

**Activity measurements of the radionuclide  $^{137}\text{Cs}$**   
**for the NMIJ, Japan and the IRMM, Geel**  
**in the ongoing comparison BIPM.RI(II)-K1.Cs-137 and update of the KCRV**

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### **Abstract**

In 2004, the National Metrology Institute of Japan (NMIJ) and the Institute for Reference Materials and Measurements (IRMM, Geel) each submitted one sample of known activity of  $^{137}\text{Cs}$  to the International Reference System (SIR). The NMIJ submission replaces their 1994 SIR measurement and the IRMM is their first measurement in the SIR. The values of the activity submitted were about 2.7 MBq and 0.98 MBq. The key comparison reference value (KCRV) and the degrees of equivalence have been recalculated to include these latest values. The results are given in the form of a matrix that now contains 13 results, comparison identifier BIPM.RI(II)-K1.Cs-137, to which the five remaining eligible results from the CCRI(II)-K2.Cs-137 held in 1982 are linked.

### **1. Introduction**

The SIR for activity measurements of  $\gamma$ -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  $^{226}\text{Ra}$  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 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 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, ten 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 two NMIs are linked to the BIPM key comparison through this CCRI(II) comparison [4].

## 2. Participants

In addition to the NMIJ and the IRMM, fourteen NMIs and two other laboratories have submitted ampoules for the comparison of  $^{137}\text{Cs}$  activity measurements since 1976. The laboratory details are given in Table 1 while the details of the participants in CCRI(II)-K2.Cs-137 are given in [4].

**Table 1. Details of the participants in the comparison 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
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

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

<b>Original acronym</b>	<b>NMI</b>	<b>Full name</b>	<b>Country</b>	<b>Regional metrology organization</b>	<b>Date of measurement at the BIPM</b>
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1983-07-19 2001-11-28
LMRI LPRI	LNE- LNHB	Laboratoire national de métrologie et d'essais – 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 2005-04-28
–	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
–	IRMM	Institute for Reference Materials and Measurements	European Union	EUROMET	2004-01-20

\* 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 method for the laboratory, the activities submitted and the relative standard uncertainties ( $k = 1$ ) are given in Table 2. The uncertainty budgets for the latest participants are given in Appendix 1; the earlier contributions are in [4].

The half-life used in the SIR for these two recent submissions is 11 020 (60) d as recommended by the IAEA [5], while 11 020.8 (1.2) d [6] was used for all the previous submissions [4].

**Table 2. Standardization methods of the participants for  $^{137}\text{Cs}$**

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 $^{134}\text{Cs}$ and $^{60}\text{Co}$ 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 $^{134}\text{Cs}$ 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	77-06-01	0.05	1.92
			554	0 h UT		

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**Table 2 continued. Standardization methods of the participants for  $^{137}\text{Cs}$** 

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
OMH	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with $^{134}\text{Cs}$ efficiency tracer 4P-PC-BP-NA-GR-CT	29.901 (45) a [7]	3 426 3 425	77-04-30 12 h UT	0.2	0.8
	$4\pi\beta\text{-}\gamma$ coinc. with $^{134}\text{Cs}$ efficiency tracer 4P-PC-BP-NA-GR-CT	[5]	1 891	97-10-01 0 h UT	0.06	0.35
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 $^{134}\text{Cs}$ 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 $^{134}\text{Cs}$ 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 $^{60}\text{Co}$ efficiency tracer [8] 4P-LS-BP-NA-GR-CT	–	66 760 60 690	80-05-21 10 h UT	0.04	0.99
BIPM <sup>a</sup>	$4\pi(\text{PC})\beta\text{-}\gamma$ coinc. / selective sampling, with $^{134}\text{Cs}$ 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 $^{134}\text{Cs}$ effic. tracer 4P-PP-BP-NA-GR-AT	30.07 (3) a	1 190	01-11-15 12 h UT	0.05	0.34

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**Table 2 continued. Standardization methods of the participants for  $^{137}\text{Cs}$** 

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
LNE-LNHB	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1982 by $4\pi(\text{PC})\beta\text{-}\gamma$ coinc. with $^{134}\text{Cs}$ 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 $^{134}\text{Cs}$ 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
P3KRBiN	$4\pi\beta\text{-}\gamma$ coinc. with $^{134}\text{Cs}$ 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 $^{134}\text{Cs}$ 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 $^{60}\text{Co}$ efficiency tracer 4P-PC-BP-NA-GR-CT	–	4 133	94-12-01 12 h UT	0.10	0.40
	$4\pi\beta\text{-}\gamma$ coinc. with $^{134}\text{Cs}$ efficiency tracer 4P-PC-BP-NA-GR-CT	10 971.8	2 687	05-02-15 0 h UTC	0.4	0.3
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 $^{134}\text{Cs}$ 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

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**Table 2 continued. Standardization methods of the participants for  $^{137}\text{Cs}$** 

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
NIM	Pressurized IC 4P-IC-GR-00-00-00 calibrated by $4\pi\beta\text{-}\gamma$ coincidence with $^{134}\text{Cs}$ efficiency tracer 4P-PC-BP-NA-GR-CT	–	5 504 5 562	98-11-23 0 h UT	0.3	0.5
IRMM	4 $\pi$ (PPC)– $\gamma$ (NaI well) coinc. with $^{134}\text{Cs}$ efficiency tracer 4P-PP-BP-NA-GR-CT	[5]	980.8	03-07-01 0 h UTC	0.5	0.4
	Liquid scintillation by CIEMAT/NIST 4P-LS-BP-00-00-CN		980.8		0.10	0.69

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

<sup>a</sup> the three ampoules measured by the BIPM for the CCRI(II)-K2.Cs-137 and measured in the SIR were used to make the link for the CCRI(II) key comparison.

Details regarding the NMIJ and IRMM solutions 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 [9]. The CCRI(II) agreed in 1999 [10] that this method should be followed according to the protocol described in [11] when an NMI makes such a request or when there appear to be discrepancies. No supplementary measurements were made at the BIPM for these latest submissions.

**Table 3. Details of the NMIJ and IRMM solution of  $^{137}\text{Cs}$  submitted**

NMI	Chemical composition	Solvent conc. / ( $\text{mol dm}^{-3}$ )	Carrier: conc. /( $\mu\text{g g}^{-1}$ )	Density /( $\text{g cm}^{-3}$ )	Relative activity of any impurities <sup>†</sup>
NMIJ	CsCl in HCl	0.1	CsCl : 50	1.00	–
			CsCl : 100	1.002	–
IRMM	CsCl in HCl	0.1	CsCl : 50	–	$^{134}\text{Cs}$ : 0.013 (1) %

<sup>†</sup> the ratio of the activity of the impurity to the activity of  $^{137}\text{Cs}$  at the reference date

#### 4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The activity measurements for  $^{137}\text{Cs}$  arise from thirty-six ampoules and the SIR equivalent activity,  $A_{ei}$ , 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 uncertainty arising from the measurements in the SIR is 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  $^{226}\text{Ra}$ , all the SIR results are normalized to the radium source number 5 [1].

The SIR correction for impurities is negligible for the IRMM submission and is lower than 1.01 in all other cases [4].

**Table 4a. Results of SIR measurements of  $^{137}\text{Cs}$**

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)	2 044	3	27 289	$6 \times 10^{-4}$	94
	3.611 6 (2)	2 040		27 286		94
	3.644 22	3 902	3	27 914	$8 \times 10^{-4}$	76
	3.647 83	3 906		27 930 †		76
	3.657 33	19 734	5	27 600	$5 \times 10^{-4}$	100
CMI-IIR	0.957 64 *	15 265	4	27 490	$5 \times 10^{-4}$	110
	3.590 34	4 474	3	27 730	$8 \times 10^{-4}$	130
	3.592 60	4 212	3	27 530 #	$7 \times 10^{-4}$	130 #
NPL †	3.617 8	566	2	27 307	$9 \times 10^{-4}$	530
	3.543 7	554		27 269 #		520 #
OMH	3.604 3	3 426	3	27 386	$6 \times 10^{-4}$	230
	3.603 2	3 425		27 395		230
	3.624 6	1 891	3	27 628	$7 \times 10^{-4}$	99
ANSTO	3.593 0	1 362	3	27 460	$9 \times 10^{-4}$	540
	3.711	4 483	3	27 470	$6 \times 10^{-4}$	370
AECL †	0.526 80	2 634	3	27 596	$8 \times 10^{-4}$	66
	0.507 58 **	2 538		27 583	$7 \times 10^{-4}$	65
CSIR-NML †	3.601	66 760	5	27 658	$5 \times 10^{-4}$	280
	3.603 ***	60 690		27 653 #		280 #

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**Table 4a continued. Results of SIR measurements of  $^{137}\text{Cs}$** 

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
BIPM	3.657 94	2 219.3	3	27 613	$6 \times 10^{-4}$	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	$6 \times 10^{-4}$	93
	3.649 1 (2)	1 190	2	27 634	$8 \times 10^{-4}$	98
LNE-LNHB	3.571 45	1 047	2	27 561	$9 \times 10^{-4}$	79
	3.561 31	1 044		27 500	$8 \times 10^{-4}$	78
	3.595	3 187	3	27 514	$6 \times 10^{-4}$	84
P3KRBiN	3.556	2 462	3	27 966	$6 \times 10^{-4}$	26
	3.797	2 629		27 962		25
CNEA	2.029 65	1 465	3	27 450	$15 \times 10^{-4}$	180
NMIJ	3.612 2	4 133	3	27 750	$6 \times 10^{-4}$	120
	3.621 22	2 687	3	27 720	$6 \times 10^{-4}$	140
IRA	3.591 1 (1)	2 365	3	27 552	$6 \times 10^{-4}$	47
	3.626 5 (1)	2 159	3	27 458	$6 \times 10^{-4}$	50
BARC	3.642 2	545	2	27 380	$9 \times 10^{-4}$	220
BEV	3.611	8 164	4	27 320	$5 \times 10^{-4}$	200
NIM <sup>†</sup>	3.616 48	5 504	4	27 255	$5 \times 10^{-4}$	170
	3.654 61	5 562		27 270		170
IRMM	3.632 55	980.8 <sup>§</sup>	2	27 340	$9 \times 10^{-4}$	160

<sup>†</sup> the mean of the  $A_e$  values is used with an averaged uncertainty, as attributed to an individual entry [12]

<sup>§</sup> the mean value and standard uncertainty of the two results submitted, as evaluated by the IRMM:  $A = 980.8$  kBq,  $u = 5.8$  kBq.

<sup>#</sup> 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.

The SIR correction for impurities is negligible for the IRMM submission and is lower than 1.01 in all other cases [4].

The results of the international comparison CCRI(II)-K2.Cs-137 have been published [3] and seven of them were 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  $^{134}\text{Cs}$  efficiency tracer at the BIPM for the international comparison [4]. The result for the IRMM that was linked through the international comparison is given in Table 4b. The value agrees within the combined standard uncertainty with their recently submitted SIR equivalent activity value and is now superseded by the value in Table 4a for the results presented in the KCDB.

**Table 4b. Result of the 1982 CCRI(II) IRMM measurement of  $^{137}\text{Cs}$  linked to the SIR**

NMI	Activity concentration* ( $A/m$ ) <sub><i>i</i></sub> / (kBq g <sup>-1</sup> )	Relative standard uncertainty $u_i \times 10^2$	Linked SIR $A_{ei}$ / kBq	$u(A_{ei})$ / kBq
IRMM	604.5	0.30	27 510	84

\* reference date 1982-05-01 00 h UT

Apart from one earlier submission that was withdrawn, the results of each NMI in Table 1 are eligible for entry to Appendix B of the MRA except that of the AECL and the P3KRBiN, as these are not designated laboratories of the NRC, Canada and of the KIM-LIPI, Indonesia, respectively. 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 latter comparison also enables the linking of another two NMIs, the IFIN and the NRC to the SIR key 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.

The key comparison reference value for  $^{137}\text{Cs}$  given in [4] is 27 549 (44) kBq. At the CCRI(II) meeting in May 2005, the KCRV was modified to include the IRMM and NMIJ values, the latter replacing their earlier result of 1994. Consequently, the KCRV is now 27 534 (42) kBq [13] using the results in Table 4a from the ASMW, NPL, AECL, CMI-IIR (1980), CSIR-NML, BIPM, NIST (1983), CNEA, ANSTO (1994),

BARC (1997), OMH (1997), PTB (1997), LNE-LNHB (1998), NIM (1999), IRMM and the NMIJ (2005).

## 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 results are taken from Table 4a.

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 [14].

### 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 thirteen values from the SIR. The additional matrix cells show the six remaining results from the 1982 international (CCRI(II)) comparison linked to those of the SIR. 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. The black square symbols in the figure represent results that are more than 20 years old. The graph 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  $^{137}\text{Cs}$ , BIPM.RI(II)-K1.Cs-137 currently comprises thirteen 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 five other NMIs that took part in the CCRI(II)-K2.Cs-137 comparison in 1982 are 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  $^{137}\text{Cs}$  activity measurements to this comparison or take part in other linked comparisons.

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## References

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Table 5. Table of degrees of equivalence and introductory text for  $^{137}\text{Cs}$

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

MEASURAND : Equivalent activity of  $^{137}\text{Cs}$

Key comparison reference value: the SIR reference value for this radionuclide is  $x_R = 27.534 \text{ MBq}$  with a standard uncertainty,  $u_R = 0.042 \text{ MBq}$  (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  $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_i^2)^{1/2}$  is used with  $l$  being the linking laboratory when both laboratories are linked, and  $f$  is the correlation coefficient.

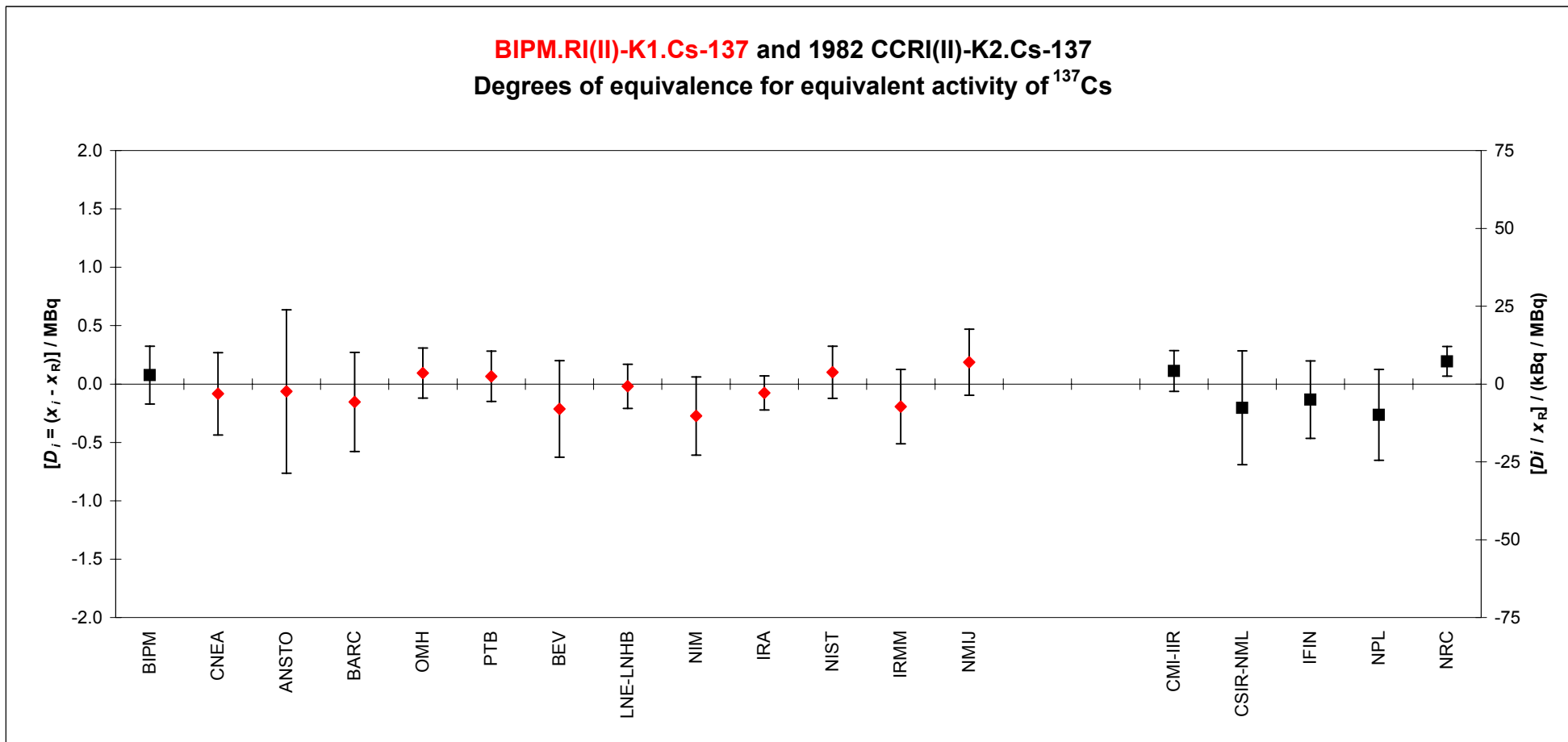
These statements make it possible to extend the BIPM.RI(II)-K1.Cs-137 matrices of equivalence to the other participants in CCRI(II)-K2.Cs-137

Table 5 continued

Lab <i>i</i> ↓	$D_i$ / MBq		$U_i$		Lab <i>j</i> →																		
	$D_{ij}$ / MBq	$U_{ij}$	$D_{ij}$ / MBq	$U_{ij}$	BIPM		CNEA		ANSTO		BARC		OMH		PTB		BEV		LNE-LNHB		NIM		
BIPM	0.08	0.25																					
CNEA	-0.08	0.35			-0.16	0.43			-0.02	0.82	0.07	0.57	-0.18	0.41	-0.15	0.41	0.13	0.54	-0.06	0.40	0.19	0.50	
ANSTO	-0.06	0.70			-0.14	0.78	0.02	0.82			0.09	0.86	-0.16	0.77	-0.13	0.77	0.15	0.84	-0.04	0.76	0.21	0.81	
BARC	-0.15	0.43			-0.23	0.50	-0.07	0.57	-0.09	0.86			-0.25	0.48	-0.22	0.48	0.06	0.59	-0.13	0.47	0.12	0.56	
OMH	0.09	0.21			0.02	0.31	0.18	0.41	0.16	0.77	0.25	0.48			0.03	0.28	0.31	0.45	0.11	0.26	0.37	0.39	
PTB	0.07	0.22			-0.01	0.31	0.15	0.41	0.13	0.77	0.22	0.48	-0.03	0.28			0.28	0.45	0.09	0.26	0.34	0.39	
BEV	-0.21	0.41			-0.29	0.47	-0.13	0.54	-0.15	0.84	-0.06	0.59	-0.31	0.45	-0.28	0.45			-0.19	0.43	0.06	0.52	
LNE-LNHB	-0.02	0.19			-0.10	0.29	0.06	0.40	0.04	0.76	0.13	0.47	-0.11	0.26	-0.09	0.26	0.19	0.43			0.25	0.38	
NIM	-0.27	0.34			-0.35	0.42	-0.19	0.50	-0.21	0.81	-0.12	0.56	-0.37	0.39	-0.34	0.39	-0.06	0.52	-0.25	0.38			
IRA	-0.08	0.15			-0.15	0.26	0.01	0.37	-0.01	0.75	0.08	0.45	-0.17	0.22	-0.14	0.22	0.14	0.41	-0.06	0.20	0.20	0.35	
NIST	0.10	0.22			0.02	0.31	0.18	0.41	0.16	0.77	0.25	0.48	0.01	0.28	0.03	0.28	0.31	0.45	0.12	0.26	0.37	0.39	
IRMM	-0.19	0.32			-0.27	0.40	-0.11	0.48	-0.13	0.81	-0.04	0.54	-0.29	0.38	-0.26	0.38	0.02	0.51	-0.17	0.36	0.08	0.47	
NMIJ	0.19	0.28			0.11	0.37	0.27	0.46	0.25	0.79	0.34	0.52	0.09	0.34	0.12	0.34	0.40	0.49	0.21	0.33	0.46	0.44	
CMI-IIR	0.11	0.17			0.04	0.28	0.20	0.39	0.18	0.76	0.27	0.47	0.02	0.25	0.05	0.25	0.33	0.43	0.13	0.23	0.39	0.37	
CSIR-NML	-0.20	0.49			-0.28	0.53	-0.12	0.60	-0.14	0.88	-0.05	0.65	-0.30	0.52	-0.27	0.52	0.01	0.62	-0.18	0.51	0.07	0.59	
IFIN	-0.13	0.33			-0.21	0.40	-0.05	0.48	-0.07	0.81	0.02	0.54	-0.23	0.38	-0.20	0.38	0.08	0.51	-0.11	0.36	0.14	0.47	
NPL	-0.26	0.39			-0.34	0.45	-0.18	0.52	-0.20	0.83	-0.11	0.58	-0.36	0.43	-0.33	0.43	-0.05	0.06	-0.24	0.42	0.01	0.51	
NRC	0.19	0.13			0.12	0.25	0.28	0.37	0.26	0.75	0.35	0.45	0.10	0.22	0.13	0.22	0.41	0.41	0.21	0.19	0.47	0.35	

Lab <i>i</i> ↓	$D_i$ / MBq		$U_i$		Lab <i>j</i> →																		
	$D_{ij}$ / MBq	$U_{ij}$	$D_{ij}$ / MBq	$U_{ij}$	IRA		NIST		IRMM		NMIJ		CMI-IIR		CSIR-NML		IFIN		NPL		NRC		
BIPM	0.08	0.25			0.15	0.26	-0.02	0.31	0.27	0.40	-0.11	0.37	-0.04	0.28	0.28	0.53	0.21	0.40	0.34	0.45	-0.12	0.25	
CNEA	-0.08	0.35			-0.01	0.37	-0.18	0.41	0.11	0.48	-0.27	0.46	-0.20	0.39	0.12	0.60	0.05	0.48	0.18	0.52	-0.28	0.37	
ANSTO	-0.06	0.70			0.01	0.75	-0.16	0.77	0.13	0.81	-0.25	0.79	-0.18	0.76	0.14	0.88	0.07	0.81	0.20	0.83	-0.26	0.75	
BARC	-0.15	0.43			-0.08	0.45	-0.25	0.48	0.04	0.54	-0.34	0.52	-0.27	0.47	0.05	0.65	-0.02	0.54	0.11	0.58	-0.35	0.45	
OMH	0.09	0.21			0.17	0.22	-0.01	0.28	0.29	0.38	-0.09	0.34	-0.02	0.25	0.30	0.52	0.23	0.38	0.36	0.43	-0.10	0.22	
PTB	0.07	0.22			0.14	0.22	-0.03	0.28	0.26	0.38	-0.12	0.34	-0.05	0.25	0.27	0.52	0.20	0.38	0.33	0.43	-0.13	0.22	
BEV	-0.21	0.41			-0.14	0.41	-0.31	0.45	-0.02	0.51	-0.40	0.49	-0.33	0.43	-0.01	0.62	-0.08	0.51	0.05	0.06	-0.41	0.41	
BNM-LNHB	-0.02	0.19			0.06	0.20	-0.12	0.26	0.17	0.36	-0.21	0.33	-0.13	0.23	0.18	0.51	0.11	0.36	0.24	0.42	-0.21	0.19	
NIM	-0.27	0.34			-0.20	0.35	-0.37	0.39	-0.08	0.47	-0.46	0.44	-0.39	0.37	-0.07	0.59	-0.14	0.47	-0.01	0.51	-0.47	0.35	
IRA	-0.08	0.15					-0.18	0.22	0.12	0.34	-0.26	0.30	-0.19	0.18	0.13	0.49	0.06	0.34	0.19	0.39	-0.27	0.14	
NIST	0.10	0.22			0.18	0.22			0.29	0.38	-0.09	0.34	-0.01	0.25	0.30	0.52	0.23	0.38	0.36	0.43	-0.09	0.22	
IRMM	-0.19	0.32			-0.12	0.34	-0.29	0.38			-0.38	0.43	-0.31	0.35	0.01	0.58	-0.06	0.45	0.07	0.50	-0.39	0.33	
NMIJ	0.19	0.28			0.26	0.30	0.09	0.34	0.38	0.43			0.07	0.32	0.39	0.56	0.32	0.43	0.45	0.47	-0.01	0.30	
CMI-IIR	0.11	0.17			0.19	0.18	0.01	0.25	0.31	0.35	-0.07	0.32			0.32	0.50	0.25	0.35	0.38	0.41	-0.08	0.17	
CSIR-NML	-0.20	0.49			-0.13	0.49	-0.30	0.52	-0.01	0.58	-0.39	0.56	-0.32	0.50			-0.07	0.58	0.06	0.61	-0.40	0.49	
IFIN	-0.13	0.33			-0.06	0.34	-0.23	0.38	0.06	0.45	-0.32	0.43	-0.25	0.35	0.07	0.58			0.13	0.49	-0.33	0.33	
NPL	-0.26	0.39			-0.19	0.39	-0.36	0.43	-0.07	0.50	-0.45	0.47	-0.38	0.41	-0.06	0.61	-0.13	0.49			-0.46	0.39	
NRC	0.19	0.13			0.27	0.14	0.09	0.22	0.39	0.33	0.01	0.30	0.08	0.17	0.40	0.49	0.33	0.33	0.46	0.39			

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



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

**Appendix 1. Uncertainty budgets for the activity of  $^{137}\text{Cs}$  submitted to the SIR****Uncertainty budget for the NMIJ (2004)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
<b>Contributions due to</b>		
Linear extrapolation including random component of weighing and counting	36	–
Dead time	–	1
Resolving time	–	2
Timing	–	2
Decay scheme (conversion electron emission rate)	–	25
Efficiency tracing with $^{134}\text{Cs}$	–	20
Decay correction	–	2
Gravimetric measurements	–	8
Background	–	3
<b>Quadratic summation</b>	<b>36</b>	<b>33</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>49</b>	

**Uncertainty budgets for the IRMM (2004)**

<b>Relative standard uncertainties for the method : 4P-PP-BP-NA-GR-CT</b>	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
<b>Contributions due to</b>		
Counting statistics	48	–
Beta dead time	–	2
Gamma dead time	–	1
Pile-up	–	2
Resolving time	–	2
Gandy effect	–	2
Counting time	–	5
Correction factor (1 + C)	–	25
Impurity	–	1.5
<sup>134</sup> Cs tracer activity	–	30
Decay correction for <sup>137</sup> Cs	–	1
Decay correction for <sup>134</sup> Cs	–	0.2
Gravimetric measurements	–	12
Background	5	–
<b>Quadratic summation</b>	<b>48</b>	<b>41</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>63</b>	

<b>Relative standard uncertainties for the method : 4P-LS-BP-00-00-CN</b>	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
<b>Contributions due to</b>		
Counting statistics	10	–
Dead time	–	10
Counting time	–	5
Wall effect	–	5
Instrument dependence	–	10
Sample stability	–	10
Interpolation from calibration curve	–	2
kB	–	3
Decay scheme parameters	–	63
Decay correction for <sup>137</sup> Cs	–	0.06
Gravimetric measurements	–	20
Background	1	–
Impurity	–	1.3
<b>Quadratic summation</b>	<b>10</b>	<b>69</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>70</b>	

## 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
$4\pi$	4P	proportional counter	PC
defined solid angle	SA	press. prop counter	PP
$2\pi$	2P	liquid scintillation counting	LS
undefined solid angle	UA	Nal(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
$4\pi$ (PC) $\beta$ - $\gamma$ -coincidence counting		4P-PC-BP-NA-GR-CO
$4\pi$ (PPC) $\beta$ - $\gamma$ -coincidence counting eff. trac.		4P-PP-MX-NA-GR-CT
defined solid angle $\alpha$ -particle counting with a PIPS detector		SA-PS-AP-00-00-00
$4\pi$ (PPC)AX- $\gamma$ (GeHP)-anticoincidence counting		4P-PP-MX-GH-GR-AC
$4\pi$ Csl- $\beta$ ,AX, $\gamma$ counting		4P-CS-MX-00-00-HE
calibrated IC		4P-IC-GR-00-00-00
internal gas counting		4P-PC-BP-00-00-IG