Update of the BIPM comparison BIPM.RI(II)-K1.Cs-134 of activity measurements of the radionuclide ¹³⁴Cs to include the 2014 result of the POLATOM (Poland), the 2014 result of the NRC (Canada), the 2015 result of the LNMRI-IRD (Brazil) and the 2016 result of the PTB (Germany)

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Abstract Since 1976, 23 laboratories have submitted 44 samples of ¹³⁴Cs to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM), with comparison identifier BIPM.RI(II)-K1.Cs-134. In the last few years, the POLATOM (Poland), the NRC (Canada), the LNMRI-IRD (Brazil) and the PTB (Germany) participated in the comparison and the key comparison reference value (KCRV) has been updated. The degrees of equivalence between each equivalent activity measured in the SIR or linked to the SIR from the APMP.RI(II)-K2.Cs-134 comparison and the updated KCRV have been calculated and the results are given in the form of a table. A graphical representation is also given.

1. Introduction

The SIR for activity measurements of γ -ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled with 3.6 g of the radioactive solution. For radioactive

1978-11-13

gases, a different standard ampoule is used. Each NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of 226 Ra using pressurized ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity $A_{\rm e}$, are all given in [1].

From its inception until 31 December 2021, the SIR has been used to measure 1033 ampoules to give 788 independent results for 72 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary standardizations. These comparisons are described as BIPM continuous comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the Comité International des Poids et Mesures Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Cs-134 key comparison. The results of earlier participations in this key comparison were published previously [3–6].

2. Participants

BIPM

Laboratory details are given in Table 1, with the earlier submissions being taken from [3–6]. The dates of measurement in the SIR given in Table 1 are used in the KCDB and all references in this report. The AECL (Atomic Energy of Canada Ltd) is not part of the NMI in Canada but 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).

NMI or	Previous	Full name	Country	RMO	Date of
labora-	acronyms				SIR mea- surement
\mathbf{tory}	or other				yyyy-mm-dd
	insti-				
	tutes				
AECL ^a	-	Atomic Energy of Canada	Canada	SIM	1977-05-23
		Ltd			
					1992-07-03
BARC	-	Bhabha Atomic Research	India	APMP	1981-09-03
		Centre			
					1996-11-22
					2005-07-20
BEV	IRK	Bundesamt fur Eich- und	Austria	EURAMET	2008-11-24
		Vermessungswesen			

Bureau International des

Poids et Mesures

Table 1: Details of the participants in the BIPM.RI(II)-K1.Cs-134.

... Continuation of Table 1.

NMI or labora- tory	Previous acronyms or other insti- tutes	Full name	Country	RMO	Date of SIR measurement yyyy-mm-dd
BKFH	OMH, MKEH	Government Office of the Capital City Budapest	Hungary	EURAMET	1979-01-26 1992-06-14 2004-12-08
CIEMAT	-	Centro de Investigaciones Energéticas, Medioambi- entales y Tecnologicas	Spain	EURAMET	2001-04-27
CMI	UVVVR, CMI-IIR	Czech Metrological Institute	Czechia	EURAMET	1978-04-17
CNEA	-	Comision Nacional de Energia Atomica	Argentina	SIM	1987-01-07 2005-09-12
IAEA	-	International Atomic Energy Agency			1978-05-26 1979-02-13
IFIN-HH	-	Institutul National de Cercetare - Dezvoltare in Fizica si Inginerie Nucleara- "Horia Hu- lubei"	Romania	EURAMET	2006-11-21
IRA	IER	Institut de Radiophysique	Switzerland	EURAMET	1978-02-24 2009-02-25
JRC	IRMM, CBNM	EC-JRC Institute for Reference Materials and Measurements	European Union	EURAMET	2004-01-20
KRISS	KSRI	Korea Research Institute of Standards and Science	Republic of Korea	APMP	1996-02-22
LNE- LNHB	LMRI, LPRI, BNM- LNHB	Université Paris-Saclay, CEA, List, Laboratoire National Henri Becquerel	France	EURAMET	1987-11-30 1998-10-06
					2005-04-13
LNMRI- IRD	IEA, IPEN ^b	Laboratorio Nacional de Metrologia das Radiações Ionizantes	Brazil	SIM	1987-10-19
					2015-12-15
NIST	NBS	National Institute of Standards and Technology	United States	SIM	1977-10-18
NMIJ	ETL	National Metrology Insti- tute of Japan	Japan	APMP	2002-06-21 2005-04-20
NMISA	NAC, CSIR- NML ^c	National Metrology Insti- tute of South Africa	South Africa	AFRIMETS	2010-08-05

NMI or labora- tory	Previous acronyms or other insti- tutes	Full name	Country	RMO	Date of SIR measurement yyyy-mm-dd
NPL	=	National Physical Labora-	United King-	EURAMET	1976-12-30
		tory	dom		
NRC	-	National Research Council	Canada	SIM	2014-03-18
POLATOM	IBJ, RC	National Centre for Nu-	Poland	EURAMET	2014-01-09
		clear Research Radioiso-			
		tope Centre POLATOM			
PTB	-	Physikalisch-Technische	Germany	EURAMET	1994-09-15
		Bundesanstalt			
					2004-07-05
					2016-08-17
PTKMR	PDS,	Pusat Teknologi Kesela-	Indonesia	APMP	1984-11-22
	PSPKR,	matan dan Metrologi Ra-			
	P3KRBiN	diasi			

... Continuation of Table 1.

3. NMI standardization methods

Each NMI that submits amoules to the SIR has measured the activity either by a primary standardization method or by using a secondary method, for example a calibrated ionization chamber. In the latter case, the traceability of the calibration needs to be clearly identified to ensure that appropriate correlations are taken into account.

A brief description of the standardization methods used by the laboratories, the activities submitted, the relative standard uncertainties and the half-life used by the participants are given in Table 2. The uncertainty budget for the new submission is given in Appendix D attached to this report; previous uncertainty budgets are given in the earlier K1 reports [3–6]. The list of acronyms used to summarize the methods is given in Appendix E.

Since 2014, the half-life used by the BIPM is 754.01(51) days as published in BIPM Monographie 5 vol. 7 [7]. The half-life of 753.1(18) days [8] was used for the earlier results.

^a federal Crown corporation, not part of the NMI in Canada (see text)

^b IEA, IPEN are other institutes of the country.

^c NAC is another institute in the country now named iThemba LABS.

Table 2: Standardization methods of the participants for $^{134}\mathrm{Cs}.$

NMI or labora- tory	Method used and the acronym	$egin{aligned} \mathbf{Activity} \ A_i/\mathbf{kBq} \end{aligned}$	Relative standard uncertainty /10 ⁻²		Reference date	Half-life /d	
			A	В	yyyy-mm- dd		
AECL	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	884.3 ^g	0.014	0.32	1977-03-18 17:00 UT	753.1(1.8)	
		901.4	0.014	0.32			
	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	699.2	0.15	0.02	1992-05-01 17:00 UT		
BARC	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	533.2	0.03	0.7	1981-08-01 06:30 UT	-	
	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	607.5	0.36	0.3	1996-08-01 06:30 UT		
	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	1259	0.4	0.2	2005-05-15 00:00 UT	[9]	
BEV	Calibrated pressurized ionization chamber (4P-IC-GR-00-00-00)	2299	0.08	0.69	2008-12-01 00:00 UT	754.0(7)	
BIPM	$4\pi(PC)\beta-\gamma$ coincidence (4P-PC-BP-NA-GR-CO)	3005.4 ^g	0.03	0.13	1978-10-15 00:00 UT	- [9]	
		3007.8	0.03	0.13			
BKFH	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	686.4 ^g	0.05	0.3	1979-01-01 12:00 UT	752.8(1.8)	
		686.2	0.05	0.3			
	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	2804	0.05	0.19	1992-07-01 12:00 UT	- [9]	
	$4\pi\beta$ - γ coincidence, anti- coincidence (4P-PC-BP- NA-GR-CO, 4P-PC-BP- NA-GR-AC)	844.3	0.03	0.3	2004-12-15 00:00 UT	[10]	
CIEMAT	LSC(CIEMAT/NIST), $4\pi(PC)$ β - $\gamma(NaI(Tl))$ coincidence, $4\pi\gamma(NaI(TI))$ (4P-LS-MX-00-00-CN, 4P-PC-BP-NA-GR-CO, 4P-NA-GR-00-00)	1540	0.18	0.06	2001-04-01 12:00 UT	754.26	
CMI	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	4281	0.2	0.4	1978-01-17 11:00 UT	760	
CNEA	$4\pi(PC)\beta-\gamma$ coincidence $(4P-PC-BP-NA-GR-CO)$	205.4	0.6	1.6	1986-10-20 00:00 UT	-	
	$4\pi(\text{PPC})\beta$ - γ coincidence (4P-PP-BP-NA-GR-CO)	1237	0.4	0.3	2005-05-10 12:00 UT	754	
IAEA	ionization chamber (4P-IC-GR-00-00-00) ^b	3407	0.2	0.4	1978-01-17 11:00 UT	760	
	ionization chamber (4P-IC-GR-00-00) ^c	1734	0.07	0.3	1978-07-01 12:00 UT	752.4(3.7)	

... Continuation of Table 2.

NMI or	Method used and the	thod used and the Activity Relative		Reference	Half-life	
labora-	acronym	$A_i/\mathbf{k}\mathbf{B}\mathbf{q}$	standard		date	$/\mathbf{d}$
tory			uncertainty			
			$/10^{-2}$			
			A	В	yyyy-mm- dd	
IFIN-HH	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	774.8	0.19	0.52	2006-10-11 00:00 UT	- [11]
IRA	$4\pi(PC)\beta-\gamma$ coincidence (4P-PC-BP-NA-GR-CO)	3151 ^g	0.08	0.63	1977-10-01 00:00 UT	
		3174	0.08	0.63	2000 00 01	77 4 20(22)
	Ionization chamber traceable to $4\pi(PC)\beta-\gamma$ coincidence meaurements made	575.7	0.03	0.51	2008-09-01 00:00 UT	754.26(22) [11]
	in 1978 (4P-IC-GR-00-00- 00 and 4P-PC-BP-NA-GR- CO)					
JRC	$4\pi\beta(\text{PPC})$ - $\gamma(\text{NaI well})$ coincidence (4P-PP-BP-NA-GR-CO) CIEMAT/NIST (4P-LS-	688.9	(0.38	2003-07-01 12:00 UT	- [10]
TIDIGG	MX-00-00-CN)	44.40	0.00		1007.00.01	
KRISS	$4\pi(PPC)\beta-\gamma$ coincidence	4149	0.06	0.17	1995-08-01	
LNE-	(4P-PP-BP-NA-GR-CO) ionization chamber cali-	1258g	0.17	0.12	00:00 UT 1987-11-17	
LNHB	brated by $4\pi\beta$ - γ coincidence (4P-IC-GR-00-00-00 and 4P-00-BP-00-GR-CO)	1200	0.17	0.12	12:00 UT	
	,	1260	0.17	0.12		
	$4\pi\beta$ - γ coincidence, $4\pi\gamma$ well type (4P-PC-BP-NA-GR-CO, 4P-NA-GR-00-00-HE)	2774	0.14	0.14	1998-06-09 12:00 UT	754.3(2)
	$4\pi\gamma$ counting (4P-NA-GR-00-00-HE)	1476.5	0.03	0.18	2005-02-15 00:00 UT	754.26(22) [11]
LNMRI- IRD	$4\pi(PC)\beta-\gamma(NaI(Tl))$ (4P-PC-BP-NA-GR-CO)	775	0.08	0.16	1987-03-01 12:00 UT	-
	$ \begin{array}{c} 4\pi(\mathrm{LS})\beta\text{-}\gamma\text{-}(\mathrm{NaI}(\mathrm{Tl})) \\ \text{anticoincidence (4P-LS-BP-NA-GR-AC)} \end{array} $	321.6	0.37	0.06	2015-09-29 12:00 UT	754.006
NIST	ionization chamber (4P-IC-GR-00-00-00 and 4P-LS-BP-NA-GR-CO) ^d	4100 ^g 3930	0.01	1.1	1977-09-14 17:00 UT	753.1(1.8)
	ionization chamber (4P-IC-GR-00-00-00 and 4P-LS-BP-NA-GR-CO) ^d	1363	0.01	0.29	2002-06-01 17:00 UT	754.15(37)
NMIJ	$4\pi\beta$ - γ coincidence (4P-PC-BP-NA-GR-CO)	1469.8	0.18	0.08	2005-02-15 00:00 UT	753.13
NMISA	$4\pi\beta(LS)$ - γ coincidence (4P-LS-BP-NA-GR-CO)	170.4	0.11	0.21	2009-11-26 10:00 UT	754.26(22) [11]

... Continuation of Table 2.

NMI or	Method used and the	Activity	Relativ	⁄e	Reference	Half-life
labora- tory	acronym	$A_i/{f kBq}$	$egin{array}{c} \mathbf{standar} \ \mathbf{uncerta} \ /\mathbf{10^{-2}} \ \end{array}$		date	/ d
			A	В	yyyy-mm- dd	
NPL	Ionization chamber traceable to $4\pi(PC)\beta-\gamma$ coincidence measurements (4P-IC-GR-00-00-00 and 4P-PC-BP-NA-GR-CO)	684.6 ^g	0.07	0.35	1976-12-20 00:00 UT	-
		684.3	0.07	0.35		
NRC	(4P-BP-PP-GR-NA-AC)	2924	0.1	0.4	2011-12-22 17:00 UT	754.0(5)
POLATOM	(4P-LS-BP-NA-GR-	9862	0.096	0.375	2013-11-01	
	$\mathrm{CO/AC}$				12:00 UT	
PTB	Ionization chamber traceable to $4\pi(PC)\beta-\gamma$ coincidence measurements (4P-IC-GR-00-00-00 and 4P-PC-BP-NA-GR-CO)	1837	0.04	0.36	1994-08-01 00:00 UT	-
	ionization chamber (4P-IC-GR-00-00-00, 4P-PC-BP-NA-GR-CO and 4P-LS-MX-00-00-CN) ^e	5375	0.03	0.21	2004-01-01 00:00 UT	754.0(7)
	(4P-PC-MX-NA-GR-CO) ^f (4P-LS-MX-NA-GR-CO) (4P-LS-MX-00-00-CN) (4P-LS-MX-00-00-TD)	2519	0.11	0.23	2016-06-01 00:00 UT	754.0(5)
PTKMR	$4\pi(PC)\beta-\gamma$ coincidence (4P-PC-BP-NA-GR-CO)	988.3	0.32	0.5	1984-11-01 08:00 UT	-

^b traceable to the $4\pi\beta$ - γ coincidence measurements at the CMI-IIR

Details regarding the solutions submitted are shown in Table 3, including any impurities, when present, as identified by the laboratories. When given, the standard uncertainties on the evaluations are shown.

^c traceable to RCC, Amersham

^d calibrated in 1977 by $4\pi(LS)$ Beta- γ coincidence

^e calibrated in July 2004 by $4\pi(PC)$ Beta- γ coincidence and CIEMAT/NIST method

f The final result is the weighted mean of the results of four methods. The relative uncertainty (0.25%) of 4P-PC-MX-NA-GR-CO is adopted for the final results. This is larger than the internal and external relative uncertainties of the weighted mean.

g Several samples submitted

Table 3: Details of each solution of $^{134}\mathrm{Cs}$ submitted.

NMI or	Chemical	Solvent conc.	Carrier	Density	Relative activity of
laboratory	composi-		conc.		any impurity ^c
/ CID	tion	// 11 -3)		-3)	
/ SIR year	G GI : TTGI	$/(\text{mol dm}^{-3})$	$/(\mu g g^{-1})$	$/(g \text{cm}^{-3})$	0.00 (4
AECL 1977	CsCl in HCl	0.3	Cs ⁺ :10	1	<0.02 %
1992	CsCl in HCl	0.3	CsCl:25	1.003	<0.02(1) %
BARC 1981	$CsNO_3$ in	0.01	$CsNO_3:40$	-	-
1000	HNO ₃	0.01	G 110 25		
1996	$CsNO_3$ in	0.01	CsNO ₃ :37	1	-
2005	HNO ₃				
2005	CsCl in HCl	-	- 0 0 50	1	-
BEV 2008	CsCl in HCl	0.1	CsCl:50	1	127.0
BIPM 1978 ^a	CsCl in HCl	0.2	CsCl:20	-	¹³⁷ Cs: <0.01 %
BKFH 1979	CsCl in HCl	0.1	Cs:100	-	<0.01 %
1992	Cs in HCl	0.1	Cs:25	-	-
2004	Cs in HCl	0.1	Cs:25	-	-
CIEMAT	CsCl in HCl	1	Cs ⁺ :150	1.019	-
2001					
CMI 1978	CsCl in HCl	0.08	CsCl:20	-	<0.1 %
CNEA 1987	CsCl in HCl	0.1	CsCl:50	0.999	<0.1 %
2005	CsCl in HCl	0.1	CsCl:51	-	-
IAEA 1978	CsCl in HCl	0.08	CsCl:20	-	<0.1 %
1979	Cs in HCl	0.1	Cs:100	1.001	<0.05 %
IFIN-HH	CsCl in HCl	1	CsCl:100	1	$<10^{-2}$ %
2006					
IRA 1978	Cs ⁺ in HCl	0.1	Cs ⁺ :25	-	-
2009	CsCl in HCl	0.1	CsCl:50	1.0005	-
JRC 2004	CsCl in HCl	0.1	CsCl:50	-	-
KRISS 1996	CsCl in HCl	0.2	CsCl:20	1.0033	-
LNE-LNHB	CsCl in HCl	0.1	CsCl:10	0.998	$<10^{-2}$ %
1987					
1998	CsCl in HCl	0.1	CsCl:10	1.01	⁶⁰ Co: 0.0013(3) %
2005	CsCl in HCl	0.1	CsCl:100	1.002	-
LNMRI-IRD	Cs in HCl	0.1	Cs:59	1.003	$^{137}\mathrm{Cs:}\ 0.06(1)\ \%$
1987					
2015	CsCl in HCl	0.1	CsCl:50	1.005	-
NIST 1977	CsCl in HCl	0.1	CsCl:57	-	-
2002	CsCl in HCl	0.9	CsCl:20	1.015(2)	-
NMIJ 2005	CsCl in HCl	0.1	CsCl:100	1.002	_b
NMISA 2010	HCl	0.1	-	1	-
NPL 1976	CsCl in HCl	0.1	CsCl:100	-	<0.04 %
NRC 2014	HCl	0.1	-	1	-
POLATOM	CsCl in HCl	0.2	CsCl:31.7	1	<0.1 %
2014					
PTB 1994	CsCl in HCl	0.1	CsCl:50	1	-
2004	CsCl in HCl	0.1	CsCl:50	1	-
2016	CsCl in HCl	0.1	CsCl:50	1	-
PTKMR	CsCl in HCl	3	CsCl:26.5	1.092(3)	-
1984					

NMI or	Chemical	Solvent conc.	Carrier	Density	Relative activity of
laboratory	composi-		conc.		any impurity ^c
	tion				
/ SIR year		/ (mol dm ⁻³)	$/(\mu \mathrm{g}\mathrm{g}^{-1})$	$/(g \text{cm}^{-3})$	

... Continuation of Table 3.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a dedicated database [12]. The latest submission has added 4 ampoules for the activity measurements for ¹³⁴Cs giving rise to 44 ampoules in total.

The SIR equivalent activity, A_{ei} , for each ampoule received from each NMI, i, including both previous and new results, is given in Table 4. The relative standard uncertainties arising from the measurements in the SIR are also shown. This uncertainty is additional to that declared by the NMI $(u(A_i))$ for the activity measurement shown in Table 2. Although submitted activities are compared with a given source of 226 Ra, all the SIR results are normalized to the radium source number 5 [1]. Table 4 also shows the comparison results selected for the KCRV as explained in section 4.1.

Table 4: Results of SIR measurement of $^{134}\mathrm{Cs}.$

NMI or labo-	m_i	A_i	$^{226}\mathrm{Ra}$	$A_{\mathbf{e}i}$	Relative	$u_{\mathbf{c}i}$	$A_{\mathbf{e}i}$ for
ratory			source		uncert.		KCRV
					from		
					SIR		
/ SIR year	$/\mathbf{g}$	/kBq		/kBq	$/10^{-4}$	$/{f kBq}$	$/\mathbf{kBq}$
AECL 1977	1.565 03 ^a	884.3	3	10 094	7	33	-
	1.595 22	901.4	3	10 088	7	33	-
1992	0.171 51	699.2	3	10 144	6 ^a	17	10 144(17)
BARC 1981	3.597 8	533.2	3	10 094	7	71	-
1996	3.616	607.5	3	10 128	7	48	-
2005	3.600 03	1259	3	10 143	7	48	10 143(48)
BEV 2008	3.615	2299	4	10 090	5	70	-
BIPM 1978	3.618 91	3005.4	4	10 091	5	14	10 092(14) ^c
	3.621 89	3007.8	4	10092	5	14	-
BKFH 1979	3.605 1	686.4	3	10 125	6	3	-
	3.603 9	686.2	3	10 122	6	1	-
1992	3.610 6	2804	4	10 119	5	21	-
2004	3.630 7	844.3	3	10 131	6	31	10 131(31)
CIEMAT 2001	3.648 9	1540	3	9936	6	20	_d
CMI 1978	3.591 62	4281	4	10 124	7	46	10 124(46)
CNEA 1987	3.663 8	205.4	1	10 010	12	170	-
2005	3.659 54	1237	3	10 190	6	48	10 190(48)

 $^{^{\}rm a}$ the solution used for the 1978 CCRI(II)-K2.Cs-134 comparison

^b the solution used for the 2004 APMP.RI(II)-K2.Cs-134 comparison

^c The ratio of the activity of the impurity to the activity of ¹³⁴Cs at the reference date

 \dots Continuation of Table 4.

NMI or labo-	m_i	A_i	$^{226}\mathbf{Ra}$	$A_{\mathbf{e}i}$	Relative	$u_{\mathbf{c},i}$	$A_{\mathbf{e}}$ for KCRV
ratory			source		uncert.	,	
					from		
					SIR		
/ SIR year	$/\mathbf{g}$	/kBq		/kBq	$/10^{-4}$	$/\mathbf{kBq}$	$/k\mathbf{Bq}$
IAEA 1978	3.594 18	3407	4	10 117	8	46	-
1979	3.624 4	1734	3	10 159	14	34	-
IFIN-HH 2006	3.596 9	774.8	3	10 222	6	56	10 222(56)
IRA 1978	3.009 38	3151	4	10 022	9	64	10 023(64) ^c
	3.031 90	3174	4	10 024	9	64	_
2009	3.568 17(9)	575.7	2	10 031	12	52	-
JRC 2004	3.629 39	688.9	3	10 047	8	39	10 047(39)
KRISS 1996	3.599 23	4149	4	10 214	7	20	10 214(20)
LNE-LNHB	3.613 43	1258	3	10 125	6	2	-
1987							
	3.619 76	1260	3	10 129	6	2	-
1998	3.613	2774	4	10 129	6	16] <u>-</u>
2005	3.602	1476.5	3	10 124	7	20	10 124(20)
LNMRI-IRD	3.521 97	775	3	9998	8	19	-
1987							
2015	3.548 756	321.6	2	10 087	11	39	10 087(39)
NIST 1977	3.720 72	4100	4	10 121	5	1	-
	3.568 63	3930	4	10 114	5	1	-
2002	3.635 6(2)	1363	3	10 141	6	31	10 141(31)
NMIJ 2005	3.589 36	1469.8	3	10 104	6 ^b	19	10 104(19)
NMISA 2010	3.570 39	170.4	1	10 101	16	29	10 101(29)
NPL 1976	3.639 6	684.6	3	10 087	6	36	10 089(36) ^c
	3.637 8	684.3	3	10 091	6	36	-
NRC 2014	3.605 2	2924	3	10 131	8	43	10 131(43)
POLATOM	3.652 66	9862	5	10 107	5	39	10 107(39)
2014							
PTB 1994	3.646	1837	3	10 069	6	37	-
2004	3.636 3(9)	5375	4	10 080	6	23	-
2016	3.629 78	2519	4	10 081	5	26	10 081(26)
PTKMR 1984	3.61	988.3	3	10 188	6	61	10 188(61)

^a mass of active solution before dilution

The APMP.RI(II)-K2.Cs-134 comparison held in 2005 [14]. The results were linked to the BIPM.RI(II)-K1.Cs-134 comparison through the measurement in the SIR of at least one ampoule of the APMP comparison, as explained in [5].

4.1. The key comparison reference value

In May 2013, the CCRI(II) decided to calculate the key comparison reference value (KCRV) by using the power-moderated weighted mean [15] rather than an unweighted mean, as had been the policy. This type of weighted mean is similar to a Mandel-Paule

^b result used to link the 2004 APMP key comparison

^c An average value and average uncertainty between all submitted samples is used for the KCDB [13].

d result identified as an outlier

mean in that the NMIs' uncertainties may be increased until the reduced chi-squared value is one. In addition, it allows for a power α smaller than two in the weighting factor. As proposed in [15], α is taken as 2-3/N where N is the number of results selected for the KCRV. Therefore, all SIR key comparison results can be selected for the KCRV with the following provisions:

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

Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are made only by the CCRI(II) during one of its biennial meetings, or by consensus through electronic means (e.g., email) as discussed at the CCRI(II) meeting in 2013.

Consequently, using the recent result produces an updated KCRV for 134 Cs in 2016 of **10 123(10)** kBq with the power $\alpha=1.85$ that has been calculated using the previously published results, selected as shown in Table 4, for the NPL (1976), BIPM (1978), IRA (1978), CMI (1978), PTKMR (1984), AECL (1992), KRISS (1996), NIST (2002), BKFH (2004), JRC (2004), LNE-LNHB (2005), NMIJ (2005), BARC (2005), CNEA (2005), IFIN-HH (2006), NMISA (2010), NRC (2014), POLATOM (2014), LNMRI-IRD (2015), and the present PTB (2016) result. This can be compared with the previous KCRV values of 10 096(16) kBq published in 2003 [3] and 10 116(13) kBq published in 2007 [5].

4.2. Degrees of equivalence

Every participant in a comparison is entitled to have one result included in the KCDB as long as the NMI is a signatory or designated institute listed in the CIPM MRA and the result is valid (i.e., not older than 20 years). No recent submission has been identified as a pilot study so the most recent result of each NMI is normally eligible for inclusion on the KCDB platform of the CIPM MRA [2]. An NMI may withdraw its result only if all other participants agree.

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

4.2.1. Comparison of a given NMI result with the KCRV

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

$$D_i = A_{ei} - KCRV \tag{1}$$

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

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

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

$$u^{2}(D_{i}) = (1 - 2w_{i})u_{i}^{2} + u^{2}(KCRV)$$
(3)

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

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

The introductory text in Appendix A is the one agreed by the CCRI(II) for all the K1 comparisons.

4.2.2. Comparison between pairs of NMI results

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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \tag{5}$$

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

$$u^{2}(D_{ij}) = u_{i}^{2} + u_{i}^{2} - 2u(A_{ei}, A_{ej})$$

$$(6)$$

where any obvious correlations between the NMIs (such as a traceable calibration, correlations normally coming from the SIR, or from the linking factor in the case of linked comparison) are subtracted using the covariance $u(A_{ei}, A_{ej})$ (see [16] for more detail). However, the CCRI decided in 2011 that these pair-wise degrees of equivalence no longer need to be published as long as the methodology is explained.

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

5. Conclusion

The BIPM continuous key comparison for ¹³⁴Cs, BIPM.RI(II)-K1.Cs-134, currently comprises 15 results. The KCRV has been recalculated to include the latest result from the POLATOM (Poland), LNMRI-IRD (Brazil), PTB (Germany), and the NRC (Canada). The SIR results, together with the previously published APMP.RI(II)-K2.Cs-134 results, have been analyzed with respect to the updated KCRV, providing degrees of equivalence for 17 national metrology institutes. Other results may be added when other NMIs contribute ¹³⁴Cs activity measurements to this comparison or take part in other linked comparisons.

6. References

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Appendix A. Introductory text for ¹³⁴Cs degrees of equivalence

Key comparison BIPM.RI(II)-K1.Cs-134

MEASURAND: Equivalent activity of ¹³⁴Cs

Key comparison reference value: the SIR reference value $x_{\rm R}$ for this radionuclide is 10123 kBq, with a standard uncertainty, $u_{\rm R}$ equal to 10 kBq (see Section 4.1 of the Final Report). The value x_i is taken as the equivalent activity for a laboratory i.

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

Appendix B. Table of degrees of equivalence for BIPM.RI(II)-K1.Cs-134

Table B1: The table of degrees of equivalence for ${\rm BIPM.RI(II)\text{-}K1.Cs\text{-}134}$

NMI i	$A_{\mathbf{e}i}/\mathbf{kBq}$	u_i / \mathbf{kBq}	D_i / \mathbf{kBq}	U_i / \mathbf{kBq}
NIST	10141	31	18	62
JRC	10047	39	-76	77
BKFH	10131	31	8	62
LNE-LNHB	10124	20	1	42
NMIJ	10104	19	-19	41
BARC	10143	48	20	95
CNEA	10190	48	67	95
IFIN-HH	10222	56	99	111
BEV	10090	70	-33	142
IRA	10031	52	-92	106
NMISA	10101	29	-22	58
POLATOM	10107	39	-16	77
NRC	10131	43	8	85
LNMRI-IRD	10087	39	-36	77
PTB	10081	26	-42	53

Table B2: The table of degrees of equivalence for the APMP.RI(II)-K2.Cs-134(2005) comparison

NMI i	$A_{\mathbf{e}i}/\mathbf{k}\mathbf{B}\mathbf{q}$	u_i / \mathbf{kBq}	D_i / \mathbf{kBq}	U_i / \mathbf{kBq}
INER	10178	18	55	42
VNIIM	10094	26	-29	56

Appendix C. Graph of degrees of equivalence with the KCRV for ¹³⁴Cs (as it appears in Appendix B of the MRA)

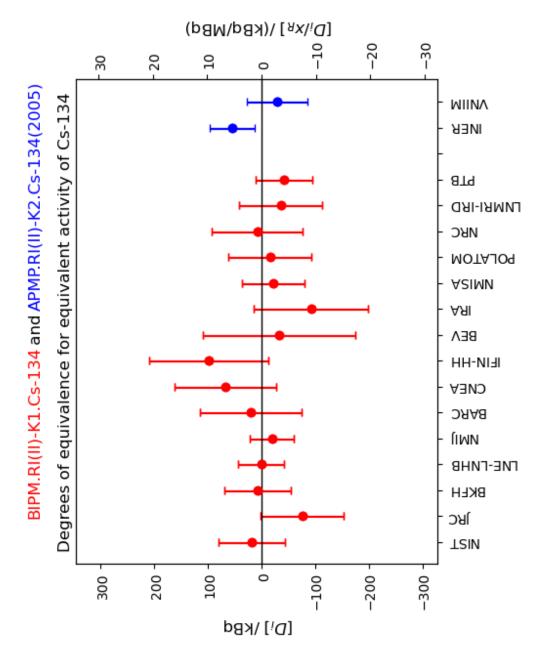


Figure C1. Degrees of equivalence for equivalent activity of ¹³⁴Cs.

Appendix D. Uncertainty budgets for the activity of $^{134}\mathrm{Cs}$ submitted to the SIR

Detailed Uncertainty Budget

Laboratory: LNMRI/IRD; Radionuclide: Cs-134; Ampoule number: 73L15

Uncertainty components*, in % of the activity concentration, due to

		Remarks	Evaluation
			type (A or B)
counting statistics	0.33	standard desviation	A
weighing	0.05		В
Live time	0.01		В
background	0.04		В
pile-up			
counting time			
adsorption	====		
impurities			
tracer		المعارض بمعارض المعارض المعارض المعارضة المعارضة	
input parameters and statistical model			
quenching			######################################
interpolation from calibration curve			
decay-scheme parameters			
half life ($T_{1/2} = 2.0644 \text{ y}$;			
u = 0.0014 y)	< 0.01		В
self absorption	~~~~~		
extrapolation of efficiency curve	0.17	Mean unt. 10 extrap	A
other effects (if relevant) (explain)			
combined uncertainty (as quadratic sum of all uncertainty components)			0.37 %

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, ISO, corrected and reprinted 1995).

The NRC (2014) uncertainty budget:

Relative standard uncertainties	u _i x 10 ⁴ Evaluated by method		
Contributions due to	A	В	Comment
Counting statistics	10	_	External uncertainty of weighted average of 9 sources
Weighing	_	< 3	Balance calibration
Live-time		< 0.1	$\Delta t_{ m live}/t_{ m live}$
Background		1	
Extrapolation of efficiency curve		40	Mean std error on N_0
Quadratic summation	10	40	
Combined standard uncertainty	2	11	

Detailed Uncertainty Budget

Laboratory: Laboratory of Radioactivity Standards, Radioisotope Centre POLATOM, National Centre for Nuclear Research;

Radionuclide: ¹³⁴Cs;

Ampoule number: BW/34/13

Uncertainty components*, in % of the activity concentration, due to:

		Remarks	Evaluation
			type (A or B)
counting statistics	0.096		A
weighing	0.224		В
dead time			
background			
pile-up			
counting time	0.001	,	В
adsorption			
impurities			
tracer			
input parameters and statistical model			
quenching			
interpolation from calibration curve			
decay-scheme parameters			
half life $(T_{1/2} = 2.0644 \text{ y})$; u = 0.0014 y	0.001		В
self absorption			
extrapolation of efficiency curve	0.3		В
other effects (if relevant) (explain)	0.014	coincidence gate	В
combined uncertainty (as quadratic sum of all uncertainty components)	0.387		

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, ISO, corrected and reprinted 1995).

Detailed Uncertainty Budget (4P-LS-MX-00-00-TD)

Laboratory: PTB; Radionuclide: Cs-134; Ampoule number: 2016-1320.

Uncertainty components*, in % of the activity concentration, due to

		Remarks	Evaluation
		type (A	or B)
counting statistics	0.01	std. dev. of mean of 4 samples	A
weighing	0.03		B
dead time	0.03		B
background	0.03		A
pile-up	n.a.		
counting time	0.01		B
adsorption	0.05		B
impurities	<0.03	no impurities detected	B
TDCR value and fit	0.1		B
model + nucl. decay data	0.2	e.g. shape of beta spectra	B
ionization quenching			
and kB	0.1		B
decay correction	<0.03		B
quenching indicator (SQP(E),tSIE)	n.a.		B
dilution	0.05	two dilution steps	B
PMT asymmetry	0.05		B
sample (in)stability	0.05	long-term (in)stability of LS samples	B
combined uncertainty	0.28		

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, <u>JCGM 100:2008</u>

Detailed Uncertainty Budget (4P-LS-MX-00-00-CN)

Laboratory: PTB;	Radionuclide:Cs-13	34; Ampoule number: 2016-	1320.
Uncertainty components*, in % of the activity concentration, due to			
counting statistics	0.02	std. dev. of mean of 4 samples	A
weighing	0.03		B
dead time	0.1	*****	B
background	0.05		A
pile-up	п.а.		
counting time	0.01		B
adsorption	0.05		B
impurities	<0.03	no impurities detected	B
tracer and fit	0.06		B
model + nucl. decay data	0.2	e.g. shape of beta spectra	B
ionization quenching			
and kB	0.06		B
decay correction	< 0.03		B
quenching indicator (SQP(E),tSIE)	n.a.		B
dilution	0.05	two dilution steps	B
PMT asymmetry	0.07		B
sample (in)stability	0.05	long-term (in)stability of LS samples	B
combined uncertainty	0.28		

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, <u>JCGM 100:2008</u>

Detailed Uncertainty Budget (4P-LS-MX-NA-GR-CO)

Laboratory: PTB; Radionuclide:Cs-134; Ampoule number: 2016-1320. Uncertainty components*, in % of the activity concentration, due to counting statistics 0.07 correlations taken into account A weighing 0.03 ___B___ dead time 0.20 __B___ _____ 0.03 background correlations taken into account ___A____ pile-up n.a. _____ counting time negligible __B___ _____ 0.05 adsorption __B___ _____ impurities < 0.03 __B___ no impurities detected __B___ resolving time 0.20 _____ __B___ < 0.03 decay correction _____ nucl. decay data negligible extrapolation of efficiency ____B___ 0.07 curve fitting deviation 0.15 __B___ dilution 0.05 two dilution steps combined uncertainty 0.35

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, <u>JCGM 100:2008</u>

Detailed Uncertainty Budget (4P-PC-MX-NA-GR-CO)

Laboratory: **PTB**; Radionuclide: Cs-134; Ampoule number: 2016-1320. *Uncertainty components**, in % of the activity concentration, due to

counting statistics	0.11	correlations taken into account	A
weighing	0.04		B
dead time	negligible		B
background	0.02	correlations taken into account	A
pile-up	n.a.		
counting time	negligible		B
adsorption	0.05		B
impurities	< 0.03	no impurities detected	B
resolving time	negligible		B
decay correction	<0.03		B
nucl. decay data	negligible		B
extrapolation of efficiency			
curve	0.07		B
fitting deviation	0.19		B
dilution	0.05	two dilution steps	B
combined uncertainty	0.25		

^{*} The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, <u>JCGM 100:2008</u>

Appendix E. Acronyms used to identify different measurement methods

Each acronym has six components, geometry-detector (1)-radiation (1)-detector (2)-radiation (2)-mode. When a component is unknown, ?? is used and when it is not applicable 00 is used.

Geometry	acronym	Detector	acronym
4 π	4P	proportional counter	PC
defined solid angle	SA	press. Prop. Counter	PP
2 π	2P	liquid scintillation counting	LS
undefined solid angle	UA	NaI(Tl)	NA
		Ge(HP)	GH
		Ge(Li)	GL
		Si(Li)	SL
		CsI(Tl)	CS
		ionization chamber	IC
		grid ionization chamber	GC
		Cerenkov detector	CD
		calorimeter	CA
		solid plastic scintillator	SP
		PIPS detector	PS
		CeBr3	СВ

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	СО
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with	СТ
		efficiency tracing	
x-rays	XR	anti-coincidence counting	AT
		with efficiency tracing	
photons $(x + \gamma)$	PH	triple-to-double coincidence	TD
		ratio counting	
${ m photons} + { m electrons}$	PE	selective sampling	SS
alpha particle	AP	high efficiency	HE
mixture of various radi-	MX	digital coincidence counting	DC
ation			

Examples of methods	acronym
$4\pi(PC)\beta-\gamma$ coincidence counting	4P-PC-BP-NA-GR-CO
4π (PPC)β- γ coincidence counting	4P-PP-MX-NA-GR-CT
eff. trac	
defined solid angle α -particle	SA-PS-AP-00-00-00
counting with a PIPS detector	
$4\pi(PPC)AX-\gamma(GeHP)-$	4P-PP-MX-GH-GR-AC
anticoincidence counting	
$4\pi \text{CsI-}\beta, AX, \gamma \text{ counting}$	4P-CS-MX-00-00-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG