

Update of the BIPM comparison BIPM.RI(II)-K1.Ba-133
of activity measurements of the radionuclide ^{133}Ba to include the results of the
IFIN-HH (Romania) and the NMIJ (Japan)

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Abstract

Since 2005, the Institutul de Fizica si Inginerie Nucleara- "Horia Hulubei" (IFIN-HH) of Romania and the National Metrology Institute of Japan (NMIJ) have each submitted samples of known activity of ^{133}Ba to the International Reference System (SIR). The values of the activity submitted were about 1.4 MBq and 6.6 MBq. The IFIN result is their first contribution to the SIR for this radionuclide and for the NMIJ this key comparison result has replaced their earlier result dating from 1986. Each of these two new results has contributed to the updating of the KCRV for this radionuclide and is included in the matrix of degrees of equivalence in the key comparison database that now contains nine results for the comparison identified as BIPM.RI(II)-K1.Ba-133.

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 form or a different standard ampoule for radioactive gases. 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 reference 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 2006, the SIR has measured 894 ampoules to give 655 independent results for 63 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 BIPM key comparison database (KCDB) that was set up under the CIPM Mutual Recognition Arrangement (CIPM MRA) [2]. The results described in this report form part of the BIPM.RI(II)-K1.Ba-133 key comparison [3].

In addition, an international comparison was held in 1984 for this radionuclide, CCRI(II)-K2.Ba-133 [4] and this comparison has been given the status of having provisional equivalence in the KCDB. Consequently, these results are included in the degrees of equivalence published in the KCDB.

2. Participants

Fifteen NMIs and three other laboratories have submitted thirty-five ampoules for the comparison of ^{133}Ba activity measurements since 1977. All the laboratory details are given in Table 1 as the KCRV has been updated. 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. The AECL was an invited participant in various SIR comparisons, as in the early years, J.G.V. Taylor of the AECL was a personal member of the predecessor to the CCRI(II).

Table 1. Details of the participants in the BIPM.RI(II)-K1.Ba-133

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM YYYY-MM-DD
OMH	MKEH	Magyar Kereskedelmi Engedélyezési Hivatal	Hungary	EUROMET	1977-05-26 1978-10-26 1980-06-30 1991-04-10 1996-12-10
ASMW*	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1978-03-13 1978-11-08 1981-10-13
AAEC	ANSTO	Australian Nuclear Science and Technology Organisation	Australia	SIM	1978-09-01
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1978-11-06 1980-02-07
AECL	–	Atomic Energy of Canada Ltd	Canada	SIM	1978-11-09

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Table 1 continued. Details of the participants in the BIPM.RI(II)-K1.Ba-133

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
IAEA	–	International Atomic Energy Agency	–	–	1979-02-14
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1979-03-05 1986-07-24 1998-11-16
LMRI	LNE-LNHB	Laboratoire national de métrologie et d'essai - Laboratoire national Henri Becquerel	France	EUROMET	1979-11-07
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1982-05-04
BIPM	–	Bureau International des Poids et Mesures	–	–	1983-11-16
IMM	VNIIM	D.I. Mendeleev Institute for Metrology	Russia	COOMET	1984-04-18
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1986-02-07 2006-07-07
PSPKR	PTKMR	Pusat Teknologi Keselamatan dan Metrologi Radiasi	Indonesia	APMP	1991-08-21
–	KRISS	Korea Research Institute of Standards and Science	Republic of Korea	APMP	1995-01-19
–	LNMRI	Laboratorio Nacional de Metrologia das Radiações Ionizantes	Brazil	SIM	1995-11-21

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Table 1 continued. Details of the participants in the BIPM.RI(II)-K1.Ba-133

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
–	CNEA	Comision Nacional de Energia Atomica	Argentina	SIM	1996-12-20
–	BEV	Bundesamt für Eich- und Vermessungswesen	Austria	EUROMET	1998-06-25
–	IFIN-HH	Institutul de Fizica si Inginerie Nucleara- "Horia Hulubei"	Romania	EUROMET	2005-12-08

* another laboratory in 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 any correlations are taken into account. For this comparison, the BEV ionization chamber is traceable to the NPL.

A brief description of the standardization methods for each laboratory, the activities submitted and the relative uncertainties are given in Table 2. The uncertainty budgets for the two recent results are given in Appendix 1.

The half-life used by the BIPM is 3835 (37) d [5]. The data could be revised using the half-life from the Monographie 5 [6], 3850 (2) d. However, the updated degrees of equivalence would not differ significantly as the SIR measurements were performed within less than one year following the reference date. In the extreme case of nine months, for the IAEA, the relative change in A_e is about 2×10^{-4} . In the 1984 CCRI(II)-K2.Ba-133 comparison a half life of 3846 (15) days being the arithmetic mean of the measurements in [7] was used.

Details regarding the solution submitted are shown in Table 3, including any impurities, when present, as identified by the laboratories. The BIPM standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer is described in [8]. The CCRI(II) agreed in 1999 [9] that this method should be followed according to the protocol described in [10] when an NMI makes such a request or when there appear to be discrepancies. No impurity measurements were carried out at the BIPM for the two latest submissions.

Table 2. Standardization methods of the participants for ^{133}Ba

NMI	Method used and acronym (see Appendix 2)	Half-life	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
MKEH	4 π (e,x)- γ coincidence 4P-PP-MX-NA-GR-CO	10.5 (1) a	3535	77-04-30	0.10	0.80
			3534	12 h UT		
	973.4		78-08-01	0.07	0.60	
	973.8		12 h UT			
	4 π (e _x ,x)- γ coincidence 4P-PP-MX-NA-GR-CO	3578	80-10-01	0.02	0.80	
	3476	91-07-01	0.02	0.17		
	4 π (e,x)- γ coincidence 4P-PP-MX-NA-GR-CO	3862 (15) d	1585	96-12-01 0 h UT	0.02	0.20
PTB	4 π (PC)- γ and 4P-PC-00-NA-GR-CO 4 π (PPC)- γ coincidence 4P-PP-00-NA-GR-CO	–	1924	78-01-01 0 h UT	0.03	0.20
	4 π (PC)- γ coincidence 4P-PC-00-NA-GR-CO	–	3235 3365	78-10-01 12 h UT	0.10	0.20
	Pressurized IC 4P-IC-GR-00-00-00	–	4531 4506	82-01-01 0 h UT	0.08	2.01
ANSTO	4 π β - γ coincidence 4P-00-BR-NA-GR-CO	–	347.7	78-07-01 0 h UT	0.20	0.27
CMI-IIR	x- γ coincidence 4P-00-XR-NA-GR-CO	10.35 a	3637	78-09-08 11 h UT	0.12	0.62
	4 π x- γ coincidence 4P-00-XR-NA-GR-CO		4303	79-11-28 11 h UT	0.12	0.62
AECL	4 π (PC)- γ coincidence 4P-PC-MX-NA-GR-CO	–	3849 4200	78-02-28 17 h UT	0.05	0.25
IAEA /RCC †	–	10.8 a	1618	78-05-01 12 h UT	0.10	0.70

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Table 2 continued. Standardization methods of the participants for ^{133}Ba

NMI	Method used and acronym (see Appendix 2)	Half-life	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
NIST	$4\pi(e,x)\text{-}\gamma$ coincidence 4P-00-MX-NA-GR-CO	10.74 a	2004	78-09-01 17 h UT	0.13	0.29
	Pressurized IC ^b 4P-IC-GR-00-00-00	–	3585	86-07-01 0 h UT	0.07	0.21
	Pressurized IC ^c 4P-IC-GR-00-00-00	10.52 (13) a	1259	98-11-16 12 h UT	0.04	0.26
LNE-LNHB	$4\pi(\text{NaI}(\text{Tl}))\gamma$ 4P-NA-GR-00-00-00	3835 (37) d	4083 4067	79-04-11 0 h UT	0.01	0.14
NPL	Pressurized IC ^d 4P-IC-GR-00-00-00	–	2663	82-04-28 0 h UT	0.03	0.96
BIPM ^a	$4\pi(\text{PC})\text{-}\gamma$ coincidence and selective sampling 4P-PC-00-NA-GR-CO 4P-PC-00-NA-GR-SS	3846 (15) d	4174.0 4176.0 4175.6	84-03-15 0 h UT	0.03	0.11
VNIIM	$4\pi(e,x)\text{-}\gamma$ coincidence 4P-00-MX-NA-GR-CO	–	6876 6779	83-11-28 12 h UT	0.21	0.77
NMIJ	$4\pi(\text{PC})\text{x-}\gamma$ coincidence 4P-PC-XR-NA-GR-CO	–	2148 2085	86-02-04 12 h UT	0.09	0.34
	$4\pi(e,x)\text{-}\gamma$ coincidence 4P-PC-MX-NA-GR-CO	[6]	6629	06-04-01 0 h UT	0.21	0.03
PTKMR	$4\pi(\text{PC})\text{-}\gamma$ coincidence 4P-PC-00-NA-GR-CO	–	568.6	91-05-23 0 h UT	0.07	–
KRISS	$4\pi(e,x)\text{-}\gamma$ coincidence 4P-PP-MX-NA-GR-CO	–	507	94-10-01 0 h UT	0.30	0.45
LNMRI	$4\pi(e_A,x)\text{-}\gamma$ coincidence 4P-00-MX-NA-GR-CO	–	820	95-07-31 0 h UT	0.06	0.29
CNEA	$4\pi(\text{PC})\text{-}\gamma$ coincidence 4P-PC-00-NA-GR-CO	–	3275	96-12-03 12 h UT	0.20	0.80

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Table 2 continued. Standardization methods of the participants for ^{133}Ba

NMI	Method used and acronym (see Appendix 3)	Half-life	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
BEV	Pressurized IC ^e	3848 d	767	98-06-01 0 h UT	0.28	0.66
IFIN-HH	$4\pi\beta\text{-}\gamma$ coincidence 4P-PC-MX-NA-GR-CO	10.57 a	1431	05-11-14 12 h UT	0.10	0.27

[†] The Radiochemical Centre Ltd, Amersham

^a the three ampoules measured by the BIPM for the CCRI(II)-K2.Ba-133 and measured in the SIR are used to make the link for the CCRI(II) key comparison

^b calibrated by $4\pi\beta(\text{PPC})\text{-}\gamma$ coincidence for the nuclide considered

^c calibrated by $4\pi(\text{e,x})\text{-}\gamma$ anticoincidence for the nuclide considered in 1983

^d calibrated by a primary method for the nuclide considered

^e calibrated by the NPL.

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 ^{133}Ba arise from thirty-five ampoules and the SIR equivalent activity for each ampoule is given in Table 4. 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. 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].

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, six of these results, for the ANSTO, LNE-LNHB, CMI-IIR, NPL, PTB and the VNIIM have been superseded by the international comparison that was held in 1984 [4]. The 1984 comparison also enables the linking of another three NMIs, the IRA, NMISA and the NRC, and an international laboratory, the IRMM, to the SIR key comparison. Finally, the IAEA no longer makes activity measurements and the result from the AECL is not included, as the AECL is not a designated laboratory of the NRC, Canada. Although Indonesia has signed the MRA, the result from the PTKMR is not included in the KCDB as they have not yet been designated as a national laboratory holding activity standards.

Table 3. Details of the solution of ¹³³Ba submitted

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. / (µg g ⁻¹)	Density / (g cm ⁻³)	Relative activity of the impurity [†]
MKEH	Ba in HCl	0.1	Ba : 25	—	⁶⁵ Zn : 0.0022 (5)% ¹³⁴ Cs : 0.0007 (2) %
	BaCl ₂ in HCl	0.1	BaCl ₂ : 25	—	¹³⁴ Cs : 0.0060 (15) %
	Ba in HCl	0.1	Ba: 25	—	⁶⁰ Co : 0.007 (2) %
	BaCl ₂ in HCl	0.1	BaCl ₂ : 25	—	⁶⁰ Co : 0.0020 (3) % ¹³⁴ Cs : 0.020 (5) %
				—	⁶⁰ Co : 0.0010 (3) % ¹³⁴ Cs : 0.0040 (5) %
PTB	BaCl ₂ in HCl	0.1	BaCl ₂ : 30	1.0	<0.01 %
	BaCl ₂ in HNO ₃	0.1	BaCl ₂ : 500	1.003	—
	BaCl ₂ in HCl	1	BaCl ₂ : 23	1.00	—
ANSTO	BaCl ₂ in HCl	1	BaCl ₂ : 20	1.00	—
CMI-IIR	BaCl ₂ in HCl	0.08	BaCl ₂ : 30	—	< 0.2 %
					¹³¹ Ba : 0.15 (3) %
AECL	BaCl ₂ in HCl	0.6	BaCl ₂ : ~ 10	1.02	—
IAEA /RCC	—	—	Ba : 100		< 0.05 %
NIST	BaCl ₂ in HCl	1	BaCl ₂ : 118	1.014 (2)	¹³⁴ Cs : 0.0018 (2) %
			BaCl ₂ : 62 *	1.015	< 0.01 %
			BaCl ₂ : 120	1.015	—
LNE-LNHB	BaCl ₂ in HCl	1	BaCl ₂ : 15	1.02	< 10 ⁻⁴ %
NPL	BaCl ₂ in HCl	1	Ba : 50	1.015	—
BIPM	BaCl ₂ in HCl	1	BaCl ₂ : 60 *	1.016	¹³⁴ Cs : 0.0011(5) %
VNIIM	BaCl ₂ in HCl	0.1	Ba : 40	1.001	—
NMIJ	BaCl ₂ in HCl	1	BaCl ₂ : 50	1.015	—
	BaCl ₂ in HCl	0.1	BaCl ₂ : 50	1.004	—
PTKMR	BaCl ₂ in HCl	2.7	BaCl ₂ : 28	1	—
KRISS	BaCl ₂ .2H ₂ O in HCl	0.5	BaCl ₂ .2H ₂ O : 1.9	1.0073	¹³⁴ Cs : 0.29 (1) %
LNMRI	BaCl ₂ in HCl	1	BaCl ₂ : 50	1.013	—

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Table 3 continued. Details of the solution of ¹³³Ba submitted

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. / (μg g ⁻¹)	Density / (g cm ⁻³)	Relative activity of the impurity [†]
CNEA	BaCl ₂ .2H ₂ O in HCl	0.1	BaCl ₂ .2H ₂ O : 36	0.9904	¹³⁴ Cs : 0.30 (1) % ¹³⁷ Cs : 0.070 (7) %
BEV	BaCl ₂ in HCl	0.1	BaCl ₂ : 30	1.0	—
IFIN-HH	BaCl ₂ in HCl	0.1	BaCl ₂ : 70	1	¹³⁴ Cs : 0.020 (7) %

* same solution as that for the 1984 CCRI(II)-K2.Ba-133 comparison

[†] the ratio of the activity of the impurity to the activity of ¹³³Ba at the reference date.

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:

- only primary standardized solutions 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 included¹;
- each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);
- 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;
- 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 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 only made by the CCRI(II), normally during one of its biennial meetings. This was the case in May 2007, when the CCRI(II) agreed to include the results of the IFIN-HH and the new result from the NMIJ in the KCRV.

Consequently, the KCRV for ¹³³Ba has been identified as 43 932 (67) kBq using the results in Table 4 from the PTB(1978), ANSTO, ASMW(1978), AECL, LNE-LNHB, CMI-IIR(1980), NPL, BIPM, VNIIM, KRISS, LNMRI, MKEH(1996), CNEA, NIST(1998), IFIN-HH and the NMIJ(2006).

¹ Rule modified at the CCRI(II) meeting in 2005.

Table 4. Results of SIR measurements of ^{133}Ba

NMI	Mass of solution m_i / g	Activity submitted A_i / kBq	N° of Ra source used	SIR A_e / kBq	Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / kBq
MKEH	3.605 2	3535	3	44 360	7×10^{-4}	360
	3.604 4	3534		44 340	8×10^{-4}	360
	3.601 3	973.4	2	44 540	10×10^{-4}	270
	3.602 8	973.8		44 520		270
	3.603 7	3578	3	43 990	8×10^{-4}	350
	3.614 8	3476	3	43 636	8×10^{-4}	80
3.630 1	1585	2	43 772	10×10^{-4}	97	
PTB	3.615 5(1)	1924	2	43 910	11×10^{-4}	100
	3.554 39	3235	3	43 948	8×10^{-4}	105
	3.697 75	3365		43 946 [†]		104
	3.648 63	4531	3	43 930	8×10^{-4}	890
	3.629 01	4506		43 930 ^{#†}		890
ANSTO	3.659 2	347.7	1	43 910 [#]	13×10^{-4}	160
CMI-IIR	3.630 06	3637	3	44 310	8×10^{-4}	280
	3.554 93	4303	3	43 900 [#]	8×10^{-4}	280
AECL	0.301 524	3849	3	43 788	9×10^{-4}	118
	0.329 044 [*]	4200		43 773 [†]		118
IAEA /RCC	3.601 4	1618	2	43 730	12×10^{-4}	330
NIST	3.664 17	2004	3	44 330	8×10^{-4}	150
	3.602 23	3585	3	43 730	8×10^{-4}	100
	3.626 02	1259	2	43 910	11×10^{-4}	120
LNE-LNHB	3.688 61	4083	3	43 926	9×10^{-4}	72
	3.673 46	4067		43 930 ^{#†}		72
NPL	3.646 2	2663	3	43 990 [#]	9×10^{-4}	420
BIPM	3.601 39	4174.0	3	43 784	8×10^{-4}	59
	3.603 12	4176.0		43 769	8×10^{-4}	58
	3.602 75	4175.6		43 748 [†]	8×10^{-4}	58
VNIIM	3.522 64	6876	3	43 430	8×10^{-4}	350
	3.472 69	6779		43 360 ^{#†}		350

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Table 4a continued. Results of SIR measurements of ¹³³Ba

NMI	Mass of solution / g	Activity submitted / kBq	N° of Ra source used	SIR A_e / kBq	Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / kBq
NMIJ	3.676 9	2148	3	44 189	8×10^{-4}	160
	3.569 2	2085		44 164 †		160
	3.599 44	6629	3	44 030	8×10^{-4}	100
PTKMR	3.678 4	568.6	1	41 801	14×10^{-4}	66
KRISS	3.575 98	507	1	44 300	15×10^{-4}	250
LNMRI	3.532 66	820	2	44 340	10×10^{-4}	140
CNEA	3.593 04	3275	3	44 440	9×10^{-4}	370
BEV	3.633	767	1	43 670	13×10^{-4}	320
IFIN-HH	3.641 33	1431	2	43 590	11×10^{-4}	140

† the mean of the two A_e values is used with an averaged uncertainty, as attributed to an individual entry [11]

values superseded in the KCDB by the international comparison in 1984

* mass of active solution before dilution.

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

$$D_i = A_{e_i} - \text{KCRV} \quad (1)$$

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

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

taking correlations into account as appropriate [12].

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

$$D_{ij} = D_i - D_j = A_{e_i} - A_{e_j} \quad (3)$$

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

$$u_{D_{ij}}^2 = u_i^2 + u_j^2 - 2u(A_{e_i}, A_{e_j}) \quad (4)$$

where any obvious correlations between the NMIs (such as a traceable calibration) are subtracted using the covariance $u(A_{e_i}, A_{e_j})$, 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 appear in Appendix B of the KCDB. The core of the matrix is based on nine values from the SIR. The additional matrix cells show the ten results from the 1984 international (CCRI(II)) comparison linked to those of the SIR [4]. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{e_i} 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 (identified as x_R in the KCDB), is shown in Figure 1 where, following the advice of the CCRI, measurements made prior to 1987 are indicated as black squares. This 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.

Conclusion

The BIPM ongoing key comparison for ^{133}Ba , BIPM.RI(II)-K1.Ba-133 currently comprises nine results. These have been analysed with respect to the updated 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 nine other NMIs and one international laboratory that took part in the CCRI(II)-K2.Ba-133 comparison in 1984 were previously linked to the BIPM ongoing key comparison through three ampoules of the comparison measured in the SIR [3]. These linked results are included in the matrix of degrees of equivalence approved by the CCRI(II).

Other results may be added as and when other NMIs contribute ^{133}Ba activity measurements to this comparison or take part in other linked comparisons.

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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, 14 October 1999, 45 pp. Available from the BIPM website: <http://www.bipm.org/pdf/mra.pdf>.
- [3] Ratel G., Michotte C., BIPM comparison BIPM.RI(II)-K1.Ba-133 of activity measurements of the radionuclide ^{133}Ba and links for the 1984 international comparison CCRI(II)-K2.Ba-133, 2003, *Metrologia* **40** 06011.
- [4] Ratel G., Activity concentration of a solution of ^{133}Ba : results of an international comparison, *Nucl. Inst. Meth. Phys. Res.*, **A316**, (1992), 318-323.
- [5] NCRP Report No. 58, A Handbook of radioactivity measurements procedures, 2nd ed. (1985), Bethesda.
- [6] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Helmer R., Nichols A., Schönfeld E., Dersch R., Table of radionuclides, *BIPM Monographie 5*, 2004, volume 1.
- [7] Hoppes D.D. and Schima F.J. (eds.) Nuclear data for the efficiency calibration of germanium spectrometer systems, *NBS special publication 626*, 1982.
- [8] Michotte C., Efficiency calibration of the Ge(Li) detector of the BIPM for SIR-type ampoules, *Rapport BIPM-1999/03*, 15 pp.
- [9] CCRI *Rapport de la 16^e session* (June 1999), CCRI(II) 81-82. (Paris: Off. Lib.)

- [10] Michotte C., Protocol on the use of the calibrated spectrometer of the BIPM for the measurement of impurities in ampoules submitted to the SIR, [CCRI\(II\)/01-01, 2001, 2p.](#)
- [11] Woods M.J., Reher D.F.G. and Ratel G., Equivalence in radionuclide metrology, *Applied Radiation and Isotopes*, **52**, (2000) 313-318.
- [12] Ratel G., Evaluation of the uncertainty of the degree of equivalence, 2005, [Metrologia 42, 140-144.](#)

Table 5. Table of degrees of equivalence and introductory text for ¹³³Ba

Key comparison BIPM.RI(II)-K1.Ba-133

MEASURAND : Equivalent activity of ¹³³Ba

Key comparison reference value: the SIR reference value for this radionuclide is $x_R = 43.932$ MBq with a standard uncertainty, $u_R = 0.067$ 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 $U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\sum u_i^2)^{1/2}$ 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.Ba-133 (1984) to BIPM.RI(II)-K1.Ba-133

The value x_i is the equivalent activity for laboratory i participant in CCRI(II)-K2.Ba-133 having been normalized to the value of the BIPM as the linking laboratory (see reference in Final report)

The degree of equivalence of laboratory i participant in CCRI(II)-K2.Ba-133 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.Ba-133 and one in CCRI(II)-K2.Ba-133, or both participant in CCRI(II)-K2.Ba-133, 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 $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_iu_j)^{1/2}$ is used with l being the linking laboratory when both laboratories are linked, and f is the correlation coefficient.

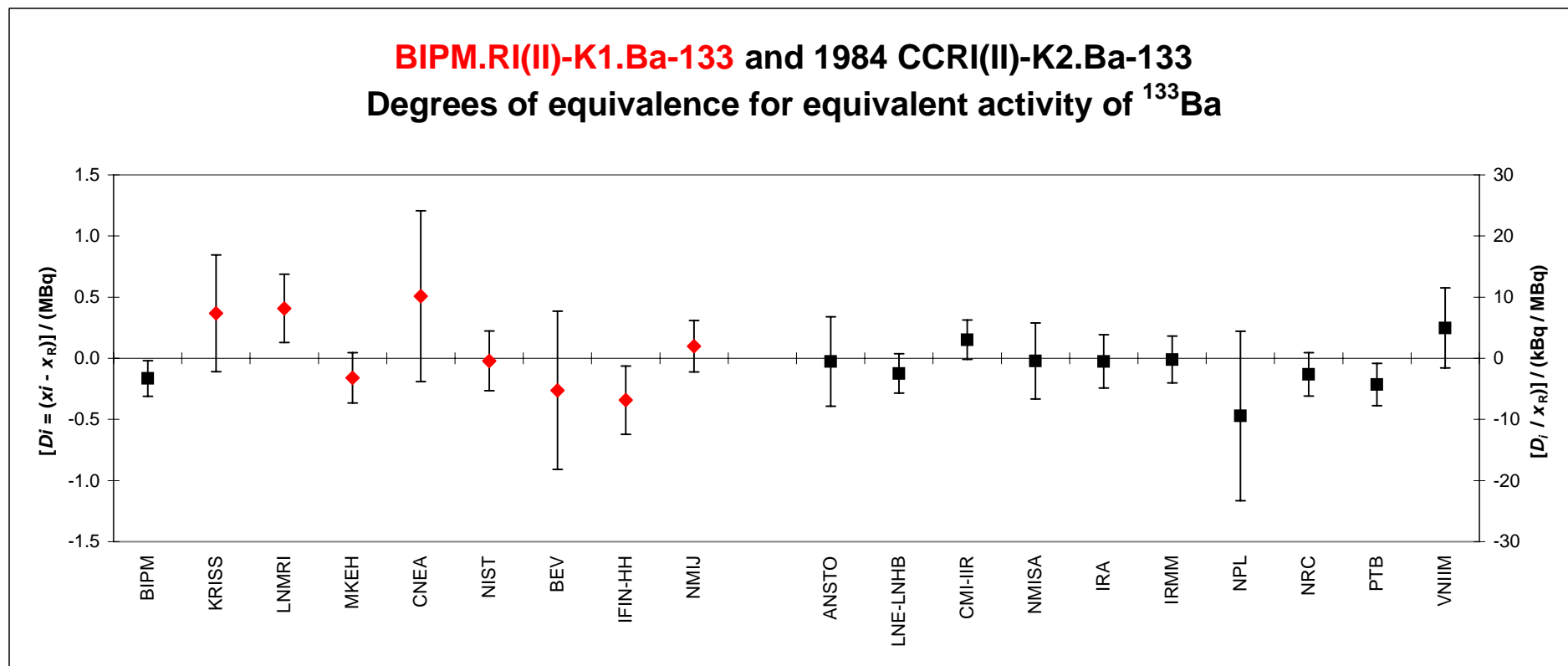
These statements make it possible to extend the BIPM.RI(II)-K1.Ba-133 matrices of equivalence to the other participants in CCRI(II)-K2.Ba-133.

Table 5 continued

Lab <i>i</i> ↓	D_i / MBq		U_i / MBq		BIPM		KRISS		LNMRI		MKEH		CNEA		NIST		BEV		IFIN-HH		NMIJ		ANSTO	
	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq
BIPM	-0.16	0.15																						
KRISS	0.37	0.48																						
LNMRI	0.41	0.28																						
MKEH	-0.16	0.21																						
CNEA	0.51	0.70																						
NIST	-0.02	0.24																						
BEV	-0.26	0.65																						
IFIN-HH	-0.34	0.28																						
NMIJ	0.10	0.21																						
ANSTO	-0.03	0.37																						
LNE-LNHB	-0.12	0.16																						
CMI-IIR	0.15	0.16																						
NMISA	-0.02	0.31																						
IRA	-0.03	0.22																						
IRMM	-0.01	0.19																						
NPL	-0.47	0.69																						
NRC	-0.13	0.18																						
PTB	-0.21	0.17																						
VNIIM	0.25	0.33																						

Lab <i>i</i> ↓	D_i / MBq		U_i / MBq		LNE-LNHB		CMI-IIR		NMISA		IRA		IRMM		NPL		NRC		PTB		VNIIM		
	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	D_{ij} / MBq	U_{ij} / MBq	
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NPL	-0.47	0.69																					
NRC	-0.13	0.18																					
PTB	-0.21	0.17																					
VNIIM	0.25	0.33																					

Figure 1. Graph of degrees of equivalence with the KCRV for ^{133}Ba
 (as it appears in Appendix B of the MRA)



N.B. The right-hand axis gives approximate relative values only

Appendix 1. Uncertainty budgets for the activity of ^{133}Ba submitted to the SIR**Uncertainty budget for the IFIN-HH measurement of 2005**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	10	–
weighing	–	15
dead time	–	3
resolving time	–	15
extrapolation of efficiency curve	–	16
background	–	1
impurities	–	0.7
Quadratic summation	10	27
Relative combined standard uncertainty, u_c	29	

Uncertainty budget for the NMIJ measurement of 2006

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	10*	–
weighing	5*	–
dead time	–	< 1
resolving time	–	1
Gandy effect	–	2
counting time	–	1
half-life	–	< 1
background	–	1
extrapolation of efficiency curve	21	–
Quadratic summation	21	3
Relative combined standard uncertainty, u_c	21	

* included in the extrapolation uncertainty

Appendix 3. 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 ($x + \gamma$)	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	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