Update of the BIPM comparison BIPM.RI(II)-K1.Ce-139 of activity measurements of the radionuclide ^{139}Ce to include the 2019 result of the NMISA (South Africa) and the 2022 result of the LNE-LNHB (France)

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Abstract Since 1976, 13 laboratories have submitted 27 samples of ^{139}Ce 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.Ce-139. Recently, the NMISA (South Africa) and the LNE-LNHB (France) participated in the comparison and the key comparison reference value (KCRV) has been updated. The degrees of equivalence between each equivalent activity measured in the SIR and the updated KCRV have been calculated and the results are given in the form of a table. A graphical presentation is also given.

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 2021, the SIR has been used to measure 1033 ampoules to give 788 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.Ce-139 key comparison. The results of earlier participations in this key comparison were published previously [3–5].

2. Participants

Laboratory details are given in Table 1, with the earlier submissions being taken from [3–5]. 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.Ce-139.

NMI or laboratory	Previous acronyms	Full name	Country	RMO	Date of SIR measurement yyyy-mm-dd
BEV	IRK	Bundesamt fur Eich- und Vermessungswesen	Austria	EURAMET	2008-12-02
BIPM	-	Bureau International des Poids et Mesures			1976-03-19
BKFH	OMH, MKEH	Government Office of the Capital City Budapest	Hungary	EURAMET	1984-06-07
CMI	UVVVR, CMI-IIR	Czech Metrological Institute	Czechia	EURAMET	1985-03-01
IRA	IER	Institut de Radiophysique	Switzerland	EURAMET	1983-12-22 2000-12-01
KAE	-	Korea Atomic Energy Research Institute	Republic of Korea	APMP	1976-06-04
LNE-LNHB	LMRI, LPRI, BNM-LNHB	Laboratoire National de métrologie et d´Essais -Laboratoire National Henri Becquerel	France	EURAMET	1997-02-26 2022-03-16
LNMRI-IRD	IEA, IPEN[a]	Laboratorio Nacional de Metrologia das Radiações Ionizantes	Brazil	SIM	1997-10-28
NIST	NBS	National Institute of Standards and Technology	United States	SIM	1988-01-05 1997-01-24
NMIJ	ETL	National Metrology Institute of Japan	Japan	APMP	1994-12-05

... Continuation of Table 1.

NMI or laboratory	Previous acronyms	Full name	Country	RMO	Date of SIR measurement yyyy-mm-dd
					2004-03-16
NMISA	NAC, CSIR-NML[b]	National Metrology Institute of South Africa	South Africa	AFRIMETS	1980-11-14 1982-04-29 1983-12-01 1999-03-17 2019-03-07
NPL	-	National Physical Laboratory	United Kingdom	EURAMET	1981-10-07
PTB	-	Physikalisch-Technische Bundesanstalt	Germany	EURAMET	1999-12-01 2008-03-14

[a] IEA, IPEN are other institutes of the country.
[b] NAC is another institute of the country.

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 Appendix D attached to this report; previous uncertainty budgets are given in the earlier K1 reports [3–5]. The list of acronyms used to summarize the methods is given in Appendix E.

The half-life used by the BIPM is 137.65(7) days as published in AECL rep 1980 [6], which is in agreement‡ with the evaluation published in 2008 in the Monographie-5, 137.641(20) days [7]. The IAEA half-life of 137.640(23) days [8] was used in the 2004 APMP.RI(II)-K2.Ce-139 comparison.

‡ An update of the half-life involves recalculating all previously published equivalent activity values. This is done only when the update has a significant impact on the results.

Table 2: Standardization methods of the participants for ^{139}Ce.

NMI or laboratory	**Method used and the acronym**	**Activity** A_i/**MBq**	**Relative standard uncertainty** /$\mathbf{10^{-2}}$		**Reference date**	**Half-life /d**
			A	**B**	**yyyy-mm-dd**	
BEV	Ionization chamber (4P-IC-GR-00-00-00)	484	0.31	0.84	2008-12-01 00:00 UT	137.641
BIPM	4π(PC)-γ coincidence (4P-PC-00-NA-GR-CO)	2493.2[f] 2493.7	0.19 0.07	1.17 0.57	1976-03-15 00:00 UT	137.65(7)
BKFH	4π(x,e)-γ coincidence (4P-PP-MX-NA-GR-CO)	3616	0.03	0.35	1984-06-30 12:00 UT	137.64(5)
CMI	4π(x,e)-γ coincidence (4P-00-MX-NA-GR-CO)	14 820	0.05	0.23	1985-01-24 12:00 UT	137.5
IRA	4π(PC)x-γ coincidence (4P-PC-XR-NA-GR-CO)	2690	0.05	0.3	1983-11-15 00:00 UT	-
	4π x-γ coincidence (4P-00-XR-NA-GR-CO)	5224	0.54	0.23	2000-12-01 12:00 UT	137.64(2)
KAE	NaI(Tl) γ spectrometry (2P-NA-GR-00-00-00)	1687[f] 1689	0.83 0.83	0.21 0.21	1976-05-19 02:00 UT	-
LNE-LNHB	$4\pi\beta$-γ coincidence (4P-PC-BP-NA-GR-CO)	2848[f] 2807	0.50 0.50	0.02 0.02	1997-02-01 12:00 UT	137.640(23) [6]
	4π x-γ counting (4P-NA-MX-00-00-HE) $4\pi\beta$-γ coincidence method (4P-LS-MX-NA-GR-AC)	2435.4[a] 2442.8	0.141 0.315	0.321[a2] 0.102	2022-01-03 12:00 UT	137.641(20) [7]
LNMRI-IRD	4π(x,e)-γ coincidence (4P-00-MX-NA-GR-CO)	1071	0.22	0.25	1997-08-01 00:00 UT	-
NIST	Ionization chamber (4P-IC-GR-00-00-00)[b]	1850	0.01	0.3	1987-12-03 17:00 UT	137.64(2)
	Ionization chamber (4P-IC-GR-00-00-00)[b]	730.9	0.06	0.24	1997-01-01 12:00 UT	-
NMIJ	4π(x,e)-γ coincidence (4P-00-MX-NA-GR-CO)	538	0.2	0.34	1994-12-01 12:00 UT	
	4π(x,e)-γ coincidence (4P-PC-MX-NA-GR-CO)	1111.2[c]	0.17	0.07	2004-03-01 00:00 UT	137.64
NMISA	4π(LS) β-γ coincidence (4P-LS-BP-NA-GR-CO)	46 640[f] 47 250	0.19 0.19	0.44 0.44	1980-10-24 10:00 UT	-
	4π(LS) β-γ coincidence (4P-LS-BP-NA-GR-CO)	42 670	0.39	0.33	1982-04-01 10:00 UT	
	4π(LS) β-γ coincidence (4P-LS-BP-NA-GR-CO)	55 170[f] 55 350	0.18 0.18	0.42 0.42	1983-11-01 12:00 UT	
	4π(LS) (x,e)-γ coincidence (4P-LS-MX-NA-GR-CO)	13 700	0.05	0.63	1999-01-12 12:00 UT	137.64

... Continuation of Table 2.

NMI or laboratory	**Method used and the acronym**	**Activity** A_i**/MBq**	**Relative standard uncertainty /**10^{-2} **A**	**B**	**Reference date yyyy-mm-dd**	**Half-life /d**
	4π(LS) (e,X)-γ coincidence (4P-LS-MX-NA-GR-CO)	1547.58	0.05	0.52	2019-01-30 10:00 UT	137.641
NPL	Ionization chamber (4P-IC-GR-00-00-00)[d]	2402	0.15	0.4	1981-10-05 00:00 UT	-
PTB	4π(PC)EC-γ coincidence (4P-PC-MX-NA-GR-CO) and 4π(PPC)EC-γ coincidence (4P-PP-MX-NA-GR-CO)	5400[g]	0.06	0.14	1999-11-01 00:00 UT	137.66(6)
	4π(PC)EC-γ coincidence (4P-PC-MX-NA-GR-CO) and 4π(PPC)EC-γ coincidence (4P-PP-MX-NA-GR-CO)	4145[e]	0.06	0.23	2008-02-01 00:00 UT	137.66(6)

[a] The activity of 2439.1 kBq is used. It corresponds to the arithmetic mean of both results.
[a2] The relative uncertainty of 0.35% is used. It corresponds to the higher value of both results.
[b] Calibrated by 4π(PC)-γ coincidence counting in 1976
[c] The ampoule measured by the NMIJ was used to make the link of the APMP.RI(II)-K2 comparison to the SIR.
[d] Calibrated by 4π(PC)-γ coincidence counting
[e] Weighted mean result taking correlation into account for the uncertainties
[f] Several samples submitted
[g] The result is the mean of the different methods.

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 ^{139}Ce 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]
BEV 2008	$CeCl_3$ in HCl	1	$CeCl_3$:35	1	-
BIPM 1976	$CeCl_3$ in HCl	0.2	$CeCl_3$:20	-	Negligible
BKFH 1984	Ce in HCl	0.1	Ce:25	-	-
CMI 1985	$CeCl_3$ in HCl	0.08	$CeCl_3$:30	-	<0.1 %
IRA 1983	Ce^{3+} in HCl	0.1	Ce^{3+}:7.5	-	-
2000	Ce^{3+} in HCl	0.1	Ce^{3+}:20	1.000(7)	-
KAE 1976	$CeCl_3$ in HCl	0.2	$CeCl_3$:15	-	-
LNE-LNHB 1997	$CeCl_3$ in HCl	0.5	$CeCl_3$:10	1	-
2022	$CeCl_3$ in HCl	0.1	$CeCl_3$:10	1	None detected

... Continuation of Table 3.

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]
LNMRI-IRD 1997	$CeCl_3$ in HCl	1	$CeCl_3$:30	0.998	-
NIST 1988	$CeCl_3$ in HCl	0.4	Ce:16	1.005	^{54}Mn:0.0017(3) %; ^{65}Zn:0.029(3) %
1997	$CeCl_3$ in HCl	0.77	Ce^{3+}:100	1.012	-
NMIJ 1994	$CeCl_3$ in HCl	0.1	$CeCl_3$:10	1	-
2004[a]	$CeCl_2$ in HCl	0.1	$CeCl_2$:100	1.002	-
NMISA 1980	$CeCl_3$ in HCl	1	Ce^{3+}:170	1.037	-
1982	$CeCl_3$ in HCl	1	Ce^{3+}:180	1.037	-
1983	$CeCl_3$ in HCl	1	Ce^{3+}:409	1.017	-
1999	$CeCl_3$ in HCl	1	Ce^{3+}:534	1.023	-
2019	$CeCl_3$ in HCl	0.5	$CeCl_3$:10	1	None detected
NPL 1981	$CeCl_3$ in HCl	0.1	$CeCl_3$:40	1.0015	-
PTB 1999	$CeCl_3$ in HCl	0.1	$CeCl_3$:35	0.999	-
2008	$CeCl_3$ in HCl	1	$CeCl_3$:35	1.016	-

[a] Solution used in the APMP-RI(II)-K2.Ce-139 comparison

[b] The ratio of the activity of the impurity to the activity of ^{139}Ce at the reference date

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a dedicated database [9]. The latest submission has added 2 ampoules for the activity measurements for ^{139}Ce giving rise to 27 ampoules in total.

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]. Table 4 also shows the comparison results selected for the KCRV as explained in section 4.1.

Repeated measurements at the BIPM after periods of up to 3 weeks gave comparison results for NMISA(2019) and LNE-LNHB(2022) in agreement with the standard uncertainty.

Table 4: Results of SIR measurement of ^{139}Ce.

NMI or laboratory / SIR year	m_i /g	A_i /MBq	^{226}**Ra source**	A_{ei} /MBq	**Relative uncert. from SIR** /10^{-4}	u_{ci} /MBq	A_{ei} **for KCRV** /MBq
BEV 2008	3.618 4	484	1	131.6	16	1.2	-
BIPM 1976	3.509 17	2493.2	1	132.28	12	1.58	132.3(12)[g]
	3.506 87	2493.7	1	132.38	12	0.78	-
BKFH 1984	3.600 2	3616	2	132.02	11	0.48	132.02(48)
CMI 1985	3.610 9	14 820	3	132.77	8	0.34	132.77(34)
IRA 1983	3.602	2690	2	132.63	11	0.42	-
2000	3.588 7(1)	5224	1	132.93	10	0.8	132.93(80)
KAE 1976	3.507 70	1687	1	124.5	12	1.1	-
	3.512 37	1689	1	124.6	12	1.1	-
LNE-LNHB 1997	3.551 0	2848	2	132.71[a]	10	0.68	-
	3.500 1	2807	2	132.75	11	0.68	-
2022	3.717 0(6)	2435.4	1	132.53	15	0.51	132.74(51)[h]
		2442.8		132.94		0.49	-
LNMRI-IRD 1997	3.592 77	1071	1	132.69	15	0.48	132.69(48)
NIST 1988	3.707 2	1850	1	133.38	13	0.44	133.38(44)
1997	3.650 63	730.9	1	134.41	14	0.38	-
NMIJ 1994	3.610 8	538	1	134.22	17	0.57	-
2004	3.563 86	1111.2	1	132.74[b]	17	0.35	132.74(35)
NMISA 1980	0.516 35[c]	46 640	4	134.95	8	0.66	-
	0.523 13	47 250	4	134.90	8	0.66	-
1982	0.520 97[d]	42 670	4	134.2	8	0.7	-
1983	0.253 975[e]	55 170	4	133.77	8	0.62	-
	0.254 797	55 350	4	133.70	8	0.62	-
1999	3.603	13 700	3	133.1	8	0.85	-
2019	3.711 05	1547.58	1	133.81	15	0.73	133.81(73)
NPL 1981	3.705 3	2402	2	132.76	11	0.59	132.76(59)
PTB 1999	3.739 78	5400	2	132.66	10	0.24	-
2008	3.677 28	4145	2	132.61	10	0.34	132.61(34)

[a] The mean of the two Ae values is used with an averaged uncertainty as attributed to an individual entry.
[b] Result used to link the APMP comparison to the SIR
[c] Mass of solution before dilution. Mass of solution after dilution 3.601 g and 3.604 g respectively
[d] Mass of solution before dilution. Mass after dilution = 3.872 g.
[e] Mass of solution before dilution. Mass after dilution = 3.67577 g and 3.61560 g respectively.
[g] An average value and average uncertainty between all submitted samples is used for the KCDB [10].
[h] A_i = 2439.1 kBq is considered for the KCDB (see Table 2).

The APMP.RI(II)-K2.Ce-139 comparison had been held in 2004. The results were linked to the BIPM.RI(II)-K1.Ce-139 comparison through the measurement in the SIR of at least one ampoule of the APMP comparison, as explained in [4].

4.1. The key comparison reference value

In May 2013, the CCRI(II) decided to calculate the key comparison reference value (KCRV) by using the power-moderated weighted mean [11] rather than an unweighted mean, as had been the policy. This type of weighted mean is similar to a Mandel-Paule mean in that the NMIs' uncertainties may be increased until the reduced chi-squared value is one. In addition, it allows for a power α smaller than two in the weighting factor. As proposed in [11], α is taken as $2 - 3/N$ where N is the number of results selected for the KCRV. Therefore, all SIR key comparison results can be selected for the KCRV with the following provisions:

(a) only results for solutions standardized by primary techniques are accepted, with the exception of radioactive gas standards (for which results from transfer instrument measurements that are directly traceable to a primary measurement in the laboratory may be included);
(b) each NMI or other laboratory may only use one result (normally the most recent result or the mean if more than one ampoule is submitted);
(c) results more than 20 years old are included in the calculation of the KCRV but are not included in data shown in the KCDB or in the plots in this report, as they have expired;
(d) possible outliers can be identified on a mathematical basis and excluded from the KCRV using the normalized error test with a test value of 2.5 and using the modified uncertainties;
(e) results can also be excluded for technical reasons; and
(f) the CCRI(II) is always the final arbiter regarding excluding any data from the calculation of the KCRV.

The data set used for the evaluation of the KCRVs is known as the KCRV file and is a reduced data set from the SIR master-file. Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are made only by the CCRI(II) during one of its biennial meetings, or by consensus through electronic means (e.g., email) as discussed at the CCRI(II) meeting in 2013.

Consequently, using the recent result produces an updated KCRV for ^{139}Ce in 2022 of **132.77(14) MBq** with the power α =1.727 that has been calculated using the previously published results, selected as shown in Table 4, for the BIPM (1976), NPL (1981), BKFH (1984), CMI (1985), NIST (1988), LNMRI-IRD (1997), IRA (2000), NMIJ (2004), PTB (2008), NMISA (2019), and the LNE-LNHB (2022) result. This can be compared with the previous KCRV values of 132.87(17) MBq published in 2003 [3] , 132.74(11) MBq published in 2005 [4] and 132.73(11) MBq published in 2011 [5].

4.2. Degrees of equivalence

Every participant in a comparison is entitled to have one result included in the KCDB as long as the NMI is a signatory or designated institute listed in the CIPM MRA and the result is valid (i.e., not older than 20 years). 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]. An NMI may withdraw its result only if all other participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the KCRV [2]. The degree of equivalence is expressed quantitatively in terms of the deviation from the key comparison reference value and the expanded uncertainty of this deviation ($k = 2$). The degree of equivalence between any pair of national measurement standards is expressed in terms of their difference and the expanded uncertainty of this difference and is independent of the choice of key comparison reference value.

4.2.1. Comparison of a given NMI result with the KCRV

The degree of equivalence of the result of a particular NMI, i, with the key comparison reference value is expressed as the difference D_i between the values

$$D_i = A_{\mathrm{e}i} - \mathrm{KCRV} \tag{1}$$

and the expanded uncertainty ($k = 2$) of this difference, U_i, known as the equivalence uncertainty; hence

$$U_i = 2u(D_i) \tag{2}$$

When the result of the NMI i is included in the KCRV with a weight w_i, then

$$u^2(D_i) = (1 - 2w_i)u_i^2 + u^2(\mathrm{KCRV}) \tag{3}$$

However, when the result of the NMI i is not included in the KCRV, then

$$u^2(D_i) = u_i^2 + u^2(\mathrm{KCRV}) \tag{4}$$

4.2.2. Comparison between pairs of NMI results

The degree of equivalence between the results of any pair of NMIs, i and j, is expressed as the difference D_{ij} in the values

$$D_{ij} = D_i - D_j = A_{\mathrm{e}i} - A_{\mathrm{e}j} \tag{5}$$

and the expanded uncertainty ($k = 2$) of this difference, $U_{ij} = 2u(D_{ij})$, where

$$u^2(D_{ij}) = u_i^2 + u_j^2 - 2u(A_{\mathrm{e}i}, A_{\mathrm{e}j}) \tag{6}$$

where any obvious correlations between the NMIs (such as a traceable calibration, or correlations normally coming from the SIR or from the linking factor in the case of linked comparison) are subtracted using the covariance $u(A_{\mathrm{e}i}, A_{\mathrm{e}j})$ (see [12] for more detail). However, the CCRI decided in 2011 that these pair-wise degrees of equivalence no longer need to be published as long as the methodology is explained.

Tables B1 show the matrix of all the degrees of equivalence as they will appear in the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with $A_{\mathrm{e}i}$ replaced by x_i. The introductory text is that agreed for the comparison. The graph of the results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV (identified as x_{R} in the KCDB), is shown in Figure C1. This graphical representation indicates in part the degree of equivalence between the NMIs but obviously does not take into account the correlations between the different NMIs. It should be noted that the final data in this paper, while correct at the time of publication, will become out-of-date as NMIs make new comparisons. The formal results under the CIPM MRA [2] are those available in the KCDB.

5. Conclusion

The BIPM continuous key comparison for ^{139}Ce, BIPM.RI(II)-K1.Ce-139, currently comprises 9 results. The KCRV has been recalculated to include the result from the LNE-LNHB (France) , and the NMISA (South Africa). The SIR results, together with the previously published APMP.RI(II)-K2.Ce-139 results, have been analyzed with respect to the updated KCRV, providing degrees of equivalence for 9 national metrology institutes. Other results may be added when other NMIs contribute ^{139}Ce activity measurements to this comparison or take part in other linked comparisons.

6. References

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Appendix A. Introductory text for ^{139}Ce degrees of equivalence

Key comparison BIPM.RI(II)-K1.Ce-139

MEASURAND: Equivalent activity of ^{139}Ce

Key comparison reference value: the SIR reference value x_{R} for this radionuclide is 132.77 MBq , with a standard uncertainty, u_{R} equal to 0.14 MBq (see Section 4.1 of the Final Report). The value x_i is taken as the equivalent activity for a laboratory i.

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_{\mathrm{R}})$ and U_i, its expanded uncertainty ($k = 2$), both expressed in MBq, and $U_i = 2((1 - 2w_i)u_i^2 + u_{\mathrm{R}}^2)^{1/2}$, where w_i is the weight of laboratory i contributing to the calculation of x_{R}.

Appendix B. Table of degrees of equivalence for BIPM.RI(II)-K1.Ce-139

Table B1: The table of degrees of equivalence for BIPM.RI(II)-K1.Ce-139

NMI i	D_i **/MBq**	U_i **/MBq**
NMIJ	-0.03	0.65
PTB	-0.16	0.63
BEV	-1.2	2.4
NMISA	1.0	1.4
LNE-LNHB	-0.03	0.98

Table B2: The table of degrees of equivalence for the APMP.RI(II)-K2.Ce-139(2004) comparison

NMI i	D_i **/MBq**	U_i **/MBq**
INER	0.1	1.0
KRISS	-0.9	1.1
NIM	2.0	1.5
VNIIM	0.29	0.63

Appendix C. Graph of degrees of equivalence with the KCRV for ^{139}Ce (as it appears in Appendix B of the MRA)

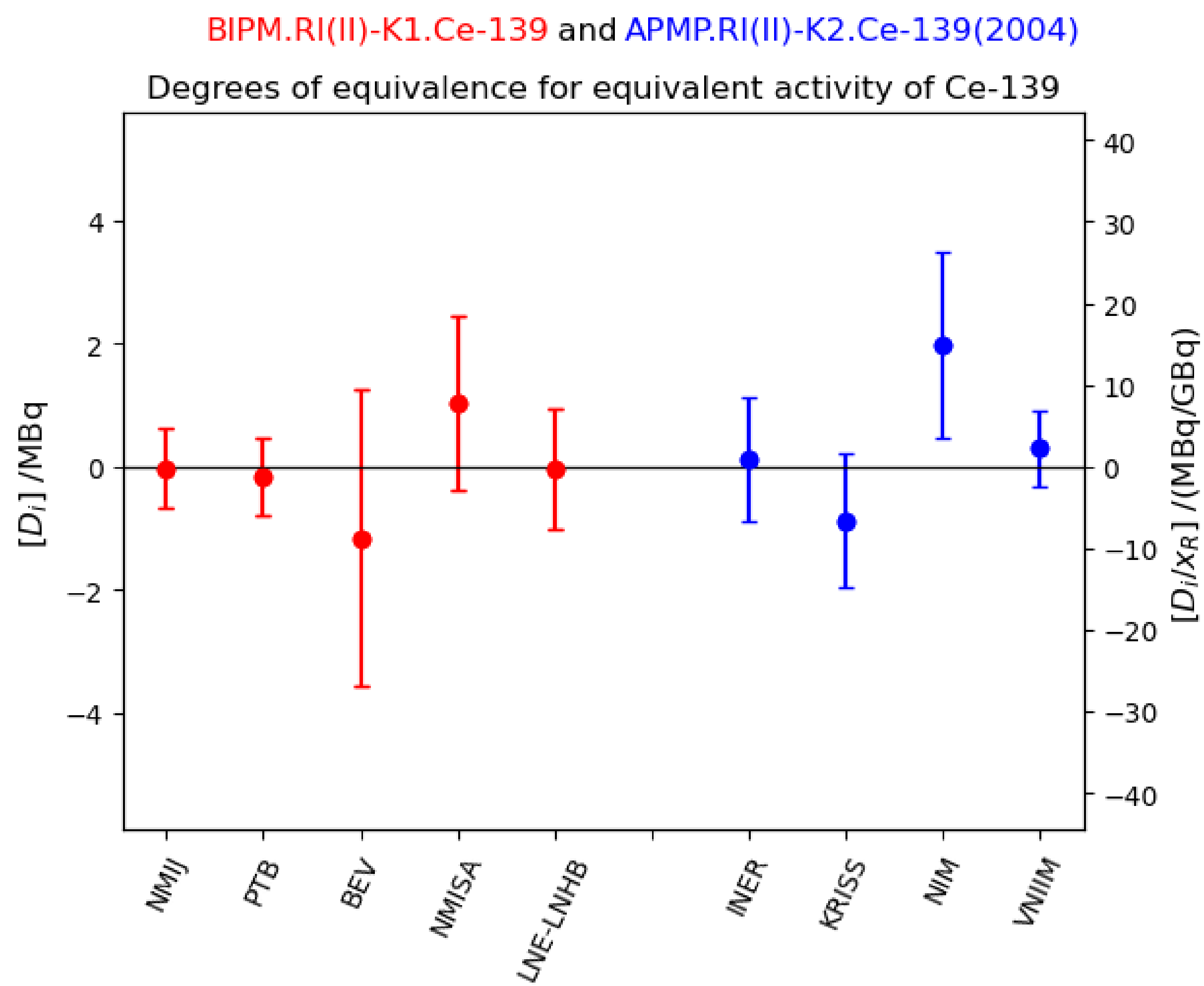

Figure C1. Degrees of equivalence for equivalent activity of ^{139}Ce.

Appendix D. Uncertainty budgets for the activity of ^{139}Ce submitted to the SIR

LNE-LNHB 4pi gamma counting

Uncertainty budget

Uncertainty component	Relative uncertainty / 10^{-2}	Evaluation type (A or B)	Comment
Counting statistics	0.100	A	Standard deviation of 11 sources
Background	0.100	A	Statistics
Weighing	0.050	B	Pycnometer technique
Dilution			
Dead time	0.010	B	Live time technique
Resolving time			
Pile-up, afterpulse			
Adsorption			
Impurities			
Decay correction	0.015	B	DDEP nuclear decay data
Zero-energy extrapolation	0.100	B	Conservative calculation
Detection efficiency	0.300	B	Monte Carlo calculation
Quenching, kB value			
Tracer			
Reproducibility			
Combined standard uncertainty	**0.350**		Quadratic sum of all uncertainty component

LNE-LNHB 4pi beta-gamma coincidence method

Uncertainty budget

Uncertainty component	Relative uncertainty / 10^{-2}	Evaluation type (A or B)	Comment
Counting statistics	0.120	A	Measurements of 10 LS sources
Background	0.080	A	
Weighing	0.100	B	pycnometer technique
Dilution			
Dead time	0.020	B	Live-time technique with MTR2 module
Resolving time			
Pile-up, afterpulse			
Adsorption			
Impurities			
Decay correction	0.010	B	
Decay data			
Extra-/Inter-polation of efficiency curve	0.280	A	Efficiency extrapolation technique carried out by PMT defocusing and grey filters
Quenching, kB value			
Tracer			
Reproducibility			
Combined standard uncertainty	**0.330**		

Appendix 1. Uncertainty budgets for the activity of ^{139}Ce submitted to the SIR

The NMISA has submitted a detailed uncertainty budget as follows:

	Relative standard uncertainties			
Contributions due to	$u_{rel,i}$ / 10^{-2}	**Method**	***Comment***	***Relative sensitivity factors***
counting statistics	0.05	A	Standard deviation of the mean of 18 values	0.24
weighing (primary source prep.) weighing (SIR ampoule prep.)	0.03 0.01	B B		1 1
dead time	0.03	B	$\Delta \tau_D \pm 0.05$ μs	0.006
background	0.01	B	Background square root statistics applied	0.003
counting time	0.001	B	Calibration of timer	1
decay correction	0.002	B	Half-life of 137.641(20) d [7]	0.11
efficiency extrapolation	0.42	B	Relative difference between 3rd O polynomial (with 2nd O coefficient = 0) and 2nd O polynomial	1
coincidence resolving time	0.03	B	$\Delta \tau_R \pm 0.01$ μs	0.01
afterpulse correction	0.3	B	Based on formula for $\Delta\theta$	0.0009
adsorption	0.01	B	Count rates after multiple rinsings relative to the expected count rates if rinsings were not done	1
radionuclide impurities	0.0	B	HPGe measurements	–
Relative combined standard uncertainty, u_c	**0.52**			

Appendix E. 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