

BIPM comparison BIPM.RI(II)-K1.Cs-137
of activity measurements of the radionuclide ^{137}Cs and links for the
1982 international comparison CCRI(II)-K2.Cs-137

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Abstract

Since 1976, fifteen national metrology institutes (NMIs) and two other laboratories have submitted thirty-four samples of known activity of ^{137}Cs to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures. The activities ranged from about 550 kBq to 67 MBq. The degrees of equivalence between each equivalent activity measured in the SIR and the key comparison reference value (KCRV) have been calculated and the results are given in the form of a matrix for eleven NMIs and the BIPM. A graphical presentation is also given. The results of this comparison have been approved by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), comparison identifier BIPM.RI(II)-K1.Cs-137. The results of a CCRI international comparison, identifier CCRI(II)-K2.Cs-137 held in 1982 that have been approved for provisional equivalence for this radionuclide, have been linked to the SIR results. This has enabled three NMIs to update their results and a further three NMIs and one international laboratory, the IRMM, to be linked to the SIR.

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 are all given in [1].

From its inception until 31 December 2002, the SIR has measured 835 ampoules to give 606 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 BIPM key comparison database (KCDB) that was set up under the Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Cs-137 key comparison.

In addition, an international comparison was held in 1982 for this radionuclide, CCRI(II)-K2.Cs-137 [3] and this comparison has been given the status of having provisional equivalence in the KCDB. Although nineteen laboratories took part in this comparison, nine of them have since submitted ampoules to the SIR. Three NMIs that had previously submitted ampoules to the SIR have updated their results and a further three NMIs and one international laboratory are eligible to be linked to the BIPM key comparison through this CCRI(II) comparison, as listed in Table 1b.

2. Participants

Fifteen NMIs and two other laboratories have submitted thirty-four ampoules for the comparison of ^{137}Cs activity measurements since 1976. An early result for another NMI has been withdrawn and is not included here. 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. 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 1a. Details of the participants in the BIPM.RI(II)-K1.Cs-137

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
ASMW*	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1976-11-16 1978-11-07 1997-11-13
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1977-02-02 1978-11-28 1980-02-08
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1977-05-18
–	OMH	Országos Mérésügyi Hivatal	Hungary	EUROMET	1977-05-27 1997-09-25

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Table 1a continued. Details of the participants in the BIPM.RI(II)-K1.Cs-137

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
AAEC	ANSTO	Australian Nuclear Science and Technology Organisation	Australia	APMP	1977-06-16 1994-06-03
AECL	–	Atomic Energy of Canada Ltd	Canada	–	1977-09-30
NAC*	CSIR-NML	National Metrology Laboratory	South Africa	SADCMET	1980-07-22
BIPM	–	Bureau International des Poids et Mesures	–	–	1982-04-07
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1983-07-19 2001-11-28
LMRI LPRI	BNM-LNHB	Bureau national de métrologie- Laboratoire national Henri Becquerel	France	EUROMET	1985-09-16 1998-12-14
PSPKR	P3KRBiN	Pusat Penelitian & Pengembangan Keselamatan Radiasi & Biomedika Nuklir	Indonesia	APMP	1989-09-25
–	CNEA	Comision Nacional de Energia Atomica	Argentina	SIM	1992-01-28
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1994-12-06
–	IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	1996-09-20 2000-12-07
–	BARC	Bhabha Atomic Research Centre	India	APMP	1997-04-30
–	BEV	Bundesamt für Eich- und Vermessungswesen	Austria	EUROMET	1998-10-14
–	NIM	National Institute of Metrology	China	APMP	1999-05-11

* another laboratory in the country

The three NMIs and the international organization that took part in the CCRI(II) international comparison, CCRI(II)-K2.Cs-137 in 1982 and are also eligible for the KCDB are shown in Table 1b together with the three NMIs that used this comparison to update their results.

Table 1b. Details of the participants in the 1982 CCRI(II)-K2.Cs-137 to be linked to BIPM.R.(II)-K1.Cs-137

Original acronym	NMI	Full name	Country	Regional metrology organization
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET
NAC*	CSIR-NML	National Metrology Laboratory	South Africa	SADCMET
–	IFIN	Institutul de Fizica si Inginerie Nucleara	Romania	EUROMET
(CBNM) IRMM	–	Institute for Reference Materials and Measurements	European Union	EUROMET
–	NPL	National Physical Laboratory	United Kingdom	EUROMET
–	NRC	National Research Council	Canada	SIM
–	SCK-CEN	Studiecentrum voor Kernenergie-Centre d'étude de l'Energie Nucléaire	Belgium	EUROMET

* 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.

A brief description of the standardization methods for each laboratory, the activities submitted and the relative uncertainties are given in Table 2. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. When submitted by the NMIs, the uncertainty budgets are given in Appendix 1 attached to this report. Consequently, uncertainty budgets are given for the IRA and the NIST.

The half-life used in the SIR is 11 020.8 (1.2) d. This is in agreement with the half-life recommended by the IAEA [5], 11 020 (60) d which will be used in the future. The value used in the 1982 CCRI(II)-K2.Cs-137 comparison was 11 100 (100) d [3].

Table 2. Standardization methods of the participants for ^{137}Cs

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
PTB	Pressurized IC 4P-IC-GR-00-00-00 calibrated by $4\pi\beta\text{-}\gamma$ coinc. with efficiency tracing 4P-PC-BP-NA-GR-CT and by $4\pi(\text{PC})\beta$ 4P-PC-BP-00-00-00	–	2 043 2 039	76-10-01 0 h UT	0.16	0.30
	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with ^{134}Cs and ^{60}Co efficiency tracers 4P-PC-BP-NA-GR-CT	–	3 902 3 906	78-10-01 12 h UT	0.07	0.25
	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1982 by $4\pi(\text{PC})\beta\text{-}\gamma$ and $4\pi(\text{PPC})\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT 4P-PP-BP-NA-GR-CT	–	19 734	98-01-01 0 h UT	0.05	0.36
CMI-IIR	4 $\pi\beta$ 4P-PC-BP-00-00-00	30.1 a	15 264	76-12-15 11 h UT	0.15	0.35
		29.9 a	4 474	78-10-24 11 h UT	0.20	0.43
			4 212	79-10-24 11 h UT	0.20	0.43
NPL	Pressurized IC* 4P-IC-GR-00-00-00	–	566 554	77-06-01 0 h UT	0.05	1.92
OMH	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	29.901 (45) a [6]	3 426 3 425	77-04-30 12 h UT	0.2	0.8
	$4\pi\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	[5]	1 891	97-10-01 0 h UT	0.06	0.35

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Table 2 continued. Standardization methods of the participants for ^{137}Cs

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
ANSTO	Pressurized IC 4P-IC-GR-00-00-00	–	1 362	77-05-01 0 h UT	0.3	2.0
	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1982 $4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	–	4 483	94-05-18 23 h UT	0.05	1.33
AECL	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	–	2 634 2 538	77-08-16 17 h UT	0.16	0.16
CSIR-NML	$4\pi(\text{LS})\beta\text{-}\gamma$ coincidence with ^{60}Co efficiency tracer [7] 4P-LS-BP-NA-GR-CT	–	66 760 60 690	80-05-21 10 h UT	0.04	0.99
BIPM ^a	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. / selective sampling, with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	11 100 (100) d	2 219.3 2 190.8 2 189.9	82-05-01 0 h UT	0.44	
NIST	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1982 by	–	2 541	82-05-01 0 h UT	0.12	0.31
	$4\pi(\text{PPC})\beta\text{-}\gamma$ anticoinc. with ^{134}Cs effic. tracer 4P-PP-BP-NA-GR-AT	30.07 (3) a	1 190	01-11-15 12 h UT	0.05	0.34
BNM-LNHB	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1982 by $4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-GL-GR-CT	–	1 047 1 044	85-06-15 12 h UT	0.03	0.27
	$4\pi(\text{PC})\beta\text{-}\gamma$ coincidence with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT and LS counting 4P-LS-BP-00-00-00	30.037 (28) a	3 187	98-04-06 12 h UT	0.03	0.30

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Table 2 continued. Standardization methods of the participants for ^{137}Cs

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
P3KRBiN	$4\pi\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	–	2 462 2 629	89-08-01 15 h UT	0.07	–
CNEA	$4\pi(\text{PC})\beta$ with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT and calibrated HPGe UA-GH-GR-00-00-00	–	1 465	92-01-01 0 h UT	0.19	0.60
NMIJ	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with ^{60}Co efficiency tracer 4P-PC-BP-NA-GR-CT	–	4 133	94-12-01 12 h UT	0.10	0.40
IRA	Pressurized IC traceable to the SIR in 1982 4P-IC-GR-00-00-00	–	2 365	96-09-01 0 h UT	0.02	0.16
		30.04 (3) a	2 159	00-12-01 12 h UT	0.03	0.17
BARC	$4\pi\beta\text{-}\gamma$ coinc. with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	–	545	97-01-01 6 h 30 UT	0.5	0.6
BEV	Pressurized IC* 4P-IC-GR-00-00-00	[5]	8 164	98-10-01 0 h UT	0.09	0.71
NIM	Pressurized IC 4P-IC-GR-00-00-00 calibrated by $4\pi\beta\text{-}\gamma$ coincidence with ^{134}Cs efficiency tracer 4P-PC-BP-NA-GR-CT	–	5 504 5 562	98-11-23 0 h UT	0.3	0.5

* traceable to primary measurements of ^{137}Cs at the NPL

^a the three ampoules measured by the BIPM for the CCRI(II)-K2.Cs-137 and measured in the SIR are used to make the link for the CCRI(II) key comparison.

Details regarding the solution submitted are shown in Table 3, including any impurities, when present, as identified by the laboratories. Recently the BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [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.

Table 3. Details of the solution of ¹³⁷Cs submitted

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. / (μg g ⁻¹)	Density / (g cm ⁻³)	Relative activity of any impurities [†]
PTB	CsCl in HCl	0.1	CsCl : 50	–	¹³⁴ Cs : 0.02 %
	CsNO ₃ in HNO ₃	0.1	CsNO ₃ : 20	1.0001	¹³⁴ Cs : 0.35 (2) %
	CsCl in HCl	0.1	CsCl : 50	1.00	–
CMI-IIR	CsCl in HCl	0.01	CsCl : 20	–	< 0.03 %
		0.08	CsCl : 20	–	¹³⁴ Cs : 0.16 (2) %
		–	–	–	¹³⁴ Cs : 0.040 (8) %
NPL	CsCl in HCl	0.1	CsCl: 100	1.001	¹³⁴ Cs < 0.01 %
OMH	Cs in HCl	0.1	Cs : 10	–	⁶⁰ Co : 0.003 (1) % ¹³⁴ Cs : 0.021 (5) %
	CsCl in HCl	0.1	CsCl : 25	–	¹³⁴ Cs : 0.005 (1) %
ANSTO	CsCl in HCl	0.1	–	1.00	¹³⁴ Cs : 0.09 (2) %
		2	–	–	–
AECL	CsCl in HCl	0.3	Cs ⁺ : 10	1	¹³⁴ Cs: 0.03 (1) %
CSIR-NML	CsCl in HCl	1.0	Cs ⁺ : 789	1.037	–
BIPM [#]	CsCl in HCl	0.2	CsCl : 20	–	¹³⁴ Cs : 0.004 %
NIST	CsCl in HCl	1	CsCl : 19	1.015	¹³⁴ Cs : 0.00106 (53) %
		1.0	CsCl: 30	1.015 (1)	–
BNM-LNHB	CsCl and BaCl ₂ in HCl	0.1	CsCl : 10 BaCl ₂ : 10	0.999	–
	CsCl in HCl	0.1	CsCl: 10	1.0001	–
P3KRBiN	CsCl in HCl	2.7	CsCl : 28	1.0920 (3)	–
CNEA	CsCl in HCl	0.1	CsCl: 50	1.000	¹³⁴ Cs : 0.10 (5) %
NMIJ	CsCl in HCl	0.1	CsCl : 50	1.00	–
IRA	CsCl in HCl	1	CsCl : 30	–	⁶⁰ Co < 3.3 × 10 ⁻⁴ %
				1.015	–
BARC	CsNO ₃ and Ba(NO ₃) ₂ in HNO ₃	0.01	CsNO ₃ and Ba(NO ₃) ₂ : 37	–	–

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Table 3 continued. Details of the solution of ^{137}Cs submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. ($\mu\text{g g}^{-1}$)	Density (g cm^{-3})	Relative activity of any impurities [†]
BEV	CoCl ₂ in HCl	0.1	CoCl ₂ : 50	1.0	–
NIM	Cs and Ba in HCl	0.1	Cs and Ba: 50	1.008	–

[†] the ratio of the activity of the impurity to the activity of ^{137}Cs at the reference date

[#] the solution used for the 1982 CCRI(II)-K2.Cs-137 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 ^{137}Cs arise from thirty-four ampoules and the SIR equivalent activity for each ampoule is given in Table 4a. The dates of measurement in the SIR are given in Table 1 and are used in the KCDB and all references in this report. 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].

The SIR corrections for impurities are generally small and the largest values are 1.0093, 1.0042, 1.0027 and 1.0024, for ASMW, CMI-IIR (1978), CNEA and ANSTO (1994), respectively.

The CNEA ampoule was measured again at the BIPM after a period of eleven years when the ^{134}Cs impurity had almost completely decayed, and this produced the same comparison result for the CNEA. This measurement confirms the validity of the impurity correction in 1992.

Apart from one earlier submission that was withdrawn, the results of each NMI in Table 1a are eligible for Appendix B of the MRA except that of the AECL, as this is not a designated laboratory of the NRC and of the P3KRBiN as Indonesia has not signed the MRA. However, three of the SIR results, for the CMI-IIR, CSIR-NML and the NPL have actually been superseded by the international comparison that was held in 1982. This comparison also enables the linking of another three NMIs, the IFIN, NRC and the SCK-CEN, and an international laboratory, the IRMM, to the SIR key comparison.

The results of the international comparison CCRI(II)-K2.Cs-137 have been published [3]. The seven laboratories to be added to the matrix of degrees of equivalence from this previous 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 the three ampoules standardized by $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence / selective sampling measurements with a ^{134}Cs efficiency tracer at the BIPM for the international comparison. The link is made using a ratio deduced from the BIPM line of Table 4a:

$$A_{ei} = (A/m)_i \times \frac{1}{3} \sum_{L=1}^3 (A_{e,BIPM,L} / (A/m)_{BIPM,L}) = (A/m)_i \times 45.508 \quad (a)$$

The results of the links for the seven laboratories are given in Table 4b. The uncertainties for the international comparison linked to the SIR are comprised of the original uncertainties together with the uncertainty in the link, 6×10^{-4} , given by the uncertainty of the SIR measurement of the BIPM ampoules of the CCRI(II)-K2.Cs-137 comparison. It is of note that the standard uncertainty of the unweighted mean of the 18 primary results of this comparison was 6.6×10^{-3} .

The BNM-LNHB (1985), PTB (1997), ANSTO (1994) and the NIST (1983 and 2001) SIR submissions are traceable to the measurements that these NMIs carried out in the frame of the comparison CCRI(II)-K2.Cs-137. In consequence, these SIR results may be compared with the corresponding results of the comparison in reference [3] through the link defined by equation (a). The agreement is within one standard uncertainty for the ANSTO, BNM-LNHB and the NIST (1983), and within two standard uncertainties for the PTB (1997) and the NIST (2001). This indicates the robustness of the KCRV determined through the SIR results.

The results for the CMI-IIR, the CSIR-NML and the NPL that are linked through the international comparison agree within the combined standard uncertainty with their original SIR equivalent activity values. However, as the CCRI comparison is more recent for these NMIs, the earlier SIR results have been superseded by the values in Table 4b for the results presented in the KCDB.

Table 4a. Results of SIR measurements of ^{137}Cs

NMI	Mass of solution / g	Activity submitted / kBq	N° of Ra source used	SIR A_e / kBq	Relative uncertainty from SIR	Total uncertainty $u_{c,i}$ / kBq
PTB	3.618 3 (2)	2 043	3	27 289	6×10^{-4}	94
	3.611 6 (2)	2 039		27 286		94
	3.644 22	3 902	3	27 914	8×10^{-4}	76
	3.647 83	3 906		27 930 †		76
3.657 33	19 734	5	27 600	5×10^{-4}	100	
CMI-IIR	0.957 64 *	15 264	4	27 490	5×10^{-4}	110
	3.590 34	4 474	3	27 730	8×10^{-4}	130
	3.592 60	4 212	3	27 530 #	7×10^{-4}	130 #
NPL †	3.617 8	566	2	27 307	9×10^{-4}	530
	3.543 7	554		27 269 #		520 #
OMH	3.604 3	3 426	3	27 386	6×10^{-4}	230
	3.603 2	3 425		27 395		230
	3.624 6	1 891	3	27 628	7×10^{-4}	99

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Table 4a continued. Results of SIR measurements of ¹³⁷Cs

NMI	Mass of solution / g	Activity submitted / kBq	N° of Ra source used	SIR A_e / kBq	Relative uncertainty from SIR	Total uncertainty $u_{e,i}$ / kBq
ANSTO	3.593 0	1 362	3	27 460	9×10^{-4}	540
	3.711	4 483	3	27 470	6×10^{-4}	370
AECL †	0.526 80	2 634	3	27 596	8×10^{-4}	66
	0.507 58 **	2 538		27 583	7×10^{-4}	65
CSIR-NML †	3.601	66 760	5	27 658	5×10^{-4}	280
	3.603 ***	60 690		27 653 #		280 #
BIPM	3.657 94	2 219.3	3	27 613	6×10^{-4}	123
	3.611 08	2 190.8		27 601		123
	3.609 48	2 189.9		27 615		123
NIST	3.615 39	2 541	3	27 577	6×10^{-4}	93
	3.649 1 (2)	1 190	2	27 634	8×10^{-4}	98
BNM-LNHB	3.561 31	1 047	2	27 561	9×10^{-4}	79
	3.571 45	1 044		27 500	8×10^{-4}	78
	3.595	3 187	3	27 514	6×10^{-4}	84
P3KRBiN	3.556	2 462	3	27 966	6×10^{-4}	26
	3.797	2 629		27 962		25
CNEA	2.029 65	1 465	3	27 450	15×10^{-4}	180
NMIJ	3.612 2	4 133	3	27 750	6×10^{-4}	120
IRA	3.591 1 (1)	2 365	3	27 552	6×10^{-4}	47
	3.626 5 (1)	2 159	3	27 458	6×10^{-4}	50
BARC	3.642 2	545	2	27 380	9×10^{-4}	220
BEV	3.611	8 164	4	27 320	5×10^{-4}	200
NIM †	3.616 48	5 504	4	27 255	5×10^{-4}	170
	3.654 61	5 562		27 270		170

† 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 1982

* mass measured at the BIPM after transfer into a NBS/BIPM ampoule.

** mass of standardized solution before dilution

*** mass of standardized solution before dilution: 1.100 95 g and 1.000 81 g respectively.

Table 4b. Results of 1982 CCRI(II) measurements of ¹³⁷Cs linked to the SIR

NMI	Activity * concentration (A/m) _i / (kBq g ⁻¹)	Relative standard uncertainty <i>u_i</i> × 10 ²	Linked SIR <i>A_{ei}</i> / kBq	<i>u(A_{ei})</i> / kBq
CMI-IIR [#]	607.5	0.27	27 646	76
CSIR-NML [#]	600.5	0.86	27 330	240
IFIN	602.1	0.59	27 400	160
IRMM	604.5	0.30	27 510	84
NPL [#]	599.2 [†]	0.70 [†]	27 270	190
NRC	609.3	0.16	27 728	47
SCK-CEN	604.8 [†]	0.18 [†]	27 523	52

* results from [3] with reference date 1982-05-01 00 h UT

[†] weighted mean of two different methods

[#] also measured in the SIR prior to this comparison.

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, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;
- each NMI 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 excluded from the KCRV, if necessary 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 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.

Consequently, the KCRV for ¹³⁷Cs has been identified as 27 549 (44) kBq using the results in Table 4a from the ASMW, AECL, CSIR-NML, NIST (1983), NPL, CMI-IIR (1980), CNEA, ANSTO (1994), NMIJ, BARC (1997), OMH (1997), PTB (1997), BNM-LNHB (1998), NIM (1999) and the BIPM.

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 (see Appendix 1).

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

$$u_{D_{ij}}^2 = u_i^2 + u_j^2 - \sum_k (f_k u_{k,\text{corr}})_i^2 - \sum_k (f_k u_{k,\text{corr}})_j^2 \quad (4)$$

and any obvious correlations between the NMIs (such as a traceable calibration) are subtracted 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. The core of the matrix is based on twelve values from the SIR. The additional matrix cells show the seven results from the 1982 international (CCRI(II)) comparison linked to those of the SIR comprising the three NMIs for which the SIR results have been superseded and the ten additional laboratories given in Table 4b. 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 (identified as x_R in the KCDB), is shown in Figure 1. 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 ^{137}Cs , BIPM.RI(II)-K1.Cs-137 currently comprises twelve results. These have been analysed with respect to the 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 six other NMIs and one international laboratory that took part in the CCRI(II)-K2.Cs-137 comparison in 1982 have been linked to the BIPM ongoing key comparison through three 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).

Other results may be added as and when NMIs contribute ^{137}Cs 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 ^{137}Cs

Key comparison BIPM.RI(II)-K1.Cs-137

MEASURAND : Equivalent activity of ^{137}Cs

Key comparison reference value: the SIR reference value for this radionuclide is $x_R = 27.549 \text{ MBq}$ with a standard uncertainty, $u_R = 0.044 \text{ MBq}$ (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.Cs-137 (1982) to BIPM.RI(II)-K1.Cs-137

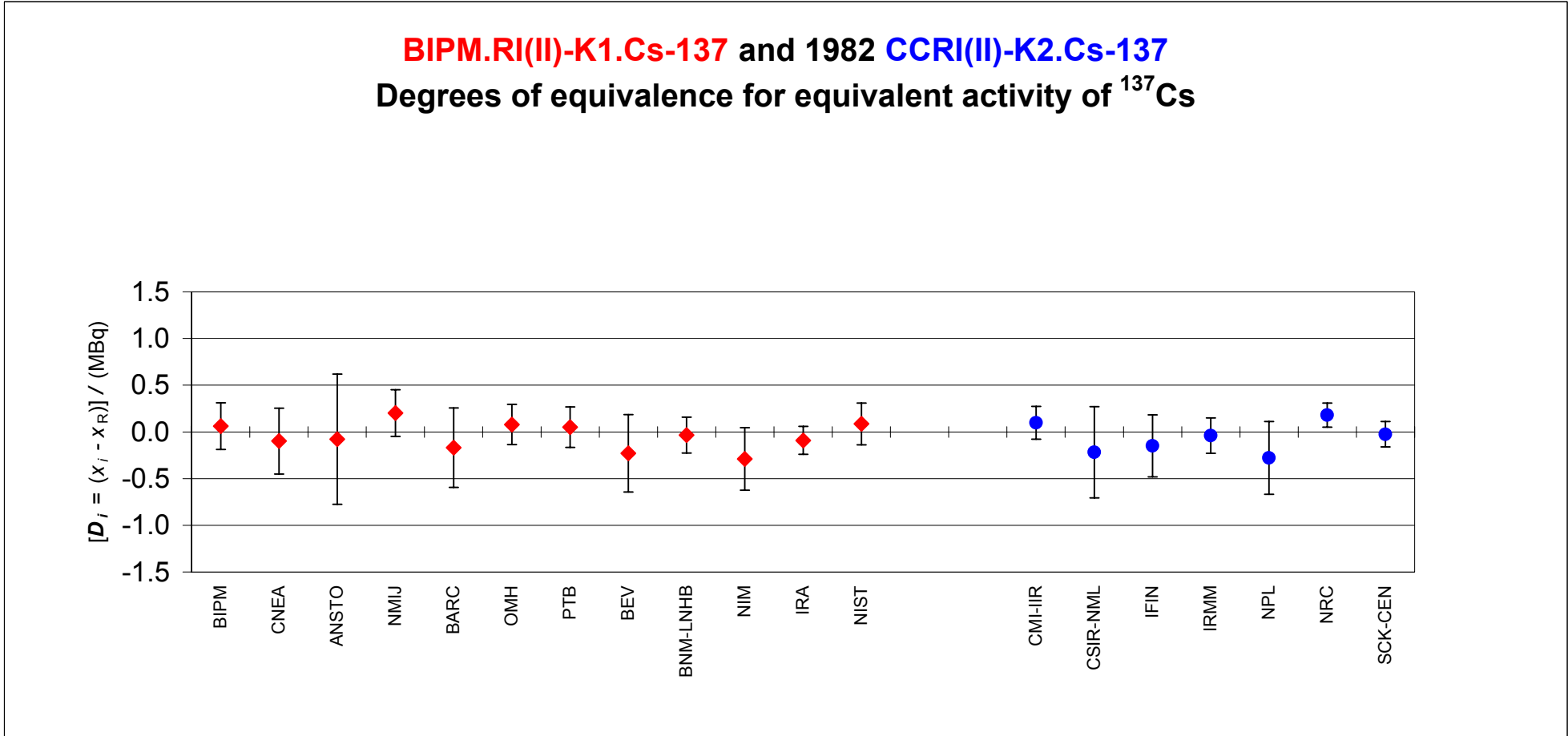
The value x_i is the equivalent activity for laboratory i participant in CCRI(II)-K2.Cs-137 having been normalized to the value of the BIPM as the linking laboratory (see Final report)

The degree of equivalence of laboratory i participant in CCRI(II)-K2.Cs-137 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.Cs-137 and one in CCRI(II)-K2.Cs-137, or both participant in CCRI(II)-K2.Cs-137, 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.Cs-137 matrices of equivalence to the other participants in CCRI(II)-K2.Cs-137.

Figure 1. Graph of degrees of equivalence with the KCRV for ^{137}Cs
 (as it appears in Appendix B of the MRA)



Appendix 1. Uncertainty budgets for the activity of ^{137}Cs submitted to the SIR**Uncertainty budget for the IRA (2000)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
Current of the first long lived reference source $\nu = 6$ *	1.8	–
Current of the second long lived reference source; $\nu = 15$	1.9	–
Current of the ampoule of ^{137}Cs under measurement; $\nu = 5$	1.9	–
Substitution factor of the reference source	–	4
Equivalent activity (calibration for ^{137}Cs)	–	16
Decay correction for the reference sources	–	2
Decay correction for ^{137}Cs	–	«0.1
Gravimetric measurements	–	0.3
Background	–	0.1
Quadratic summation	2.7 †	16.6
Relative combined standard uncertainty, u_c	17	

* number of degrees of freedom

† quadratic sum of 1.9 and 1.9 only.

Uncertainty budget for the NIST (2001)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
PIC A* net response per gram of solution SRM4233E, measured relative to RRS20	3	–
PIC A net response per Bq of ^{137}Cs in solution, measured relative to RRS20	4	–
Activity used to calibrate PIC A net response per Bq of ^{137}Cs in solution	–	32
Half-life of ^{137}Cs	«0.1	–
Half-life of ^{226}Ra	0.4	
Gravimetric measurements	–	5
Live time	–	5
PIC A charge collection	–	5
Source positioning	–	5
Photon emitting impurities	–	5
Quadratic summation	5	34
Relative combined standard uncertainty, u_c	34	

* PIC A, pressurized ionization chamber A

Appendix 2. Evaluation of the uncertainty of the degree of equivalence

Table 5 indicates for each laboratory the degree of equivalence D_i with its associated uncertainty U_i . This appendix presents the procedure used to evaluate these uncertainties.

The degree of equivalence of one laboratory is defined as the difference between the individual value of the equivalent activity A_{ei} for an NMI i and a suitable reference value which has been evaluated by the KCDB Working Group and the expanded uncertainty of this difference. Currently, the reference value, KCRV, for a given radionuclide is calculated as the arithmetic mean value of the SIR experimental entries for this radionuclide. Briefly at least four situations can occur depending on the consistency of the experimental SIR data sets :

1. All data are consistent and contribute to the reference value; this is the general case;
2. The value obtained by a laboratory that no longer exists, is used as long as it fits the usual quality criteria; it is taken into account when evaluating the reference value but does not appear in the matrices of results;
3. A value, that has been identified for example as an outlier, is not taken into account for the evaluation of the reference value but, nevertheless, the corresponding laboratory appears in the matrices of results.

The situation where a laboratory that no longer exists but contributes to the reference value and where an outlier has been identified in the data set can occur. This is a combination of both situation 2) and situation 3). The results, deduced from these two preceding cases, are also presented here, case 4.

In the following, the expression of the uncertainty for these four cases is considered on the assumption that the uncertainties of the different equivalent activities A_{ei} are not correlated. For the sake of coherence with the definition of the variables used in the text, the following notation is used :

$x_i = A_{ei}$ and $u_i = u_{A_{ei}}$ its uncertainty.

Case 1. All n laboratories contribute to the reference value, and appear in Table 5. In this case obviously we have

$$x_{\text{ref}} = \bar{x} = \frac{\sum_{j=1}^n x_j}{n} \quad (\text{A-1})$$

$$D_i = x_i - x_{\text{ref}} \quad (\text{A-2})$$

$$D_i = x_i - \frac{\sum_{j=1}^n x_j}{n} = x_i \left(1 - \frac{1}{n}\right) - \frac{\sum_{j \neq i} x_j}{n} \quad (\text{A-3})$$

At this stage the uncertainty of D_i has to be calculated. Applying the method of Gauß for the propagation of the uncertainties it is necessary to calculate the partial derivatives of D_i with respect to the x_i .

$$\text{So } \frac{\partial D_i}{\partial x_i} = \left(1 - \frac{1}{n}\right), \text{ and} \quad (\text{A-4})$$

$$\frac{\partial D_i}{\partial x_j} = -\frac{1}{n}, (j \neq i). \quad (\text{A-5})$$

Then the total combined uncertainty becomes

$$u_{c_i}^2 = \left(\frac{\partial D_i}{\partial x_i}\right)^2 u_i^2 + \sum_{j \neq i} \left(\frac{\partial D_i}{\partial x_j}\right)^2 u_j^2 \quad (\text{A-6})$$

$$= \left(1 - \frac{1}{n}\right)^2 u_i^2 + \frac{1}{n^2} \sum_{j \neq i} u_j^2 \quad (\text{A-7})$$

or, after recombination

$$= \left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2} \sum_{j=1}^n u_j^2. \quad (\text{A-8})$$

When a coverage factor of 2 is used (A-8) becomes

$$U_i^2 = 2^2 \left[\left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2} \sum_{j=1}^n u_j^2 \right]. \quad (\text{A-9})$$

Case 2. A laboratory was used to evaluate the reference value but does not appear in Table 5.

Let us assign the subscript n to the additional laboratory that contributes to the reference value. The uncertainty of this laboratory will appear only in the second part of equation (A-9). Accordingly, equation (A-9) becomes

$$U_i^2 = 2^2 \left[\left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2} \left(\sum_{j=1}^n u_j^2\right) \right], \text{ for } i = 1, n - 1. \quad (\text{A} - 10)$$

Case 3. The reference value was evaluated with all reported values except one.

For the sake of simplicity let us assign the subscript $n + 1$ to the ineligible laboratory so that the subscript for the other laboratories will run from 1 to n . Under this assumption the treatment of the ineligible laboratory will be slightly different and two formulae are deduced.

The ineligible laboratory does not contribute to the reference value, so the term $(1 - 2/n)$ in (A-9) reduces to 1 and the uncertainty is simply given by

$$U_{n+1}^2 = 2^2 \left[u_{n+1}^2 + \frac{1}{n^2} \sum_{j=1}^n u_j^2 \right]. \quad (\text{A} - 11)$$

In the evaluation of the uncertainty related to the n other laboratories the contribution from laboratory $n + 1$ disappears totally and the uncertainty remains given by the expression (A-10) without restriction over the subscript range i. e.

$$U_i^2 = 2^2 \left[\left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2} \sum_{j=1}^n u_j^2 \right]. \quad (\text{A} - 12)$$

Case 4. A laboratory that no longer exists contributes to the reference value and an outlier has been identified for another laboratory.

Let us assign the subscript n to the defunct existing laboratory so that the expression for the mean (A-1) remains applicable. In addition the outlier will be labelled by $n + 1$. For the $(n - 1)$ first laboratories which contribute to the mean value and appear in Table 5 the uncertainty of D_i is given by

$$U_i^2 = 2^2 \left[\left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2} \sum_{j=1}^n u_j^2 \right], \text{ for } i = 1, n - 1. \quad (\text{A} - 13)$$

For the laboratory $n + 1$ that is ineligible for the KCRV, its coefficient $(1 - 2/n)$ in (A-13) reduces to 1 and the expression of the uncertainty in Table 5 becomes

$$U_{n+1}^2 = 2^2 \left[u_{n+1}^2 + \frac{1}{n^2} \sum_{j=1}^n u_j^2 \right], \quad (\text{A} - 14)$$

similar to (A-11).

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	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
bremsstrahlung	BS	coincidence	CO
gamma ray	GR	anti-coincidence	AC
X - rays	XR	coincidence counting with efficiency tracing	CT
alpha - particle	AP	anti-coincidence counting with efficiency tracing	AT
mixture of various radiation e.g. X and gamma	MX	triple-to-double coincidence ratio counting	TD
		selective sampling	SS

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-00-00-00
calibrated IC		4P-IC-GR-00-00-00
internal gas counting		4P-PC-BP-00-00-IG