Final report of the new BIPM comparison BIPM.RI(II)-K1.Ac-225 of activity measurements of the radionuclide ^{225}Ac to include the 2019 result of the PTB (Germany)

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Abstract In 2019, the PTB(Germany) has submitted a sample of ^{225}Ac to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM), with comparison identifier BIPM.RI(II)-K1.Ac-225. As PTB is the first participant in the comparison no key comparison reference value (KCRV) and no degrees of equivalence can be calculated.

1. Introduction

The SIR for activity measurements of γ-ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled with 3.6 g of the radioactive solution. For radioactive gases, a different standard ampoule is used. Each NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of ^{226}Ra using pressurized ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity A_e, are all given in [1].

From its inception until 31 December 2020, the SIR has been used to measure 1021 ampoules to give 776 independent results for 72 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary standardizations. These comparisons are described as BIPM continuous comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the Comité International des Poids et Mesures Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Ac-225 key comparison.

2. Participants

Laboratory details are given in Table 1. The dates of measurement in the SIR given in Table 1 are used in the KCDB and all references in this report.

Table 1: Details of the participants in the BIPM.RI(II)-K1.Ac-225.

NMI or laboratory	**Previous acronyms**	**Full name**	**Country**	**RMO**	**Date of SIR measurement** yyyy-mm-dd
PTB	-	Physikalisch-Technische Bundesanstalt	Germany	EURAMET	2019-06-21

3. NMI standardization methods

Each NMI that submits ampoules to the SIR has measured the activity either by a primary standardization method or by using a secondary method, for example a calibrated ionization chamber. In the latter case, the traceability of the calibration needs to be clearly identified to ensure that appropriate correlations are taken into account.

A brief description of the standardization methods used by the laboratories, the activities submitted, the relative standard uncertainties and the half-life used by the participants are given in Table 2. The uncertainty budget for the new submission is given in 7 attached to this report. The list of acronyms used to summarize the methods is given in 8.

The half-life used by the BIPM is 9.920(3) days as published by S. Pommé et al. [3].

Table 2: Standardization methods of the participants for ^{225}Ac.

NMI or laboratory	**Method used and the acronym**	**Activity** A_i/**kBq**	**Relative standard uncertainty** /10^{-2}		**Reference date** yyyy-mm-dd	**Half-life** /d
			A	**B**		
PTB	TDCR (4P-LS-MX-00-00-TD)[a]	10 066	0.05	0.25	2019-06-14 00:00 UT	9.920(3)

[a] See details in [4]

Details regarding the solutions submitted are shown in Table 3, including any impurities, when present, as identified by the laboratories. When given, the standard uncertainties on the evaluations are shown.

Table 3: Details of each solution of ^{225}Ac submitted.

NMI or laboratory / SIR year	**Chemical composition**	**Solvent conc.** /(mol dm^{-3})	**Carrier conc.** /(µg g^{-1})	**Density** /(g cm^{-3})	**Relative activity of any impurity**[b]
PTB 2019	^{225}Ac in HCl	0.1	-	approx. 1.0	^{225}Ra: 9.8(25)x10^{-3} %[a]

[a] Impurity not at equilibrium; correction for the SIR measurement assuming the source production date is equal to the reference date

[b] the ratio of the activity of the impurity to the activity of ^{225}Ac at the reference date

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". The activity measurements for ^{225}Ac now have a total of 1 ampoule. The SIR equivalent activity, A_{ei}, for each ampoule received from each NMI, i, including both previous and new results, is given in Table 4.

The relative standard uncertainties arising from the measurements in the SIR are also shown. This uncertainty is additional to that declared by the NMI ($u(A_i)$) for the activity measurement shown in Table 2. Although submitted activities are compared with a given source of ^{226}Ra, all the SIR results are normalized to the radium source number 5 [1]. No recent submission has been identified as a pilot study so the most recent result of each NMI is normally eligible for inclusion on the KCDB platform of the CIPM MRA [2].

A change of the assumed source production date (see Table 3) by 10 days increases by a factor of two the SIR correction of 1.00016 for the ^{225}Ra impurity which is not at equilibrium. However the effect on the SIR result is negligible. Measurements repeated at the BIPM after up to 2 half-lives later produced results in agreement within standard uncertainty.

Table 4: Results of SIR measurement of ^{225}Ac.

NMI or laboratory / SIR year	m_i /g	A_i /kBq	^{226}**Ra source**	A_{ei} /kBq	**Relative uncert. from SIR** /10^{-4}	u_{ci} /kBq	A_{ei} **for KCRV** /kBq
PTB 2019	3.608 85	10 066	3	74 519	8	200	74 520(200)

4.1. The key comparison reference value

As there is only one participant in the comparison, no key comparison reference value (KCRV) can be calculated. However, the result can be compared with the estimation of 75 840(530) kBq obtained using the SIRIC efficiency curve of the SIR [5] and nuclear data for ^{225}Ac [6] in equilibrium with ^{221}Fr, ^{213}Bi and ^{209}Tl [7], [8], and neglecting the contribution from ^{217}At, ^{213}Po and ^{209}Pb.

4.2. Degrees of equivalence

As there is only one participant in the comparison, no degrees of equivalence can be calculated.

5. Conclusion

The BIPM continuous key comparison for ^{225}Ac, BIPM.RI(II)-K1.Ac-225, comprises now 1 result. Other results may be added when other NMIs contribute ^{225}Ac activity measurements to this comparison or take part in other linked comparisons.

6. References

[1] Ratel, G. The Système International de Référence and its application in key comparisons, *Metrologia*, 2007, **44**(4), S7-S16.

[2] CIPM MRA: *Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes*, International Committee for Weights and Measures, 1999, pp. 45, Technical Supplement revised in October 2003 (pages 38-41).

[3] S. Pommé S., Marouli M., Suliman G. , Dikmen H., Van Ammel R., Jobbágy V., Dirican A., Stroh H., Paepen J., Bruchertseifer F., Apostolidis C., and Morgenstern A., Measurement of the ^{225}Ac half-life, *Applied Radiation and Isotopes*, 2012, **70**, 2608-2614.

[4] Kossert K., Takács M. P. and Nähle O., Determination of the activity of ^{225}Ac and of the half-lives of ^{213}Po and ^{225}Ac, *Applied Radiation and Isotopes*, 2020, **156**, 109020.

[5] Cox M.G., Michotte C., Pearce A.K., Measurement modelling of the International Reference System (SIR) for gamma-emitting radionuclides, 2007, Monographie BIPM-7, 48 pp.

[6] Bé M.-M., Chisté V., Dulieu C., Mougeot X., Chechev V.P., Kondev F.G., Nichols A.L., Huang X., Wang B., *Table of radionuclides*, Monographie BIPM-5, 2013, Vol. 7.

[7] Bé M.-M., Chisté V., Dulieu C., Mougeot X., Browne E., Chechev V., Kuzmenko N., Kondev F., Luca A., Galán M., Nichols A.L., Arinc A., Huang X., *Table of radionuclides*, Monographie BIPM-5, 2010, Vol. 5.

[8] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Kondev F., Luca A., Galán M., Pearce A., Huang X., *Table of radionuclides*, Monographie BIPM-5, 2008, Vol. 4.

7. Uncertainty budgets for the activity of ^{225}Ac submitted to the SIR

Detailed Uncertainty Budget

Laboratory: **PTB** Radionuclide: **Ac-225**; Ampoule number: **2019-1444**

*Uncertainty components**, in % of the activity concentration, due to

		Remarks	Evaluation type (A or B)	Relative sensitivity Factor
counting statistics	**0.036**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **A**_ _ _ _	_ _ _ _ _ _ _
weighing	**0.033**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
dead time	**0.07**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
background	**0.03**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **A**_ _ _	_ _ _ _ _ _ _
pile-up	**n.a.**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
counting time	**negligible**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _
adsorption	**0.05**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
impurities	**0.2**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
decay correction	**0.015**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
fitting and extrapolation of efficiency curve	**0.11**	combined with model_ _ _ and decay data	_ _ _ **B**_ _ _ _ _ _ _ _ _ _	_ _ _ _ _ _ _ _ _ _ _ _ _ _
dilution	**0.048**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ **B**_ _ _	_ _ _ _ _ _ _
combined uncertainty (as quadratic sum of all uncertainty components)	**0.256**	_ _ _ _ _ _ _ _ _ _ _ _ _ _	_ _ _ _ _ _ _	_ _ _ _ _ _ _

* The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, **17**, 73 and *Guide to expression of uncertainty in measurement*, http://www.bipm.org/en/publications/guides/

8. Acronyms used to identify different measurement methods

Each acronym has six components, geometry-detector (1)-radiation (1)-detector (2)-radiation (2)-mode. When a component is unknown, ?? is used and when it is not applicable 00 is used.

Geometry	**acronym**	**Detector**	**acronym**
4 π	4P	proportional counter	PC
defined solid angle	SA	press. Prop. Counter	PP
2 π	2P	liquid scintillation counting	LS
undefined solid angle	UA	NaI(Tl)	NA
		Ge(HP)	GH
		Ge(Li)	GL
		Si(Li)	SL
		CsI(Tl)	CS
		ionization chamber	IC
		grid ionization chamber	GC
		Cerenkov detector	CD
		calorimeter	CA
		solid plastic scintillator	SP
		PIPS detector	PS
		CeBr3	CB

Radiation	**acronym**	**Mode**	**acronym**
positron	PO	efficiency tracing	ET
beta particle	BP	internal gas counting	IG
Auger electron	AE	CIEMAT/NIST	CN
conversion electron	CE	sum counting	SC
mixed electrons	ME	coincidence	CO
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with efficiency tracing	CT
x-rays	XR	anti-coincidence counting with efficiency tracing	AT
photons ($x + \gamma$)	PH	triple-to-double coincidence ratio counting	TD
photons + electrons	PE	selective sampling	SS
alpha particle	AP	high efficiency	HE
mixture of various radiation	MX	digital coincidence counting	DC

Examples of methods	**acronym**
4π(PC)β-γ coincidence counting	4P-PC-BP-NA-GR-CO
4π(PPC)β-γ coincidence counting eff. trac	4P-PP-MX-NA-GR-CT
defined solid angle α-particle counting with a PIPS detector	SA-PS-AP-00-00-00
4π(PPC)AX-γ(GeHP)-anticoincidence counting	4P-PP-MX-GH-GR-AC
4πCsI-β,AX,γ counting	4P-CS-MX-00-00-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG