	Relative standard deviation of activities obtained with *kB* = 0.0075 and 0.012 cm/MeV, and 2 prescriptions for the stopping powers used to calculate Q(E) (ESTAR and Tan & Xia).

			Dilution factor	0.006	
			Timing	0.003	Time base error
			Sample repeatability	0.19	Relative standard deviation of activities of the 9 sources measured thrice on two Tricarbs

Table A7. Uncertainty budgets for the results submitted by IRA (4P-NA-MX-00-00-HE and 4P-PS-PO-4P-NA-GR-CO).

IRA - 4P-NA-MX-00-00-HE			IRA - 4P-PS-PO-4P-NA-GR-CO		
Quantity	***u*(*a*)/*a* in %**	**Comments**	**Quantity**	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.1	Statistical standard deviation of the mean count rates observed during repeated counting of sources	Counting statistics	0.369	
Weighing	0.059	Δm/m for the lightest source of the sources used	Weighing	0.017	
Background	0.072	ΔBγ/Rγmin where ΔBγ the maximum dispersion of the γ-background rate during the campaign, while Rγmin is the smallest γ-count rate measured	Background	0.023	
Dead/live time	0.02	Δτ × ργ where Δτ is the uncertainty of the deadtime and ργ is a typical true gamma countrate	Dead/live time	0.02	
Decay data	0.2	Propagation of the decay scheme parameter uncertainties on the MC efficiency	Resolving time	0.62	
Decay correction	0.03	Propagation of the half-life uncertainty on decay correction factor	Extra-/inter-polation of efficiency curve	0.776	
Timing	0.002	Worst case time base error	Decay correction	0.092	
Energy threshold	0.264	Propagation of the energy threshold uncertainty (+/- 2 keV) on the Monte Carlo efficiency	Reproducibility	1.295	
Source geometry	0.219	Propagation of the volume uncertainty of GSV/PSV sources on the MC efficiency	Timing	0.035	
Efficiency calculation	0.071	Statistical uncertainty in the Monte Carlo computation of the efficiency			
Sample variability	0.374	Relative standard deviation of the activities obtained for the 12 sources used.			

Table A8. Uncertainty budgets for the results submitted by IRA (4P-NA-MX-00-00-HE) and LNHB (4P-LS-MX-00-00-TD / Hidex 300SL system).

IRA - UA-Ge(HP)-GR-00-00-00			LNHB - 4P-LS-MX-00-00-TD / Hidex 300SL		
Quantity	***u*(*a*)/*a* in %**	**Comments**	**Quantity**	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.1		Counting statistics	0.08	Standard deviation calculated from the measurement of 8 LS vials
Weighing	0.01		Weighing	0.1	Pycnometer technique
Background	0.025		Background	0.01	
Decay data	0.0138		Dead/live time	0.13	Hidex dead-time management verified at LNHB
Efficiency curve	1.4		Decay data	0.12	DDEP recommended values
Decay correction	0.03		Decay correction	0.05	
			Dilution	0.2	
			TDCR model	0.2	

Table A9. Uncertainty budgets for the results submitted by LNHB (4P-LS-MX-00-00-TD / RCTD1 system) and (4P-NA-MX-00-00-HE).

LNHB - 4P-LS-MX-00-00-TD / RCTD1 system			LNHB - 4P-NA-MX-00-00-HE		
Quantity	***u*(*a*)/*a* in %**	**Comments**	**Quantity**	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.14	Standard deviation calculated from the measurement of 8 LS vials	Counting statistics	0.1	Standard deviation of 8 sources
Weighing	0.1	Pycnometer technique	Weighing	0.05	Weighing with AX504 balance
Dead/live time	0.02	Live time technique using extendable dead-times	Background	0.1	Statistics
Background	0.05		Dead/live time	0.01	Live time technique using a MTR2 module
Decay data	0.12		Dilution factor	0.2	Conservative calculation
Decay correction	0.05		Zero-energy extrapolation	0.3	Conservative calculation
Dilution	0.2		Detection efficiency	0.3	Geant4 Monte Carlo simulation
TDCR model	0.2		Decay correction		Negligible

Table A10. Uncertainty budgets for the results submitted by NPL (4P-LS-MX-00-00-TD) and POLATOM (4P-LS-MX-00-00-TD).

NPL - 4P-LS-MX-00-00-TD			POLATOM - 4P-LS-MX-00-00-TD		
Quantity	***u*(*a*)/*a* in %**	**Comments**	**Quantity**	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.0126		Counting statistics	0.13	
Weighing	0.0729		Weighing	0.15	
Background	0.09		Background	0.008	
Dead/live time	0.0607		Dead/live time	0.001	
Pile-up	0.15		Resolving time	0.001	
Decay data	0.07		Decay data	0.02	
Decay correction	0.008		extra-/inter-polation of efficiency curve	0.2	
Impurities	0.0547		Decay correction	0.009	
Wall effects	0.02		Impurities	0.03	
Asymmetry	0.1		Adsorption	0.04	
Choice of Bi-213 BR	0.04		Measurement method	0.2	
Dilution factor	0.0607		Dilution	0.004	
			kB value	0.04	

Table A11. Uncertainty budgets for the result submitted by CIEMAT (2P-IC-AP-00-00-00).

The measurements were carried out using a 2π-α grid ionization chamber NUMELEC NU14B with an incorporated preamplifier connected to a spectroscopy amplifier CAEN N968 module. A non-extending dead time equal to 3.0(1) μs was applied.

CIEMAT - 2P-IC-AP-00-00-00		
Quantity	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.4	Includes standard deviation of individual results
Weighing	0.2	From calibration certificate
Background	0.05	
Dead/live time	0.0001	
Adsorption	0.05	Estimated
Self-adsorption		Estimated
Backscattering	0.1	
Dilution	0.1	
Tail extrapolation	0.1	

Table A12. Uncertainty budgets for the result submitted by IRA (UA-Ge(HP)-GR-00-00).

IRA - UA-Ge(HP)-GR-00-00		
Quantity	***u*(*a*)/*a* in %**	**Comments**
Counting statistics	0.1	
Weighing	0.01	
Background	0.025	
Decay data	0.0138	
Efficiency curve	1.4	
Decay correction	0.03	