Update of the BIPM comparison BIPM.RI(II)-K1.Ce-139 of activity measurements of the radionuclide ^{139}Ce to include the 2004 NMIJ result and links for the 2004 regional comparison APMP.RI(II)-K2.Ce-139

G. Ratel*, C. Michotte*, Y. Hino**
*BIPM and **NMIJ, Japan

Abstract

Since 1976, ten national metrology institutes (NMIs) and another laboratory have submitted twenty-one samples of known activity of ^{139}Ce to the International Reference System (SIR) for activity comparison in the BIPM.RI(II)-K1.Ce-139 comparison at the Bureau International des Poids et Mesures. The activities ranged from about 0.5 MBq to 55 MBq. The key comparison reference value (KCRV) has been recalculated to include the new National Metrology Institute of Japan (NMIJ) result. The degrees of equivalence between each equivalent activity measured in the SIR and the new KCRV have been calculated and the results are given in the form of a matrix for nine NMIs and the BIPM. A graphical presentation is also given. The results of an APMP.RI(II)-K2.Ce-139 comparison held in 2004 have been linked to the SIR results through the submission to the SIR of an ampoule by the pilot laboratory, the NMIJ. This has enabled one NMI to update its 1999 SIR entry and one NMI to update their previously published 1976 CCRI(II) comparison results and three other NMIs to have degrees of equivalence for activity measurements for this radionuclide.

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 (3.6 g) with the radionuclide in liquid (or gaseous) form. The 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].

Since its inception until 31 December 2004, the SIR has measured 872 ampoules to give 634 independent results for 62 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 realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the CIPM key comparison database (KCDB) of the Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Ce-139 key comparison. Although the details of the earlier submissions have already been published in the KCDB [3], the KCRV has been updated by the inclusion of the latest result from the NMIJ and consequently all the previously reported data are reproduced in this report for transparency of the calculation.

In addition, a regional key comparison was held in 2004 for this radionuclide, APMP.RI(II)-K2.Ce-139, piloted by the NMIJ. Nine laboratories from three RMOs took part in this comparison, including the NMIJ. Five NMIs made primary standardizations and are eligible to be linked to the BIPM key comparison as listed in Table 1b. Three results are new entries in the KCDB, one NMI has updated their previous SIR result and one has used this regional comparison to update their previously published 1976 CCRI(II)-K2.Ce-139 comparison results [3]. Three other laboratories, the Bhabha Atomic Research Centre (BARC), India, the Office of Atoms for Peace (OAP), Thailand and the Pusat Penelitian & Pengembangan Keselamatan Radiasi & Biomedika Nuklir (P3KRBiN), Indonesia used the comparison results to calibrate their secondary standard instruments for this radionuclide.

2. Participants

Ten NMIs, one other laboratory and the BIPM have submitted twenty-one ampoules for the comparison of ^{139}Ce activity measurements since 1976, the most recent submission being an ampoule standardized by the NMIJ in 2004. The laboratory details are given in Table 1a. In cases where the laboratory has changed its name since the original submission, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB.

Table 1a. Details of the participants in the BIPM.RI(II)-K1.Ce-139

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
BIPM	–	Bureau International des Poids et Mesures	–	–	1976-03-19
KAE	–	Korea Atomic Energy Research Institute	Republic of Korea	APMP	1976-06-04

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Table 1a continued. Details of the participants in the BIPM.RI(II)-K1.Ce-139

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
NAC*	CSIR-NML	National Metrology Laboratory	South Africa	SADCMET	1980-11-14 1982-04-29 1983-12-01 1999-03-17
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1981-10-07
IER	IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	1983-12-22 2000-12-01
–	OMH	Országos Mérésügyi Hivatal	Hungary	EUROMET	1984-06-07
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1985-03-01
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1988-01-05 1997-01-24
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1994-12-05 2004-03-16
LPRI	LNE**-LNHB	Laboratoire national de métrologie et d'essais -Laboratoire national Henri Becquerel	France	EUROMET	1997-02-26
–	LNMRI	Laboratorio Nacional de Metrologia das Radiaçoes Ionizantes	Brazil	SIM	1997-10-28
–	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1999-12-01

*another laboratory in the country
** note change of affiliation in 2005.

The six NMIs that took part in the regional comparison, APMP.RI(II)-K2.Ce-139 in 2004 are shown in Table 1b.

Table 1b. Details of the participants in the 2004 APMP.RI(II)-K2.Ce-139

NMI	**Full name**	**Country**	**Regional metrology organization**
CSIR-NML	National Metrology Laboratory	South Africa	SADCMET
INER	Institute of Nuclear Energy Research	Chinese Taipei	APMP
KRISS	Korea Research Institute of Standards and Science	Republic of Korea	APMP
NIM	National Institute of Metrology	China	APMP
NMIJ	National Metrology Institute of Japan	Japan	APMP
VNIIM	D.I. Mendeleyev Institute for Metrology	Russian Federation	COOMET

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 any correlations are taken into account.

A brief description of the standardization methods for each laboratory, the activities submitted and the relative standard uncertainties (k = 1) are given in Table 2. Details concerning the standardization methods and the uncertainty budgets used in the regional comparison are given in [4]. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. The SIR uncertainty budgets for the previous participants, the IRA and the CSIR-NML, are in [3] while that for the NMIJ is given in Appendix 1 attached to this report.

The half-life used by the BIPM is 137.65 (7) days [5], which is in agreement with the value recommended by the IAEA, 137.640 (23) d [6]. The IAEA half life was used in the 2004 APMP.RI(II)-K2.Ce-139 comparison.

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

NMI	Method used and acronym (see Appendix 2	Half-life /d	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation A	B
BIPM*	4π(PC) - γ coincidence 4P-PC-00-NA-GR-CO	137.65 (7)	2 493.2 2 493.7	1976-03-15 0 h UT	0.19 0.07	1.17 0.57
KAE	NaI(Tl) γ spectrometry 2P-NA-GR-00-00-00	–	1 687 1 689	1976-05-19 2 h UT	0.83	0.21
CSIR-NML	4π(LS)β-γ coincidence 4P-LS-BP-NA-GR-CO	–	46 640 47 250	1980-10-24 10 h UT	0.19	0.44
			42 670	1982-04-01 10 h UT	0.39	0.33
			55 170 55 350	1983-11-01 12 h UT	0.18	0.42
	4π(LS)(x,e)-γ coincidence 4P-LS-MX-NA-GR-CO	137.64	13 700	1999-01-12 12 h UT	0.05	0.63
NPL	Pressurized IC † 4P-IC-GR-00-00-00	–	2 402	1981-10-05 0 h UT	0.15	0.40
IRA	4π(PC)x_e- γ coincidence 4P-PC-XR-NA-GR-CO	–	2 690	1983-11-15 0 h UT	0.05	0.30
	4πx_e- γ coincidence 4P-00-XR-NA-GR-CO	137.64 (2)	5 224	2000-12-01 12 h UT	0.54	0.23
OMH	4π(x,e_x)- γ coincidence 4P-PP-MX-NA-GR-CO	137.64 (5)	3 616	1984-06-30 12 h UT	0.03	0.35
CMI-IIR	4π(x,e_x)- γ coincidence 4P-00-MX-NA-GR-CO	137.5	14 820	1985-01-24 12 h UT	0.05	0.23
NIST	Pressurized IC †† 4P-IC-GR-00-00-00	137.64 (2)	1 850	1987-12-03 17 h UT	0.01	0.30
		–	730.9	1997-01-01 12 h UT	0.06	0.24
NMIJ	4π(x,e_x)- γ coincidence 4P-00-MX-NA-GR-CO	–	538.0	1994-12-01 12 h UT	0.20	0.34
	4π(x,e)- γ coincidence 4P-PC-MX-NA-GR-CO	137.64	1 111.2 **	2004-03-01 0 h UT	0.17	0.07
LNE-LNHB	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	137.640 (23) d [5]	2 848 2 807	1997-02-01 12 h UT	0.50	0.02

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

NMI	Method used and acronym (see Appendix 3)	Half-life /d	Activity / kBq	Reference date	Relative standard uncertainty × 100 by method of evaluation	
				YY-MM-DD	A	B
LNMRI	4π(x,e_x)- γ coincidence 4P-00-MX-NA-GR-CO	–	1 071	1997-08-01 0 h UT	0.22	0.25
PTB	4π(PC and PPC)EC-γ coincidence 4P-PC-MX-NA-GR-CO 4P-PP-MX-NA-GR-CO	137.66 (6)	5 400	1999-11-01 0 h UT	0.06	0.14

* the two ampoules measured by the BIPM for the CCRI(II)-K2.Ce-139 and measured in the SIR were used to make the link for the CCRI(II) key comparison
** the ampoule measured by the NMIJ for the APMP.RI(II)-K2.Ce-139 and measured in the SIR was used to make the link for the APMP key comparison
† calibrated by 4π(PC)- γ coincidence counting for the nuclide considered
†† calibrated by 4π(PC)- γ coincidence counting for the nuclide considered in 1976.

Details regarding the solution 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. Recently the BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [7]. The CCRI(II) agreed in 1999 [8] that this method should be followed according to the protocol described in [9] when an NMI makes such a request or when there appear to be discrepancies. No impurity measurement was carried out at the BIPM for ^{139}Ce.

Table 3. Details of the solution of ^{139}Ce submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity†
BIPM	$CeCl_3$ in HCl*	0.2	$CeCl_3$:20	–	Negligible [3]
KAE	$CeCl_3$ in HCl	0.2	$CeCl_3$: 15	–	–
CSIR-NML	$CeCl_3$ in HCl	1	Ce^{3+} : 170	1.037	–
			Ce^{3+} : 180	1.037	–
			Ce^{3+} : 409	1.017	–
			Ce^{3+} : 534	1.023	–

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Table 3 continued. Details of the solution of ^{139}Ce submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity†
NPL	$CeCl_3$ in HCl	0.1	$CeCl_3$: 40	1.0015	–
IRA	Ce^{+3} in HCl	0.1	Ce^{+3} : 7.5	–	–
			Ce^{+3} : 20	1.000 (7)	–
OMH	Ce in HCl	0.1	Ce : 25	–	–
CMI-IIR	$CeCl_3$ in HCl	0.08	$CeCl_3$: 30	–	< 0.1 %
NIST	$CeCl_3$ in HCl	0.4	Ce : 16	1.005	^{54}Mn: 0.0017 (3) % ^{65}Zn: 0.029 (3) %
		0.77	Ce^{+3} : 100	1.012	–
NMIJ	$CeCl_3$ in HCl	0.1	$CeCl_3$: 10	1.00	–
	$CeCl_2$ in HCl**	0.1	$CeCl_2$: 100	1.002	–
LNE-LNHB	$CeCl_3$ in HCl	0.5	$CeCl_3$: 10	1	–
LNMRI	$CeCl_3$ in HCl	1	$CeCl_3$: 30	0.998	–
PTB	$CeCl_3$ in HCl	0.1	$CeCl_3$: 35	0.999	–

† the ratio of the activity of the impurity to the activity of ^{139}Ce at the reference date.
* solution used in the CCRI(II)-K2.Ce-139 comparison
** solution used in the APMP.RI(II)-K2.Ce-139 comparison

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The activity measurements for ^{139}Ce arise from twenty-one ampoules and the SIR equivalent activity for each ampoule, A_{ei}, is given in Table 4a for each NMI, i. The dates of measurement in the SIR are given in Table 1.

The relative standard uncertainties arising from the measurements in the SIR are also shown in Table 4a. This uncertainty is additional to that declared by the NMI for the activity measurement shown in Table 2. Although activities submitted are compared with a given source of ^{226}Ra, all the SIR results are normalized to the radium source number 5 [1].

Measurements repeated at the BIPM after periods of up to 5 months later produced the same comparison results for the KAE, CSIR-NML (1982), OMH, CMI-IIR and for the NMIJ (2004). These measurements confirm the validity of the half-life value used and exclude the possibility of a non-corrected impurity for the low KAE result.

Table 4a. Results of SIR measurements of ^{139}Ce

NMI	Mass of solution m_i / g	Activity submitted A_i / kBq	N° of Ra source used	SIR A_e / MBq	Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / MBq
BIPM [a]	3.509 17 3.506 87	2 493.2 2 493.7	1	132.28 132.38*	12×10^{-4}	1.58 0.78
KAE	3.507 70 3.512 37	1 687 1 689	1	124.5 124.6	12×10^{-4}	1.1 1.1
CSIR-NML	0.516 35 [b] 0.523 13	46 640 47 250	4	134.95 134.90	8×10^{-4}	0.66 0.66
	0.520 97 [c]	42 670	4	134.20	8×10^{-4}	0.70
	0.253 975 [d] 0.254 797	55 170 55 350	4	133.77 133.70	8×10^{-4}	0.62 0.62
	3.603	13 700	3	133.10 [e]	8×10^{-4}	0.85
NPL	3.705 3	2 402	2	132.76	11×10^{-4}	0.59
IRA	3.602 0	2 690	2	132.63	11×10^{-4}	0.42
	3.588 7 (1)	5 224	1	132.93	10×10^{-4}	0.80
OMH	3.600 2	3 616	2	132.02	11×10^{-4}	0.48
CMI-IIR	3.610 9	14 820	3	132.77	8×10^{-4}	0.34
NIST	3.707 2	1 850	1	133.38	13×10^{-4}	0.44
	3.650 63	730.9	1	134.41	14×10^{-4}	0.38
NMIJ	3.610 8	538.0	1	134.22	17×10^{-4}	0.57
	3.563 86	1 111.2	1	132.74**	17×10^{-4}	0.35
LNE-LNHB [a]	3.551 0 3.500 1	2 848 2 807	2	132.71 132.75	10×10^{-4} 11×10^{-4}	0.68 0.68
LNMRI	3.592 77	1 071	1	132.69	15×10^{-4}	0.48
PTB	3.739 78	5 400	2	132.66	10×10^{-4}	0.24

[a] the mean of the two A_e values is used with an averaged uncertainty, as attributed to an individual entry [10]
[b] mass of solution before dilution. Mass after dilution = 3.601 g and 3.604 g respectively
[c] mass of solution before dilution. Mass after dilution = 3.872 g
[d] mass of solution before dilution. Mass after dilution = 3.675 77 g and 3.615 60 g respectively
[e] value superseded in the KCDB by the APMP comparison result
* results used to link the CCRI(II) comparison to the SIR.
** result used to link the APMP comparison to the SIR.

Apart from one earlier result that was withdrawn from the SIR, no recent submission has been identified as a pilot study so the results of each NMI are eligible for Appendix B of the MRA. However, the KAE is neither an NMI nor a designated institute of the Republic of Korea and consequently their result is not eligible for Appendix B. The result for the CSIR-NML has been updated by their participation in the APMP.RI(II)-K2.Ce-139 comparison.

The results of the regional comparison have been published [4]. The five laboratories to be added to the matrix of degrees of equivalence from this publication are those given in Table 1b. The results $(A/m)_i$ for these laboratories are linked to the SIR through the measurement in the SIR of an ampoule of the same solution standardized by the NMIJ. The link is made using a normalization ratio deduced from the line indicated in Table 4a:

$$A_{ei} = (A/m)_i \times (A_{e,\mathrm{NMIJ}} / (A/m)_{\mathrm{NMIJ}}) = (A/m)_i \times 425.72 \qquad (1)$$

The details of the links are given in Table 4b. The uncertainties for the regional comparison linked to the SIR are comprised of the original uncertainties together with the uncertainty in the link, 17×10^{-4}, given by the uncertainty of the SIR measurement of the NMIJ ampoule.

The three NMIs that used the comparison results to calibrate their ionization chambers and consequently cannot be linked to the SIR through this comparison, need to make subsequent bi-lateral comparisons for this radionuclide or to send a submission directly to the SIR.

4.1 The key comparison reference value

The key comparison reference value is derived from the unweighted mean of all the results submitted to the SIR with the following provisions:

a) only primary standardized solutions are accepted, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;
b) each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);
c) any outliers are identified using a reduced chi-squared test and, if necessary, excluded from the KCRV using the normalized error test with a test value of four;
d) exclusions must be approved by the CCRI(II).

The reduced data set used for the evaluation of the KCRVs is known as the KCRV file and is the reduced data set from the SIR mother-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 only made by the CCRI(II), normally during one of its biennial meetings. In this case, the new result of the NMIJ has been used in preference to their earlier result with the endorsement of the CCRI(II). Consequently, the KCRV for ^{139}Ce has been calculated as 132.74 (11) MBq using the results from the BIPM, NPL, OMH, CMI-IIR, NIST(1988), LNE-LNHB, LNMRI, CSIR-NML(1999), PTB, IRA(2000) and the NMIJ(2004) which has the effect of reducing the value of the KCRV by 10^{-3} in relative terms and the uncertainty on the KCRV from 170 kBq to 110 kBq.

Table 4b. Results of 2004 regional comparison primary measurements of ^{139}Ce and links to the SIR

NMI	Measurement method and acronym (see Appendix 2)	Activity * concentration measured $(A/m)_i$ / (Bq mg^{-1})	Evaluation by category of relative standard uncertainty × 100		Equivalent SIR activity A_{ei} / kBq	Combined standard uncertainty $u_{c\,i}$ / kBq
			A	B		
CSIR-NML	4π(LS)(x,e)-γ 4P-LS-MX-NA-GR-CO	312.90	0.09	0.52	133 210	740
INER	4π(PC)(x,e)-γ 4P-PC-MX-NA-GR-CO	312.12	0.10	0.30	132 880	480
KRISS	4π(PP)(x,e)-γ 4P-PP-MX-NA-GR-CO	309.80	0.23	0.27	131 890	530
NIM	4π(PC)(x,e)-γ 4P-PC-MX-NA-GR-CO	316.50	0.25	0.45	134 740	740
NMIJ	4π(x,e_x)- γ coincidence 4P-PC-MX-NA-GR-CO	311.80	0.17	0.07	132 740	350
VNIIM	4π(PC)(x,e)-γ 4P-PC-MX-NA-GR-CO	312.54	0.06	0.10	133060	280

*referenced to 2004-03-01 0 h UT

4.2 Degrees of equivalence

Every NMI that has submitted ampoules to the SIR is entitled to have one result included in Appendix B of the KCDB as long as the NMI is a signatory or designated institute listed in the MRA. Normally, the most recent result is the one included. Any NMI may withdraw its result only if all the participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the key comparison reference value [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 with the KCRV*

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

$$D_i = A_{ei} - \text{KCRV} \qquad (2)$$

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

$$U_i = 2u_{D_i}, \qquad (3)$$

taking correlations into account as appropriate [11].

4.2.2 *Comparison of any two NMIs with each other*

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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \qquad (4)$$

and the expanded uncertainty of this difference U_{ij} where

$$u_{D_{ij}}{}^2 = u_i{}^2 + u_j{}^2 - 2u(A_{e,i}, A_{e,j}) \qquad (5)$$

where any obvious correlations between the NMIs (such as a traceable calibration) are subtracted using the covariance $u(A_{ei}, A_{ej})$, as are normally those correlations coming from the SIR.

The uncertainties of the differences between the values assigned by individual NMIs and the key comparison reference value (KCRV) are not necessarily the same uncertainties that enter into the calculation of the uncertainties in the degrees of equivalence between a pair of participants. Consequently, the uncertainties in the table of degrees of equivalence cannot be generated from the column in the table that gives the uncertainty of each participant with respect to the KCRV. However, the effects of correlations have been treated in a simplified way as the degree of confidence in the uncertainties themselves does not warrant a more rigorous approach.

Table 5 shows the matrix of all the degrees of equivalence as they will appear in Appendix B of the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{ei} replaced by x_i. The introductory text is that agreed for the comparison. The graph of the first column of results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV, is shown in Figure 1 where, following the advice of the CCRI, measurements made prior to 1983 are indicated as black squares. This graphical representation indicates in part the degree of equivalence between the NMIs but does not take into account the correlations between the different NMIs. However, the matrix of degrees of equivalence shown in yellow in Table 5 does take the known correlations into account.

The results of the 1976 CCRI(II)-K2.Ce-139 international comparison have already been linked to those of the SIR through the measurement in the SIR of the BIPM ampoules of the comparison [3]. For completeness, the degrees of equivalence to the presently updated KCRV are given as the extension of the matrix in Table 5 and as the second set of values in Figure 1. The degrees of equivalence between all pairs of NMIs are also given in Table 5. The correlations associated with the distribution of the same solution in the international comparison have been ignored in the analysis as the overall uncertainties are quite large. The correlation coming from the link to the SIR has been taken into account.

The results of the 2004 APMP.RI(II)-K2.Ce-139 regional comparison, linked through the SIR measurement of the NMIJ, are given as the third extension of the matrix in Table 5 and as the third set of values in Figure 1. The correlations associated with the distribution of the same solution in the regional comparison have been ignored in the analysis as the overall uncertainties are quite large. The correlation coming from the link to the SIR through the NMIJ has been taken into account.

Conclusion

The BIPM ongoing key comparison for ^{139}Ce, BIPM.RI(II)-K1.Ce-139 currently comprises ten results as that of the CSIR-NML has been superseded by the regional comparison in which they took part . The results have been analysed with respect to the new KCRV determined for this radionuclide, and with respect to each other. The matrix of degrees of equivalence has been approved by the CCRI(II) and is published in the BIPM key comparison database.

The results of five other NMIs and one international laboratory that took part in the CCRI(II)-K2.Ce-139 comparison in 1976 were linked in 2003 to the BIPM ongoing key comparison through two ampoules of the comparison measured in the SIR. These linked results are included in the matrix of degrees of equivalence approved by the CCRI(II).

The results of five other NMIs that took part in the APMP.RI(II)-K2.Ce-139 comparison in 2004 have been linked to the BIPM ongoing comparison through an ampoule of the comparison solution standardized by the NMIJ and measured in the SIR. These linked results are included in the matrix of degrees of equivalence approved by the CCRI(II) and have enabled one NMI to update its 1999 SIR result and one NMI to update their 1976 CCRI results.

Other results may be added as and when other NMIs contribute ^{139}Ce activity measurements to this comparison or take part in other linked comparisons.

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Table 5. Table of degrees of equivalence and introductory text for ^{139}Ce

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

MEASURAND : **Equivalent activity of ^{139}Ce**

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 132.74 MBq
with a standard uncertainty, u_R = 0.11 MBq (see Section 4.1 of the Final Report).
The value x_i is the equivalent activity for laboratory *i*.

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms:
$D_i = (x_i - x_R)$ and U_i, its expanded uncertainty (k = 2), both expressed in MBq, and
$U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\Sigma u_i^2)^{1/2}$ when each laboratory has contributed to the calculation of x_R, with n the number of laboratories.

The degree of equivalence between two laboratories is given by a pair of terms:
$D_{ij} = D_i - D_j = (x_i - x_j)$ and U_{ij}, its expanded uncertainty (k = 2), both expressed in MBq.

The approximation $U_{ij} \sim 2(u_i^2 + u_j^2)^{1/2}$ is used in the following table.

Linking CCRI(II)-K2.Ce-139 (1976) to BIPM.RI(II)-K1.Ce-139

The value x_i is the equivalent activity for laboratory *i* participant in CCRI(II)-K2.Ce-139
having been normalized to the value of the BIPM as the linking laboratory (see Final report).

The degree of equivalence of laboratory *i* participant in CCRI(II)-K2.Ce-139 with respect to the key comparison reference value is given
by a pair of terms: $D_i = (x_i - x_R)$ and U_i, its expanded uncertainty (k = 2), both expressed in MBq.
The approximation $U_i = 2(u_i^2 + u_R^2)^{1/2}$ is used in the following table.

The degree of equivalence between two laboratories *i* and *j*, one participant in BIPM.RI(II)-K1.Ce-139 and one in CCRI(II)-K2.Ce-139,
or both participant in CCRI(II)-K2.Ce-139, is given by a pair of terms: $D_{ij} = D_i - D_j$ and U_{ij}, its expanded uncertainty (k = 2),
both expressed in MBq, where the approximation $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_l^2)^{1/2}$ is used where *l* is the linking laboratory
when both laboratories are linked and *f* is the correlation coefficient.

Linking APMP.RI(II)-K2.Ce-139 (2004) to BIPM.RI(II)-K1.Ce-139

The value x_i is the equivalent activity for laboratory *i* participant in APMP.RI(II)-K2.Ce-139
having been normalized to the value of the NMIJ as the linking laboratory (see Final report).

The degree of equivalence of laboratory *i* participant in APMP.RI(II)-K2.Ce-139 with respect to the key comparison reference value is given
by a pair of terms: $D_i = (x_i - x_R)$ and U_i, its expanded uncertainty (k = 2), both expressed in MBq.
The approximation $U_i = 2(u_i^2 + u_R^2)^{1/2}$ is used in the following table.

The degree of equivalence between two laboratories *i* and *j*, is given by a pair of terms: $D_{ij} = D_i - D_j$
and U_{ij}, its expanded uncertainty (k = 2), both expressed in MBq, where the approximation $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_l^2)^{1/2}$ is used where *l* is
the appropriate linking laboratory when both laboratories are linked and *f* is the correlation coefficient.

These statements make it possible to extend the BIPM.RI(II)-K1.Ce-139 matrices of equivalence to the other participants in CCRI(II)-K2.Ce-139 and
in APMP.RI(II)-K2.Ce-139

Table 5 continued. Degrees of equivalence for ^{139}Ce

Lab *j* ⇒

Lab *i* ⇓

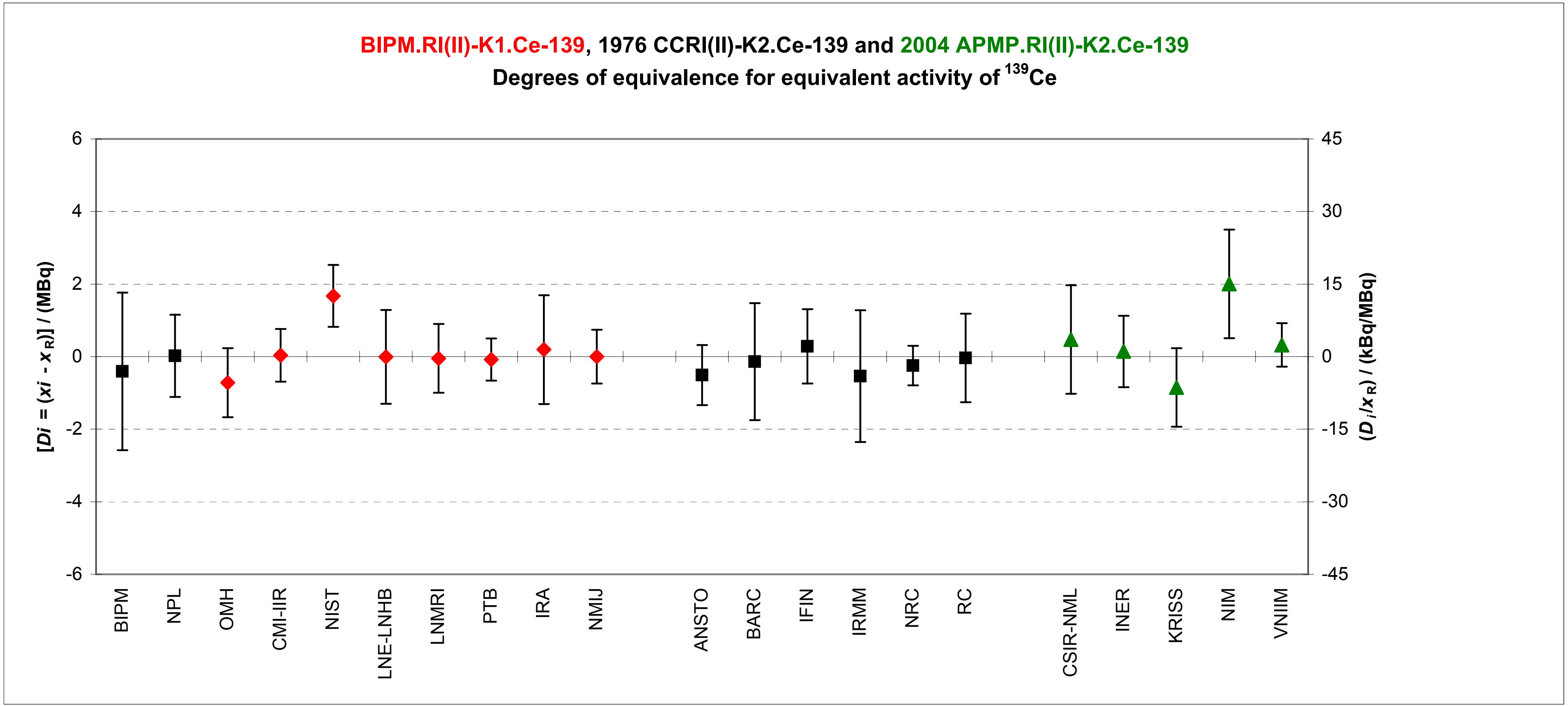
Lab *i*	D_i / MBq	U_i / MBq	BIPM D_{ij} / MBq	BIPM U_{ij} / MBq	NPL D_{ij}	NPL U_{ij}	OMH D_{ij}	OMH U_{ij}	CMI-IIR D_{ij}	CMI-IIR U_{ij}	NIST D_{ij}	NIST U_{ij}	LNE-LNHB D_{ij}	LNE-LNHB U_{ij}	LNMRI D_{ij}	LNMRI U_{ij}	PTB D_{ij}	PTB U_{ij}	IRA D_{ij}	IRA U_{ij}	NMIJ D_{ij}	NMIJ U_{ij}	ANSTO D_{ij}	ANSTO U_{ij}
BIPM	**-0.4**	2.2			**-0.4**	2.6	**0.3**	2.5	**-0.4**	2.5	**-2.1**	2.5	**-0.4**	2.7	**-0.4**	2.5	**-0.3**	2.4	**-0.6**	2.9	**-0.4**	2.5	**0.1**	2.5
NPL	**0.0**	1.1	**0.4**	2.6			**0.7**	1.5	**0.0**	1.4	**-1.7**	1.4	**0.0**	1.8	**0.1**	1.5	**0.1**	1.3	**-0.2**	2.0	**0.0**	1.4	**0.5**	1.4
OMH	**-0.7**	1.0	**-0.3**	2.5	**-0.7**	1.5			**-0.8**	1.2	**-2.4**	1.2	**-0.7**	1.7	**-0.7**	1.4	**-0.6**	1.1	**-0.9**	1.9	**-0.7**	1.2	**-0.2**	1.2
CMI-IIR	**0.0**	0.7	**0.4**	2.5	**0.0**	1.4	**0.8**	1.2			**-1.6**	1.0	**0.0**	1.5	**0.1**	1.2	**0.1**	0.8	**-0.2**	1.7	**0.0**	1.0	**0.5**	1.0
NIST	**1.7**	0.9	**2.1**	2.5	**1.7**	1.4	**2.4**	1.2	**1.6**	1.0			**1.7**	1.6	**1.7**	1.2	**1.8**	0.9	**1.5**	1.8	**1.7**	1.0	**2.2**	1.1
LNE-LNHB	**0.0**	1.3	**0.4**	2.7	**0.0**	1.8	**0.7**	1.7	**0.0**	1.5	**-1.7**	1.6			**0.0**	1.7	**0.1**	1.4	**-0.2**	2.1	**0.0**	1.5	**0.5**	1.6
LNMRI	**0.0**	1.0	**0.4**	2.5	**-0.1**	1.5	**0.7**	1.4	**-0.1**	1.2	**-1.7**	1.2	**0.0**	1.7			**0.0**	1.1	**-0.2**	1.9	**-0.1**	1.2	**0.5**	1.2
PTB	**-0.1**	0.6	**0.3**	2.4	**-0.1**	1.3	**0.6**	1.1	**-0.1**	0.8	**-1.8**	0.9	**-0.1**	1.4	**0.0**	1.1			**-0.3**	1.7	**-0.1**	0.8	**0.4**	0.9
IRA	**0.2**	1.5	**0.6**	2.9	**0.2**	2.0	**0.9**	1.9	**0.2**	1.7	**-1.5**	1.8	**0.2**	2.1	**0.2**	1.9	**0.3**	1.7			**0.2**	1.7	**0.7**	1.8
NMIJ	**0.0**	0.7	**0.4**	2.5	**0.0**	1.4	**0.7**	1.2	**0.0**	1.0	**-1.7**	1.0	**0.0**	1.5	**0.1**	1.2	**0.1**	0.8	**-0.2**	1.7			**0.5**	1.1
ANSTO	**-0.5**	0.8	**-0.1**	2.5	**-0.5**	1.4	**0.2**	1.2	**-0.5**	1.0	**-2.2**	1.1	**-0.5**	1.6	**-0.5**	1.2	**-0.4**	0.9	**-0.7**	1.8	**-0.5**	1.1		
BARC	**-0.1**	1.6	**0.3**	2.8	**-0.2**	2.0	**0.6**	1.9	**-0.2**	1.7	**-1.8**	1.8	**-0.1**	2.1	**-0.1**	1.9	**-0.1**	1.7	**-0.3**	2.3	**-0.1**	1.7	**0.4**	1.7
IFIN	**0.3**	1.0	**0.7**	2.5	**0.3**	1.5	**1.0**	1.4	**0.3**	1.2	**-1.4**	1.3	**0.3**	1.7	**0.3**	1.4	**0.4**	1.1	**0.1**	1.9	**0.3**	1.2	**0.8**	1.2
IRMM	**-0.5**	1.8	**-0.1**	2.9	**-0.6**	2.2	**0.2**	2.0	**-0.6**	1.9	**-2.2**	2.0	**-0.5**	2.3	**-0.5**	2.0	**-0.5**	1.9	**-0.7**	2.4	**-0.5**	1.9	**0.0**	1.9
NRC	**-0.2**	0.5	**0.2**	2.4	**-0.3**	1.3	**0.5**	1.1	**-0.3**	0.8	**-1.9**	0.9	**-0.2**	1.4	**-0.2**	1.1	**-0.2**	0.7	**-0.4**	1.7	**-0.3**	0.9	**0.3**	0.8
RC	**0.0**	1.2	**0.4**	2.6	**-0.1**	1.7	**0.7**	1.5	**-0.1**	1.4	**-1.7**	1.4	**0.0**	1.8	**0.0**	1.5	**0.0**	1.3	**-0.2**	2.0	**0.0**	1.4	**0.5**	1.4
CSIR-NML	**0.5**	1.5	**0.9**	2.8	**0.5**	1.9	**1.2**	1.8	**0.4**	1.6	**-1.2**	1.7	**0.5**	2.0	**0.5**	1.8	**0.6**	1.6	**0.3**	2.2	**0.5**	1.5	**1.0**	1.7
INER	**0.1**	1.0	**0.6**	2.5	**0.1**	1.5	**0.9**	1.4	**0.1**	1.2	**-1.5**	1.2	**0.2**	1.7	**0.2**	1.4	**0.2**	1.1	**-0.1**	1.9	**0.1**	1.0	**0.7**	1.2
KRISS	**-0.8**	1.1	**-0.4**	2.6	**-0.9**	1.6	**-0.1**	1.4	**-0.9**	1.3	**-2.5**	1.3	**-0.8**	1.7	**-0.8**	1.4	**-0.8**	1.2	**-1.0**	1.9	**-0.9**	1.1	**-0.3**	1.3
NIM	**2.0**	1.5	**2.4**	2.8	**2.0**	1.9	**2.7**	1.8	**2.0**	1.6	**0.3**	1.7	**2.0**	2.0	**2.1**	1.8	**2.1**	1.6	**1.8**	2.2	**2.0**	1.5	**2.5**	1.7
VNIIM	**0.3**	0.6	**0.7**	2.4	**0.3**	1.3	**1.0**	1.1	**0.3**	0.9	**-1.4**	0.9	**0.3**	1.5	**0.4**	1.1	**0.4**	0.7	**0.1**	1.7	**0.3**	0.6	**0.8**	1.0

Lab *j* ⇒

Lab *i* ⇓

Lab *i*	D_i / MBq	U_i / MBq	BARC D_{ij} / MBq	BARC U_{ij} / MBq	IFIN D_{ij}	IFIN U_{ij}	IRMM D_{ij}	IRMM U_{ij}	NRC D_{ij}	NRC U_{ij}	RC D_{ij}	RC U_{ij}	CSIR-NML D_{ij}	CSIR-NML U_{ij}	INER D_{ij}	INER U_{ij}	KRISS D_{ij}	KRISS U_{ij}	NIM D_{ij}	NIM U_{ij}	VNIIM D_{ij}	VNIIM U_{ij}
BIPM	**-0.4**	2.2	**-0.3**	2.8	**-0.7**	2.5	**0.1**	2.9	**-0.2**	2.4	**-0.4**	2.6	**-0.9**	2.8	**-0.6**	2.5	**0.4**	2.6	**-2.4**	2.8	**-0.7**	2.4
NPL	**0.0**	1.1	**0.2**	2.0	**-0.3**	1.5	**0.6**	2.2	**0.3**	1.3	**0.1**	1.7	**-0.5**	1.9	**-0.1**	1.5	**0.9**	1.6	**-2.0**	1.9	**-0.3**	1.3
OMH	**-0.7**	1.0	**-0.6**	1.9	**-1.0**	1.4	**-0.2**	2.0	**-0.5**	1.1	**-0.7**	1.5	**-1.2**	1.8	**-0.9**	1.4	**0.1**	1.4	**-2.7**	1.8	**-1.0**	1.1
CMI-IIR	**0.0**	0.7	**0.2**	1.7	**-0.3**	1.2	**0.6**	1.9	**0.3**	0.8	**0.1**	1.4	**-0.4**	1.6	**-0.1**	1.2	**0.9**	1.3	**-2.0**	1.6	**-0.3**	0.9
NIST	**1.7**	0.9	**1.8**	1.8	**1.4**	1.3	**2.2**	2.0	**1.9**	0.9	**1.7**	1.4	**1.2**	1.7	**1.5**	1.2	**2.5**	1.3	**-0.3**	1.7	**1.4**	0.9
LNE-LNHB	**0.0**	1.3	**0.1**	2.1	**-0.3**	1.7	**0.5**	2.3	**0.2**	1.4	**0.0**	1.8	**-0.5**	2.0	**-0.2**	1.7	**0.8**	1.7	**-2.0**	2.0	**-0.3**	1.5
LNMRI	**0.0**	1.0	**0.1**	1.9	**-0.3**	1.4	**0.5**	2.0	**0.2**	1.1	**0.0**	1.5	**-0.5**	1.8	**-0.2**	1.4	**0.8**	1.4	**-2.1**	1.8	**-0.4**	1.1
PTB	**-0.1**	0.6	**0.1**	1.7	**-0.4**	1.1	**0.5**	1.9	**0.2**	0.7	**0.0**	1.3	**-0.6**	1.6	**-0.2**	1.1	**0.8**	1.2	**-2.1**	1.6	**-0.4**	0.7
IRA	**0.2**	1.5	**0.3**	2.3	**-0.1**	1.9	**0.7**	2.4	**0.4**	1.7	**0.2**	2.0	**-0.3**	2.2	**0.1**	1.9	**1.0**	1.9	**-1.8**	2.2	**-0.1**	1.7
NMIJ	**0.0**	0.7	**0.1**	1.7	**-0.3**	1.2	**0.5**	1.9	**0.3**	0.9	**0.0**	1.4	**-0.5**	1.5	**-0.1**	1.0	**0.9**	1.1	**-2.0**	1.5	**-0.3**	0.6
ANSTO	**-0.5**	0.8	**-0.4**	1.7	**-0.8**	1.2	**0.0**	1.9	**-0.3**	0.8	**-0.5**	1.4	**-1.0**	1.7	**-0.7**	1.2	**0.3**	1.3	**-2.5**	1.7	**-0.8**	1.0
BARC	**-0.1**	1.6			**-0.4**	1.8	**0.4**	2.4	**0.1**	1.6	**-0.1**	1.9	**-0.6**	2.2	**-0.3**	1.9	**0.7**	1.9	**-2.1**	2.2	**-0.5**	1.7
IFIN	**0.3**	1.0	**0.4**	1.8			**0.8**	2.0	**0.5**	1.0	**0.3**	1.5	**-0.2**	1.8	**0.1**	1.4	**1.1**	1.5	**-1.7**	1.8	**0.0**	1.1
IRMM	**-0.5**	1.8	**-0.4**	2.4	**-0.8**	2.0			**-0.3**	1.8	**-0.5**	2.1	**-1.0**	2.3	**-0.7**	2.0	**0.3**	2.1	**-2.5**	2.3	**-0.9**	1.9
NRC	**-0.2**	0.5	**-0.1**	1.6	**-0.5**	1.0	**0.3**	1.8			**-0.2**	1.2	**-0.7**	1.6	**-0.4**	1.1	**0.6**	1.2	**-2.3**	1.6	**-0.6**	0.8
RC	**0.0**	1.2	**0.1**	1.9	**-0.3**	1.5	**0.5**	2.1	**0.2**	1.2			**-0.5**	1.9	**-0.2**	1.5	**0.8**	1.6	**-2.0**	1.9	**-0.4**	1.3
CSIR-NML	**0.5**	1.5	**0.6**	2.2	**0.2**	1.8	**1.0**	2.3	**0.7**	1.6	**0.5**	1.9			**0.3**	1.6	**1.3**	1.7	**-1.5**	2.0	**0.2**	1.4
INER	**0.1**	1.0	**0.3**	1.9	**-0.1**	1.4	**0.7**	2.0	**0.4**	1.1	**0.2**	1.5	**-0.3**	1.6			**1.0**	1.3	**-1.9**	1.6	**-0.2**	0.9
KRISS	**-0.8**	1.1	**-0.7**	1.9	**-1.1**	1.5	**-0.3**	2.1	**-0.6**	1.2	**-0.8**	1.6	**-1.3**	1.7	**-1.0**	1.3			**-2.9**	1.7	**-1.2**	1.0
NIM	**2.0**	1.5	**2.1**	2.2	**1.7**	1.8	**2.5**	2.3	**2.3**	1.6	**2.0**	1.9	**1.5**	2.0	**1.9**	1.6	**2.9**	1.7			**1.7**	1.4
VNIIM	**0.3**	0.6	**0.5**	1.7	**0.0**	1.1	**0.9**	1.9	**0.6**	0.8	**0.4**	1.3	**-0.2**	1.4	**0.2**	0.9	**1.2**	1.0	**-1.7**	1.4		

Figure 1. **Graph of degrees of equivalence with the KCRV for ^{139}Ce**
(as it appears in Appendix B of the MRA)



N.B. the right hand axis indicates approximate relative values only

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

Uncertainty budget for the NMIJ measurement of 2004

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
linear extrapolation, including weighing and counting type A uncertainties	17	–
weighing	–	5
background	–	1
fixed dead time	–	1
resolving time	–	3
timing	–	3
decay correction	–	1
Quadratic summation	**17**	**7**
Relative combined standard uncertainty, u_c	**18**	

Appendix 2. 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**
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop counter	PP
2π	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
		bolometer	BO
		calorimeter	CA
		PIPS detector	PS

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 + γ)	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 **method**	**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
4π CsI-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