Update of the BIPM comparison BIPM.RI(II)-K1.Cs-134 of activity measurements of the radionuclide ^{134}Cs to include the 2005 results of the BARC (India) and the CNEA (Argentina), the 2006 result of the IFIN-HH (Romania) and the link for the 2005 regional comparison APMP.RI(II)-K2.Cs-134 to include the VNIIM and the INER

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Abstract

In 2005, the Bhabha Atomic Research Centre, (BARC) India and the Comisión Nacional de Energía Atómica (CNEA), Argentina each submitted one sample of known activity of ^{134}Cs to the International Reference System (SIR). This was followed in 2006 by the submission of an ampoule by the Institutul de Fizica si Inginerie Nucleara (IFIN-HH), Romania. The values of the activity submitted were between about 0.7 MBq and 1.3 MBq. The 2005 results replace earlier SIR measurements while the 2006 result of the IFIN-HH replaces a CCRI(II) comparison result of 1978. Consequently, there are now thirteen results in the BIPM.RI(II)-K1.Cs-134 comparison for which the key comparison reference value has been updated. Also, in 2005, the APMP ran a key comparison for the activity of this radionuclide and the results are now linked to the BIPM.RI(II)-K1.Cs-134 comparison through the previous submissions of the pilot laboratory, NMIJ (Japan) and of the LNE-LNHB (France). This has enabled the VNIIM (Russian Federation) to update its result from the 1978 CCRI(II)-K2.Cs-134 comparison and the INER (Chinese Taipei) to be linked for the first time. In addition, the remaining eight eligible results from the 1978 CCRI(II) comparison are still linked provisionally to the SIR results.

1. Introduction

The SIR for activity measurements of γ-ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid form, or a different standard ampoule for radioactive gases. The NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of ^{226}Ra using pressurized ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity, A_e, are all given in [1].

From its inception until 31 December 2006, the SIR has measured 894 ampoules to give 655 independent results for 63 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the CIPM key comparison database (KCDB) of the Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Cs-134 key comparison and the earlier results have been published [3, 4].

In May 2007, the CCRI(II) decided to change the key comparison reference value (KCRV) for this activity comparison and consequently all the previous details that are relevant to the values to be included in the KCRV are also given in this report.

In addition, a regional key comparison was held in 2004 for this radionuclide, APMP.RI(II)-K2.Cs-134, piloted by the NMIJ [5]. Four laboratories from three RMOs took part in this comparison, including the NMIJ. All these NMIs made primary standardizations, two laboratories are linking laboratories and the other two are eligible to be linked to the BIPM key comparison as listed in Table 1b. The VNIIM has used this regional comparison to update their previously published 1978 CCRI(II)-K2.Cs-134 comparison result.

2. Participation

In addition to the two ampoules submitted by the BARC and the CNEA in 2005, which replace their earlier SIR submissions, and the one submitted by the IFIN-HH in 2006, which is their first submission, fourteen NMIs and four other laboratories have submitted 34 ampoules for the comparison of ^{134}Cs activity measurements since 1976. The recent BARC, CNEA and the IFIN-HH details are given in Table 1a together with the details of the earlier participations from [3, 4].

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

Original acronym	**NMI**	**Full name**	**Country**	**Regional metrology organization**	**Date of measurement at the BIPM** YYYY-MM-DD
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1976-12-30
AECL	–	Atomic Energy of Canada Ltd	Canada	SIM	1977-05-23 1992-07-03
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1977-10-18 2002-06-21

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

Original acronym	**NMI**	**Full name**	**Country**	**Regional metrology organization**	**Date of measurement at the BIPM** YYYY-MM-DD
IER	IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	1978-02-24
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1978-04-17
IAEA	–	International Atomic Energy Agency	–	–	1978-05-26 1979-02-13
BIPM	–	Bureau International des Poids et Mesures	–	–	1978-11-13
OMH	MKEH	Országos Mérésügyi Hivatal	Hungary	EUROMET	1979-01-26 1992-06-14 2004-12-08
–	BARC	Bhabha Atomic Research Centre	India	APMP	1981-09-03 1996-11-22 2005-07-20
PDS	PTKMR	Pusat Teknologi Keselamatan dan Metrologi Radiasi	Indonesia	APMP	1984-11-22
–	CNEA	Comision Nacional de Energia Atomica	Argentina	SIM	1987-01-07 2005-09-12
IRD	LNMRI	Laboratorio Nacional de Metrologia das Radiaçoes Ionizantes	Brazil	SIM	1987-10-19
LMRI LPRI	LNE-LNHB	Bureau national de métrologie-Laboratoire national Henri Becquerel	France	EUROMET	1987-11-30 1998-10-06 2005-04-13

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

Original acronym	**NMI**	**Full name**	**Country**	**Regional metrology organization**	**Date of measurement at the BIPM** YYYY-MM-DD
–	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1994-09-15 2004-07-05
–	KRISS	Korea Research Institute of Standards and Science	Republic of Korea	APMP	1996-02-22
–	CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnològicas	Spain	EUROMET	2001-04-27
–	IRMM	Institute for Reference Materials and Measurements	European Union	EUROMET	2004-01-20
–	NMIJ	National Metrology Institute of Japan	Japan	APMP	2005-04-20
	IFIN-HH	Institutul de Fizica si Inginerie Nucleara-Horia Hulubei	Romania	EUROMET	2006-11-21

Four participants took part in the APMP.RI(II)-K2.Cs-134 comparison held in 2004. Their details are given in Table 1b.

3. NMI standardization methods

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

A brief description of the standardization methods for each laboratory, the activities submitted and the relative standard uncertainties ($k = 1$) are given in Table 2. Details concerning the standardization methods and the uncertainty budgets used in the regional comparison are given in [5]. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. The SIR uncertainty budgets for the previous participants are in [3, 4] while those for the BARC, CNEA and the IFIN-HH are given in Appendix 1 attached to this report.

Table 1b. Details of the participants in the APMP.RI(II)-K2.Cs-134

NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
NMIJ*	National Metrology Institute of Japan	Japan	APMP	2005-04-20#
LNE-LNHB*	Laboratoire national de métrologie et d'essais-Laboratoire national Henri Becquerel	France	EUROMET	2005-04-13
VNIIM	D.I. Mendeleyev Institute for Metrology	Russia	COOMET	–
INER	Institute of Nuclear Energy Research	Chinese Taipei	APMP	–

* linking laboratories with SIR submissions as indicated

an early activity submission for the same ampoule in February 2004 had been a pilot study based on an ionization chamber measurement only.

The half-life used in the SIR and in the 1978 CCRI(II)-K2.Cs-134 comparison is 753.1 (1.8) d [6], as used in the APMP.RI(II)-K2.Cs-134 comparison, which is in agreement with the half-life recommended by the IAEA [7], 754.28 (22) d and the BNM-CEA value of 754.26 (22) d [8].

In May 2007, the CCRI(II) agreed to change the KCRV for this comparison and consequently, the standardization methods of the new and all previous submissions are included in Table 2.

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. The BIPM has 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. However, no such impurity measurement has been carried out at the BIPM for ^{134}Cs and the SIR corrections for impurities are negligible in all cases.

Table 2. Standardization methods of the participants for ^{134}Cs

NMI	Method used and acronym (see Appendix 2)	Half-life	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
NPL	Pressurized IC[b] 4P-IC-GR-00-00-00	–	684.6 684.3[a]	76-12-20 0 h UT	0.07	0.35
AECL	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	2.062 (5) a	884.3 901.4[a]	77-03-18 17 h UT	0.014	0.32
			699.2	92-05-01 17 h UT	0.15	0.02
NIST	Pressurized IC 4P-IC-GR-00-00-00	2.062 (5) a	4100 3930[a]	77-09-14 17 h UT	0.01	1.10
	calibrated in 1977 by 4π(LS)β-γ coinc. 4P-LS-BP-NA-GR-CO	2.0648 (10) a	1363	02-06-01 17 h UT	0.09	0.29
IRA	4π(PC)β-γ coincidence 4P-PC-BP-NA-GR-CO	–	3151 3174[a]	77-10-01 0 h UT	0.08	0.63
CMI-IIR	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	2.08 a	4281	78-01-17 11 h UT	0.2	0.4
IAEA	Pressurized IC [c] 4P-IC-GR-00-00-00	2.08 a	3407	78-01-17 11 h UT	0.2	0.4
	Pressurized IC[d] 4P-IC-GR-00-00-00	2.06 (1) a	1734	78-07-01 12 h UT	0.07	0.30
BIPM [e]	4π(PC)β-γ coincidence 4P-PC-BP-NA-GR-CO	[6]	3005.4 3007.8[a]	78-10-15 0 h UT	0.03	0.13
MKEH	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	2.061 (5) a	686.4 686.2[a]	79-01-01 12 h UT	0.05	0.30
		[6]	2804	92-07-01 12 h UT	0.05	0.19
	4πβ-γ coincidence and anti-coincidence 4P-PC-BP-NA-GR-CO 4P-PC-BP-NA-GR-AC	[7]	844.3	04-12-15 0 h UT	0.03	0.30
BARC	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	–	533.2	81-08-01 6 h 30 UT	0.03	0.70
		–	607.5	96-08-01 6 h 30 UT	0.4	0.3
	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	[6]	1259	05-05-15 0 h UT	0.4	0.2

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Table 2 continued. Standardization methods of the participants for ^{134}Cs

NMI	Method used and acronym (see Appendix 2)	Half-life	Activity / kBq	Reference date	Relative standard uncertainty × 100 by method of evaluation	
				YY-MM-DD	A	B
PTKMR	4π(PC)β-γ coinc. 4P-PC-BP-NA-GR-CO	–	988.3	84-11-01 8 h UT	0.3	0.5
CNEA	4π(PC)β-γ coinc. 4P-PC-BP-NA-GR-CO	–	205.4	86-10-20 0 h UT	0.6	1.6
	4π(PPC)β-γ coinc. 4P-PP-BP-NA-GR-CO	754.0 d	1237	05-05-10 12 h UT	0.4	0.3
LNMRI	4π(PC)β-γ(NaI(Tl)) coincidence 4P-PC-BP-NA-GR-CO	–	775.0	87-03-01 12 h UT	0.08	0.16
LNE-LNHB	Pressurized IC 4P-IC-GR-00-00-00 calibrated by 4πβ-γ coincidence	–	1258 1260[a]	87-11-17 12 h UT	0.17	0.12
	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO and 4πγ well type 4P-NA-GR-00-00-00	754.3 (2) d	2774	98-06-09 12 h UT	0.14	
	4πγ counting [g] 4P-NA-GR-00-00-HE	2.0651 (6) a [8]	1476.5	05-02-15 0 h UT	0.03	0.18
PTB	Pressurized IC [b] 4P-IC-GR-00-00-00	–	1837	94-08-01 0 h UT	0.04	0.36
	Pressurized IC calibrated in July 2004 by 4π(PC)β–γ coincidence 4P-PC-BP-NA-GR-CO and CIEMAT/NIST 4P-LS-MX-00-00-CN	754.0 (7) d	5375	04-01-01 0 h UT	0.03	0.21
KRISS	4π(PPC)β-γ coinc. 4P-PP-BP-NA-GR-CO	–	4149	95-08-01 0 h UT	0.06	0.17
CIEMAT	LSC (CIEMAT/NIST) ; 4P-LS-MX-00-00-CN 4π(PC)β-γ(NaI(Tl)) coincidence ; 4P-PC-BP-NA-GR-CO 4πγ(NaI(Tl)) 4P-NA-GR-00-00-00	2.0651 a	1540[f]	01-04-01 12 h UT	0.18[f]	0.06[f]

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Table 2 continued. Standardization methods of the participants for ^{134}Cs

NMI	Method used and acronym (see Appendix 2)	Half-life	Activity / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
IRMM	$4\pi\beta$ (PPC)-γNaI well) coincidence 4P-PP-BP-NA-GR-CO CIEMAT/NIST 4P-LS-MX-00-00-CN	[7]	691.0 686.3	03-07-01 12 h UT	0.30 0.10	0.32 0.42
NMIJ	$4\pi\beta$-γ coincidence [g] 4P-PC-BP-NA-GR-CO	753.13 d	1469.8	05-02-15 0 h UT	0.18	0.08
IFIN-HH	$4\pi\beta$-γ coincidence 4P-PC-BP-NA-GR-CO	[8]	774.8	06-10-11 0 h UT	0.19	0.52

[a] two ampoules submitted

[b] calibrated by a primary method 4P-PC-BP-NA-GR-CO for the nuclide considered

[c] traceable to the $4\pi\beta$-γ coincidence measurements at the CMI-IIR

[d] traceable to RCC, Amersham

[e] the two ampoules measured by the BIPM for the CCRI(II)-K2.Cs-134 and measured in the SIR were used to make the link for the CCRI(II) key comparison

[f] the result and uncertainties given here are for the CIEMAT/NIST method only

[g] ampoules used to make the link for the 2004 APMP key comparison.

Table 3. Details of the solution of ^{134}Cs submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of any impurities[†]
NPL 1976	CsCl in HCl	0.1	CsCl : 100	–	< 0.04 %
AECL 1977	CsCl in HCl	0.3	Cs^{+} : 10	1	< 0.02 %
1992			CsCl: 25	1.003	< 0.02(1) %
NIST 1977	CsCl in HCl	0.1	CsCl : 57	–	–
2002		0.9	CsCl : 20	1.015 (2)	–
IRA 1978	Cs+ in HCl	0.1	Cs+ : 25	–	–
CMI-IIR 1978	CsCl in HCl	0.08	CsCl : 20	–	< 0.1 %
IAEA 1978	CsCl in HCl	0.08	CsCl : 20	–	< 0.1 %
1979	Cs in HCl	0.1	Cs : 100	1.001	< 0.05 %
BIPM [a] 1978	CsCl in HCl	0.2	CsCl : 20	–	^{137}Cs : < 0.01 %

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Table 3 continued. Details of the solution of ^{134}Cs submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of any impurities[†]
MKEH 1979	CsCl in HCl	0.1	Cs : 100	–	< 0.01 %
1992	Cs in HCl	0.1	Cs: 25	–	–
2004					–
BARC 1981	$CsNO_3$ in HNO_3	0.01	$CsNO_3$: 40	–	–
1996			$CsNO_3$: 37	1	–
2005	CsCl in HCl	–	–	1	–
PTKMR 1984	CsCl in HCl	3	CsCl: 26.5	1.092 (3)	–
CNEA 1987	CsCl in HCl	0.1	CsCl: 50	0.999	< 0.1 %
2005	CsCl in HCl	0.1	CsCl: 51	–	–
LNMRI 1987	Cs in HCl	0.1	Cs : 59	1.003	^{137}Cs: 0.06 (1) %
LNE-LNHB 1987	CsCl in HCl	0.1	CsCl: 10	0.998	< 10^{-2} %
1998				1.01	^{60}Co : 0.0013 (3) %
2005	CsCl in HCl[#]	0.1	CsCl: 100	1.002	–
PTB 1994	CsCl in HCl	0.1	CsCl: 50	1.00	–
2004					–
KRISS 1996	CsCl in HCl	0.2	CsCl : 20	1.0033	–
CIEMAT 2001	CsCl in HCl	1	Cs^+ : 150	1.019	–
IRMM 2004	CsCl in HCl	0.1	CsCl: 50	–	–
NMIJ 2005	CsCl in HCl[#]	0.1	CsCl: 100	1.002	–
IFIN-HH 2006	CsCl in HCl	1	CsCl: 100	1	< 10^{-2} %

[†] the ratio of the activity of the impurity to the activity of ^{134}Cs at the reference date

[a] the solution used in the 1978 CCRI(II)-K2.Cs-134 comparison

[#] the solution used in the 2005 APMP.RI(II)-K2.Cs-134 comparison.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". The activity measurements for ^{134}Cs arise from thirty-seven ampoules and the SIR equivalent activity for each ampoule, A_{ei}, is given in Table 4a for each NMI, *i*. The dates of measurement in the SIR are given in Table 1.

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

Measurements repeated at the BIPM some 8 months later produced the same SIR result for the IFIN-HH ampoule.

Table 4a. Results of SIR measurements of ^{134}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
NPL 1976	3.639 6	684.6	3	10 087[a]	6×10^{-4}	36
	3.6378	684.3		10 091[b]		36
AECL 1977	1.565 03[c]	884.3	3	10 094	7×10^{-4}	33
	1.595 22[c]	901.4		10 088		33
1992	0.171 51[c]	699.2	3	10 144	6×10^{-4}	17
NIST 1977	3.720 72	4100	4	10 121	5×10^{-4}	110
	3.568 63	3930		10 114		
2002	3.635 6 (2)	1363	3	10 141	6×10^{-4}	31
IRA 1978	3.009 38	3151	4	10 022[a]	9×10^{-4}	64
	3.031 90	3174		10 024[b]		64
CMI-IIR 1978	3.591 62	4281	4	10 124[b]	7×10^{-4}	46
IAEA 1978	3.594 18	3407	4	10 117	8×10^{-4}	46
1979	3.624 4	1734	3	10 159	14×10^{-4}	34
BIPM[d] 1978	3.618 91	3005.4	4	10 091[a]	5×10^{-4}	14
	3.621 89	3007.8		10 092		14
MKEH 1979	3.605 1	686.4	3	10 125	6×10^{-4}	31
	3.603 9	686.2		10 122		
1992	3.610 6	2804	4	10 119	5×10^{-4}	21
2004	3.630 7	844.3	3	10 131	6×10^{-4}	31

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Table 4a continued. Results of SIR measurements of ^{134}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
BARC 1981	3.597 8	533.2	3	10 094	7×10^{-4}	71
1996	3.616 0	607.5	3	10 128	7×10^{-4}	48
2005	3.600 03	1259	3	10 143	7×10^{-4}	48
PTKMR 1984	3.610	988.3	3	10 188	6×10^{-4}	61
CNEA 1987	3.663 8	205.4	1	10 010	12×10^{-4}	170
2005	3.659 54	1237	3	10 190	6×10^{-4}	48
LNMRI 1987	3.521 97	775.0	3	9 998	8×10^{-4}	19
LNE-LNHB 1987	3.613 43 3.619 76	1258 1260	3	10 125 10 129	6×10^{-4}	22
1998	3.613 0	2774	4	10 129	6×10^{-4}	16
2005[e]	3.602	1476.5	3	10 124	7×10^{-4}	20
PTB 1994	3.646	1837	3	10 069	6×10^{-4}	37
2004	3.636 3 (9)	5375	4	10 080	6×10^{-4}	23
KRISS 1996	3.599 23	4149	4	10 214	7×10^{-4}	20
CIEMAT 2001	3.648 9	1540	3	9 936	6×10^{-4}	20
IRMM 2004	3.629 39	691.0 686.3[f]	3	10 047	8×10^{-4}	39
NMIJ 2005[e]	3.589 36	1469.8	3	10 104	6×10^{-4}	19
IFIN-HH 2006	3.596 9	774.8	3	10 222	6×10^{-4}	56

[a] the mean of the two A_e values is used with an averaged uncertainty, as attributed to an individual entry [12]
[b] KCDB values superseded by the international comparison in 1978
c mass of active solution before dilution
[d] submission used to link the 1978 CCRI(II) key comparison
[e] submission used to link the 2004 APMP key comparison
[f] the mean value and standard uncertainty of the two results submitted, as evaluated by the IRMM: A = 688.9 kBq, u = 2.6 kBq.

No earlier submission was withdrawn and no recent submission has been identified as a pilot study so the results of each NMI are eligible for Appendix B of the MRA. However, three of these results, for the CMI-IIR, IRA and the NPL have been superseded by the international comparison that was held in 1978 [3]. The IAEA no longer undertakes the metrology of activity, the PTKMR is not yet a designated institute of the Puslit KIM-LIPI, Indonesia and the AECL is not a designated laboratory of the NRC, Canada, therefore none of these results is included in the KCDB.

The BARC results are self-consistent over more than twenty years within 2.5×10^{-3}.

The results of the APMP regional comparison will be published [5]. The two laboratories to be added to the matrix of degrees of equivalence from this publication are those given in Table 1b; the VNIIM and the INER. The results $(A/m)_i$ for these laboratories are linked to the SIR through the measurement in the SIR of two ampoules of the same solution standardized by the NMIJ and the LNE-LNHB. The link is made using a normalization ratio deduced from the mean of the values in the rows indicated in Table 4a:

$$A_{ei} = (A/m)_i \times \sum_{1}^{2} \left(A_{e,\text{Link}} / (A/m)_{\text{Link}}\right) = (A/m)_i \times 24.687 \tag{1}$$

The details of the links are given in Table 4b. The uncertainties for the regional comparison linked to the SIR are comprised of the original uncertainties together with the uncertainty in the link, 7×10^{-4}, given by the standard deviation of the linking values from the NMIJ and LNE-LNHB ampoules.

Table 4b. **Results of the 2004 APMP regional comparison primary measurements of ^{134}Cs and links to the SIR**

NMI	Measurement method and acronym (see Appendix 2)	Activity * concentration measured $(A/m)_i$ / (kBq g^{-1})	Evaluation by category of relative standard uncertainty × 100		Equivalent SIR activity A_{ei} / kBq	Combined standard uncertainty $u_{c\,i}$ / kBq
			A	B		
NMIJ	4P-PC-BP-NA-GR-CO	409.5	0.18	0.08	10 104$^{\#}$	19$^{\#}$
LNE-LNHB	4P- NA-GR-00-00-00	409.9	0.03	0.18	10 124$^{\#}$	20$^{\#}$
VNIIM	4P-PC-BP-NA-GR-CO 4P- NA-GR-00-00-00	410.1 407.6	0.25 0.25		10 094**	26
INER	4P-PC-BP-NA-GR-CO	412.3	0.16		10 178	18

*referenced to 2005-02-15, 0 h UT

$^{\#}$ same results as in Table 4a

** the weighted mean of the primary results, 408.9 (5) kBq g^{-1}, is used to evaluate the linked equivalent activity.

4.1 The key comparison reference value

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

a) only primary standardized solutions are accepted, 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[1];

b) each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);

c) any outliers are identified using a reduced chi-squared test and, if necessary, excluded from the KCRV using the normalized error test with a test value of four;

d) exclusions must be approved by the CCRI(II).

The reduced data set from the SIR master-file used for the evaluation of the KCRVs following the criteria above is known as the KCRV 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 and this was the case in May 2007.

Consequently, the key comparison reference value for ^{134}Cs is 10 116 (13) kBq using the results in Table 4a from the NPL, IRA, CMI-IIR, BIPM, PTKMR, AECL (1992), NIST (2002), PTB (2004), MKEH (2004), IRMM, NMIJ, LNE-LNHB (2005), BARC (2005) and the CNEA (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 degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the key comparison reference value [2]. The degree of equivalence is expressed quantitatively in terms of the deviation from the key comparison reference value and the expanded uncertainty of this deviation ($k = 2$). The degree of equivalence between any pair of national measurement standards is expressed in terms of their difference and the expanded uncertainty of this difference and is independent of the choice of key comparison reference value.

4.2.1 *Comparison of a given NMI with the KCRV*

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

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

---

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

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

$$U_i = 2u_{D_i}, \tag{3}$$

taking correlations into account as appropriate [13].

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

$$D_{ij} = D_i - D_j = A_{e\,i} - A_{e\,j} \tag{4}$$

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

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

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

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

Table 5 shows the matrix of all the degrees of equivalence as they will appear in Appendix B of the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{ei} replaced by x_i. The introductory text is that agreed for the comparison. The graph of the first column of results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV, is shown in Figure 1 where, following the advice of the CCRI, measurements made prior to 1987 are indicated as black squares. This graphical representation indicates in part the degree of equivalence between the NMIs but does not take into account the correlations between the different NMIs. However, the matrix of degrees of equivalence shown in yellow in Table 5 does take the known correlations into account.

The results of the 1978 CCRI(II)-K2.Cs-134 international comparison have already been linked to those of the SIR through the measurement in the SIR of the BIPM ampoules of the comparison [3]. For completeness, the degrees of equivalence to the presently updated KCRV are given as the extension of the matrix in Table 5 and as the second set of values in Figure 1. The degrees of equivalence between all pairs of NMIs are also given in Table 5. The correlations associated with the distribution of the same solution in the international comparison have been ignored in the analysis as the overall uncertainties are quite large. The correlation coming from the link to the SIR has been taken into account.

The results of the 2004 APMP.RI(II)-K2.Cs-134 regional comparison, linked through the SIR measurement of the NMIJ, are given as the third extension of the matrix in Table 5 and as the third set of values in Figure 1. The correlations associated with the distribution of the same solution in the regional comparison have been ignored in the analysis as the overall uncertainties are quite large. The correlation coming from the link to the SIR through the NMIJ and the LNE-LNHB has been taken into account.

5. Conclusion

The BIPM ongoing key comparison for ^{134}Cs, BIPM.RI(II)-K1.Cs-134 currently comprises thirteen results. The results have been analysed with respect to the new KCRV determined for this radionuclide, and with respect to each other. The matrix of degrees of equivalence has been approved by the CCRI(II) and is published in the BIPM key comparison database.

The results of eight other NMIs that took part in the CCRI(II)-K2.Cs-134 comparison in 1978 are linked to the BIPM ongoing key comparison through two ampoules of the comparison measured in the SIR. These linked results are included in the matrix of degrees of equivalence approved by the CCRI(II).

The results of two other NMIs that took part in the APMP.RI(II)-K2.Cs-134 comparison in 2005 have been linked to the BIPM ongoing comparison through ampoules of the comparison solution standardized by the NMIJ and the LNE-LNHB and measured in the SIR. These linked results are included in the matrix of degrees of equivalence approved by the CCRI(II) and have enabled one NMI to update its 1978 comparison result and another NMI to be linked for the first time.

Other results may be added as and when other NMIs contribute ^{134}Cs activity measurements to this comparison or take part in other linked comparisons.

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Table 5. Introductory text for ^{134}Cs and table of degrees of equivalence

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

MEASURAND : **Equivalent activity of ^{134}Cs**

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 10 116 kBq
with a standard uncertainty, u_R = 13 kBq (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 kBq, and
$U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\Sigma u_i^2)^{1/2}$ when each laboratory has contributed to the calculation of x_R, with n the number of laboratories.

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

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-134 (1978) to BIPM.RI(II)-K1.Cs-134

The value x_i is the equivalent activity for laboratory *i* participant in CCRI(II)-K2.Cs-134
having been normalized using 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-134 with respect to the key comparison reference value is giver
by a pair of terms: $D_i = (x_i - x_R)$ and U_i, its expanded uncertainty (k = 2), both expressed in kBq.
The approximation $U_i = 2(u_i^2 + u_R^2)^{1/2}$ is used in the following table.

Linking APMP.RI(II)-K2.Cs-134 (2004) to BIPM.RI(II)-K1.Cs-134

The value x_i is the equivalent activity for laboratory *i* participant in APMP.RI(II)-K2.Cs-134
having been normalized using the value of the linking laboratories, NMIJ and LNE-LNHB (see Final report).

The degree of equivalence of laboratory *i* participant in APMP.RI(II)-K2.Cs-134 with respect to the key comparison reference value is giver
by a pair of terms: $D_i = (x_i - x_R)$ and U_i, its expanded uncertainty (k = 2), both expressed in kBq.
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*, participants in one or another of the three key comparisons, is given by a pair of terms:
$D_{ij} = D_i - D_j$ and U_{ij}, its expanded uncertainty (k = 2), both expressed in kBq. Correlations between pairs of laboratories are taken into account
as explained in Section 4.2.2 on page 14 of the Final report dated September 2007.

These statements make it possible to extend the BIPM.RI(II)-K1.Cs-134 matrices of equivalence to the other participants in CCRI(II)-K2.Cs-134 and APMP.RI(II)-K2.Cs-134

Table 5 continued

Lab *j* ⇒

Lab *i* ⇓

	BIPM		LNMRI		KRISS		CIEMAT		NIST		IRMM		PTB		MKEH		LNE-LNHB		NMIJ		BARC		CNEA		IFIN-HH				
	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}		
	/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq		/ kBq			

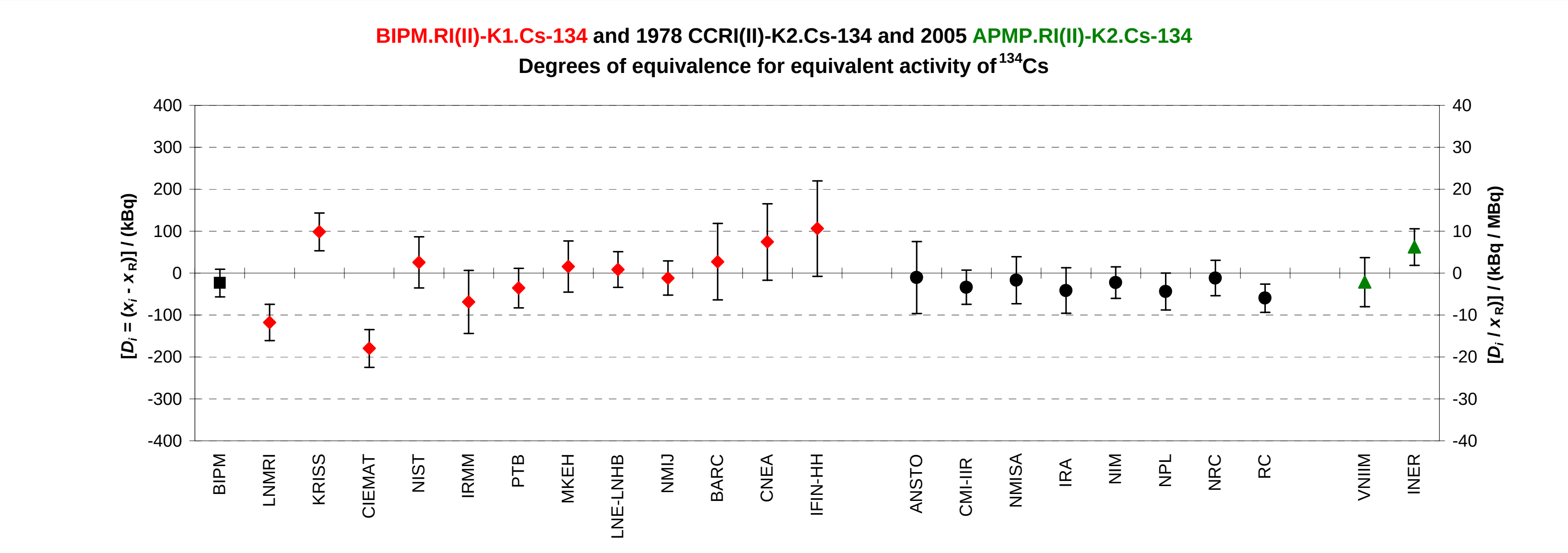
Lab *i*	D_i	U_i	BIPM		LNMRI		KRISS		CIEMAT		NIST		IRMM		PTB		MKEH		LNE-LNHB		NMIJ		BARC		CNEA		IFIN-HH	
	/ kBq		D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}
BIPM	-24	33			94	47	-122	49	156	49	-49	68	45	83	12	54	-39	68	-32	49	-12	47	-51	100	-98	100	-130	115
LNMRI	-118	43	-94	47			-216	55	62	55	-143	73	-49	87	-82	60	-133	73	-126	55	-106	54	-145	103	-192	103	-224	118
KRISS	98	45	122	49	216	55			278	57	73	74	167	88	134	61	83	74	90	57	110	55	71	104	24	104	-8	119
CIEMAT	-180	45	-156	49	-62	55	-278	57			-205	74	-111	88	-144	61	-195	74	-188	57	-168	55	-207	104	-254	104	-286	119
NIST	25	61	49	68	143	73	-73	74	205	74			94	100	61	77	10	88	17	74	37	73	-2	114	-49	114	-81	128
IRMM	-69	75	-45	83	49	87	-167	88	111	88	-94	100			-33	91	-84	100	-77	88	-57	87	-96	124	-143	124	-175	136
PTB	-36	47	-12	54	82	60	-134	61	144	61	-61	77	33	91			-51	77	-44	61	-24	60	-63	106	-110	106	-142	121
MKEH	15	61	39	68	133	73	-83	74	195	74	-10	88	84	100	51	77			7	74	27	73	-12	114	-59	114	-91	128
LNE-LNHB	8	42	32	49	126	55	-90	57	188	57	-17	74	77	88	44	61	-7	74			20	55	-19	104	-66	104	-98	119
NMIJ	-12	41	12	47	106	54	-110	55	168	55	-37	73	57	87	24	60	-27	73	-20	55			-39	103	-86	103	-118	118
BARC	27	91	51	100	145	103	-71	104	207	104	2	114	96	124	63	106	12	114	19	104	39	103			-47	136	-79	148
CNEA	74	91	98	100	192	103	-24	104	254	104	49	114	143	124	110	106	59	114	66	104	86	103	47	136			-32	148
IFIN-HH	106	114	130	115	224	118	8	119	286	119	81	128	175	136	142	121	91	128	98	119	118	118	79	148	32	148		
ANSTO	-11	86	13	85	107	90	-109	91	169	91	-36	103	58	113	25	94	-26	103	-19	91	1	90	-38	126	-85	126	-117	139
CMI-IIR	-34	41	-10	40	84	50	-132	51	146	51	-59	70	35	84	2	56	-49	70	-42	51	-22	50	-61	101	-108	101	-140	116
NMISA	-17	56	7	55	101	63	-115	64	163	64	-42	80	52	93	19	68	-32	80	-25	64	-5	63	-44	108	-91	108	-123	123
IRA	-42	54	-18	54	76	61	-140	62	138	62	-67	78	27	92	-6	66	-57	78	-50	62	-30	61	-69	107	-116	107	-148	122
NIM	-23	38	1	37	95	47	-121	49	157	49	-48	68	46	83	13	54	-38	68	-31	49	-11	47	-50	100	-97	100	-129	115
NPL	-44	44	-20	43	74	52	-142	54	136	54	-69	72	25	86	-8	58	-59	72	-52	54	-32	52	-71	103	-118	103	-150	118
NRC	-12	42	12	42	106	51	-110	52	168	52	-37	71	57	85	24	57	-27	71	-20	52	0	51	-39	102	-86	102	-118	117
RC	-60	34	-36	33	58	44	-158	46	120	46	-85	66	9	81	-24	51	-75	66	-68	46	-48	44	-87	98	-134	98	-166	114
VNIIM	-22	58	2	59	96	65	-120	66	158	66	-47	81	47	94	14	70	-37	81	-30	52	-10	52	-49	109	-96	109	-128	124
INER	62	44	86	45	180	52	-36	54	242	54	37	71	131	86	98	58	47	71	54	36	74	36	35	102	-12	102	-44	118

Lab *j* ⇒

Lab *i* ⇓

Lab *i*	D_i	U_i	ANSTO		CMI-IIR		NMISA		IRA		NIM		NPL		NRC		RC		VNIIM		INER	
	/ kBq		D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}	D_{ij} / kBq	U_{ij}
BIPM	-24	33	-13	85	10	40	-7	55	18	54	-1	37	20	43	-12	42	36	33	-2	59	-86	45
LNMRI	-118	43	-107	90	-84	50	-101	63	-76	61	-95	47	-74	52	-106	51	-58	44	-96	65	-180	52
KRISS	98	45	109	91	132	51	115	64	140	62	121	49	142	54	110	52	158	46	120	66	36	54
CIEMAT	-180	45	-169	91	-146	51	-163	64	-138	62	-157	49	-136	54	-168	52	-120	46	-158	66	-242	54
NIST	25	61	36	103	59	70	42	80	67	78	48	68	69	72	37	71	85	66	47	81	-37	71
IRMM	-69	75	-58	113	-35	84	-52	93	-27	92	-46	83	-25	86	-57	85	-9	81	-47	94	-131	86
PTB	-36	47	-25	94	-2	56	-19	68	6	66	-13	54	8	58	-24	57	24	51	-14	70	-98	58
MKEH	15	61	26	103	49	70	32	80	57	78	38	68	59	72	27	71	75	66	37	81	-47	71
LNE-LNHB	8	42	19	91	42	51	25	64	50	62	31	49	52	54	20	52	68	46	30	52	-54	36
NMIJ	-12	41	-1	90	22	50	5	63	30	61	11	47	32	52	0	51	48	44	10	52	-74	36
BARC	27	91	38	126	61	101	44	108	69	107	50	100	71	103	39	102	87	98	49	109	-35	102
CNEA	74	91	85	126	108	101	91	108	116	107	97	100	118	103	86	102	134	98	96	109	12	102
IFIN-HH	106	114	117	139	140	116	123	123	148	122	129	115	150	118	118	117	166	114	128	124	44	118
ANSTO	-11	86			23	87	6	95	31	94	12	85	33	88	1	88	49	84	11	97	-73	89
CMI-IIR	-34	41	-23	87			-17	58	8	56	-11	40	10	46	-22	44	26	36	-12	61	-96	48
NMISA	-17	56	-6	95	17	58			25	68	6	55	27	60	-5	59	43	53	5	72	-79	61
IRA	-42	54	-31	94	-8	56	-25	68			-19	54	2	58	-30	57	18	51	-20	71	-104	60
NIM	-23	38	-12	85	11	40	-6	55	19	54			21	43	-11	42	37	33	-1	59	-85	45
NPL	-44	44	-33	88	-10	46	-27	60	-2	58	-21	43			-32	47	16	40	-22	64	-106	51
NRC	-12	42	-1	88	22	44	5	59	30	57	11	42	32	47			48	38	10	62	-74	49
RC	-60	34	-49	84	-26	36	-43	53	-18	51	-37	33	-16	40	-48	38			-38	57	-122	42
VNIIM	-22	58	-11	97	12	61	-5	72	20	71	1	59	22	64	-10	62	38	57			-84	60
INER	62	44	73	89	96	48	79	61	104	60	85	45	106	51	74	49	122	42	84	60		

Figure 1 **Graph of degrees of equivalence with the KCRV for ^{134}Cs**
(as it appears in Appendix B of the CIPM MRA)



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

Red diamonds and black square : participants in BIPM.RI(II)-K1.Cs-134
Black circles : participants in CCRI(II)-K2.Cs-134 (1978)
The black colour identifies results prior to 1987
Green triangles: participants in the APMP.RI(II)-K23Cs-134

Appendix 1. Uncertainty budgets for the activity of ^{134}Cs submitted recently to the SIR

Uncertainty budget for the BARC measurement of 2005

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	10	–
weighing	–	4
background	–	1
dead time	–	4
resolving time	–	17
extrapolation of efficiency curve	42	–
decay correction	–	1
Quadratic summation	**43**	**18**
Relative combined standard uncertainty, u_c	**47**	

Uncertainty budget for the CNEA measurement of 2005

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	34	–
weighing	–	18
dead time	–	11
counting time	–	20
extrapolation of efficiency curve	12	–
input parameters	–	0.7
decay correction	–	0.4
Quadratic summation	**36**	**29**
Relative combined standard uncertainty, u_c	**46**	

Uncertainty budget for the IFIN-HH measurement of 2006

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	19	–
weighing	–	10
adsorption	–	1
beta background	–	5
gamma background	–	7.5
dead time	–	1
resolving time	–	1
impurities	–	1
extrapolation of efficiency curve	–	50
Quadratic summation	**19**	**52**
Relative combined standard uncertainty, u_c	**55**	

Appendix 2. Acronyms used to identify different measurement methods

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

Geometry	**acronym**	**Detector**	**acronym**
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop counter	PP
2π	2P	liquid scintillation counting	LS
undefined solid angle	UA	NaI(Tl)	NA
		Ge(HP)	GH
		Ge(Li)	GL
		Si(Li)	SL
		CsI(Tl)	CS
		ionization chamber	IC
		grid ionization chamber	GC
		bolometer	BO
		calorimeter	CA
		PIPS detector	PS
Radiation	**acronym**	**Mode**	**acronym**
positron	PO	efficiency tracing	ET
beta particle	BP	internal gas counting	IG
Auger electron	AE	CIEMAT/NIST	CN
conversion electron	CE	sum counting	SC
mixed electrons	ME	coincidence	CO
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with efficiency tracing	CT
X - rays	XR	anti-coincidence counting with efficiency tracing	AT
photons (x + γ)	PH	triple-to-double coincidence ratio counting	TD
photons + electrons	PE	selective sampling	SS
alpha - particle	AP	high efficiency	HE
mixture of various radiation	MX	digital coincidence counting	DC

Examples **method**	**acronym**
4π(PC)β–γ-coincidence counting	4P-PC-BP-NA-GR-CO
4π(PPC)β–γ-coincidence counting eff. trac.	4P-PP-MX-NA-GR-CT
defined solid angle α-particle counting with a PIPS detector	SA-PS-AP-00-00-00
4π(PPC)AX-γ(GeHP)-anticoincidence counting	4P-PP-MX-GH-GR-AC
4π CsI-β,AX,γ counting	4P-CS-MX-00-00-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG