Update of the BIPM comparison BIPM.RI(II)-K1.Am-241 of activity measurements of the radionuclide ^{241}Am to include the 2006 VNIIM result, links for the international 2003 CCRI(II)-K2.Am-241 comparison and links for the 2006 regional comparison COOMET.RI(II)-K2.Am-241

G. Ratel*, C. Michotte*, L. Johansson**, S. Judge**, I.A. Kharitonov#
*BIPM, **NPL, United Kingdom and #VNIIM, Russian Federation

Abstract
Since 1977, seven national metrology institutes (NMIs) have submitted eleven samples of known activity of ^{241}Am to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM). The activities ranged from about 1.9 MBq to 17 MBq. The results of the CCRI(II)-K2.Am-241 comparison held in 2003 with twenty-one participants have been linked to the BIPM.RI(II)-K1.Am-241 results using the submission to the SIR by the NPL of two ampoules of the mother solution of the CCRI(II) comparison. This has enabled four NMIs to update their earlier SIR results and fifteen NMIs, the IRMM and the BIPM to have their result published, the previous international comparison of ^{241}Am having been held over 40 years ago. The results of a COOMET.RI(II)-K2.Am-241 comparison held in 2006 have also been linked to the SIR results. This is possible through the submission to the SIR of an ampoule of the comparison by the pilot laboratory, the VNIIM. This has enabled two further NMIs to have degrees of equivalence for activity measurements for this radionuclide. The key comparison reference value (KCRV) has been updated to include the recent NPL and VNIIM results. The degree of equivalence between each equivalent activity measured in the SIR, or linked to the SIR, and the KCRV has been calculated and the results are given in the form of a matrix. A graphical presentation is also given. Degrees of equivalence between pairs of participants are similarly given as a matrix.

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].

Since 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 (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Am-241 key comparison and the results presented here update those given in [3].

The CCRI(II) held a second international comparison for activity measurements of the same ^{241}Am solution from 2001 to 2003, the CCRI(II)-K2.Am-241 (2003) comparison. The results of this comparison have been presented to the CCRI(II) , and are in publication [4]. Consequently, the opportunity has been taken to include the summary results in this report. As the previous comparison of ^{241}Am activity measurements dates from 1963 [5], the earlier results were not deemed acceptable for the KCDB.

In addition, a regional key comparison, COOMET.RI(II)-K2.Am-241, was held in 2006 for this radionuclide [6]. This was piloted by the VNIIM who sent an ampoule of the comparison solution to the SIR to make the link. Three laboratories took part in this comparison including the VNIIM. Two NMIs made primary standardizations and one NMI used a traceable secondary standard to measure the solution. The two additional NMIs are eligible to be linked to the BIPM key comparison with these new results.

2. Participants

Seven NMIs have submitted thirteen ampoules for the comparison of ^{241}Am activity measurements since 1977. In 2002, the NPL sent two ampoules of the mother solution of the CCRI(II) ^{241}Am comparison to the SIR. In addition, the VNIIM submitted one ampoule for the SIR comparison of ^{241}Am activity measurements in 2006. The laboratory details are given in Table 1a.

The 21 participants of the CCRI(II)-K2.Am-241 comparison that was held in 2003 are given in Table 1b.

Details of the three NMIs that took part in the regional comparison, COOMET.RI(II)-K2.Am-241 piloted by the VNIIM in 2006, are shown in Table 1c.

Table 1a. Details of the participants in the BIPM.RI(II)-K1.Am-241

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
OMH	MKEH	Magyar Kereskedelmi Engedéyezési Hivatal/Hungarian Trade Licensing Office	Hungary	EUROMET	1977-03-14 1979-12-13
AAEC	ANSTO	Australian Nuclear Science and Technology Organisation	Australia	APMP	1977-05-05
UVVVR	CMI-IIR	Český Metrologický Institut, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1977-05-13 1978-06-20 1979-05-18
–	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1978-03-13
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1980-10-10 2002-10-01
PSPKR	PTKMR	Pusat Teknologi Keselamatan Metrologi Radiasi	Indonesia	APMP	1989-12-01
–	VNIIM	D.I. Mendeleyev Institute for Metrology	Russian Federation	COOMET	2006-08-03 to 2007-02-01

Table 1b. Details of the participants in the 2003 CCRI(II)-K2.Am-241 to be linked to the BIPM.RI(II)-K1.Am-241 comparison

NMI	Full name	Country	Regional metrology organization
BARC	Bhabha Atomic Research Centre	India	APMP
BEV	Bundesamt für Eich- und Vermessungswesen	Austria	EUROMET
BIPM	Bureau International des Poids et Mesures	–	–
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnològicas	Spain	EUROMET
CMI-IIR	Český Metrologický Institut, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET
CNEA	Comision Nacional de Energia Atomica	Argentina	SIM
IFIN-HH	Institutul de Fizica si Inginerie Nucleara-"Horia Hulubei"	Romania	EUROMET
ININ	Instituto Nacional de Investigaciones Nucleares	Mexico	SIM
IRMM	Institute for Reference Materials and Measurements	–	EUROMET
KRISS	Korea Research Institute of Standards and Science	Republic of Korea	APMP
LNE-LNHB*	Laboratoire national de métrologie et d'essais - Laboratoire national Henri Becquerel	France	EUROMET
LNMRI	Laboratorio Nacional de Metrologia das Radiaçoes Ionizantes	Brazil	SIM
MKEH[#]	Magyar Kereskedelmi Engedéyezési Hivatal / Hungarian Trade Licensing Office	Hungary	EUROMET
NIST	National Institute of Standards and Technology	U.S.A.	SIM
NMIJ	National Metrology Institute of Japan	Japan	APMP

Continued overleaf

Table 1b continued. Details of the participants in the 2003 CCRI(II)-K2.Am-241 to be linked to the BIPM.RI(II)-K1.Am-241 comparison

NMI	Full name	Country	Regional metrology organization
NMISA§	National Metrology Institute of South Africa	South Africa	SADCMET
NPL	National Physical Laboratory	United Kingdom	EUROMET
PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET
RC	Radioisotope Centre POLATOM	Poland	EUROMET
SMU	Slovenský Metrologický Ústav	Slovakia	EUROMET
VNIIM	D.I. Mendeleyev Institute for Metrology	Russian Federation	COOMET

* formerly called BNM-LNHB
# formerly called OMH
§ formerly called CSIR-NML

Table 1c. Details of the participants in the 2006 COOMET.RI(II)-K2.Am-241 to be linked to the BIPM.RI(II)-K1.Am-241 comparison

NMI	Full name	Country	Regional metrology organization
BelGIM	Belarussian State Institute of Metrology	Belarus	COOMET
CENTIS-DMR	Centro de Isótopos. Departamento de Metrología de Radionúclidos	Cuba	COOMET
VNIIM	D.I. Mendeleyev Institute for Metrology	Russian Federation	COOMET

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 activity submitted and the relative standard uncertainties (k = 1) are given in Table 2. Full uncertainty budgets have been requested as part of the comparison protocol only since

1998. The SIR uncertainty budgets for the NPL and the VNIIM are given in Appendix 1 attached to this report.

The half-life used by the BIPM to date is 158 150 (730) d [7] which is in agreement with the value recommended by the CCRI, 158 004 (219) d [8] and with that recommended by the IAEA, 157 850 (240) d [9] used in the CCRI(II) and the COOMET comparisons. Nevertheless, in view of the long half-life of ^{241}Am, the SIR results and uncertainties do not depend on which of these the half-life values is used in the calculation.

Table 2. Standardization method of the participants for ^{241}Am

NMI	Method used and acronym (see Appendix 3)	Half-life /d	Activity / kBq	Reference date YYYY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
MKEH	4πα−γ coincidence 4P-PC-AP-NA-GR-CO	158 100 (700)	8 116 8 111	1977-03-01 12 h UT	0.14	0.30
		157 860 (180)	14 831	1979-10-01 12 h UT	0.04	0.11
ANSTO	4π(PC)α−γ coincidence 4P-PC-AP-NA-GR-CO	–	4 362	1977-04-01	0.03	0.3
CMI-IIR	4πα−γ coincidence 4P-PC-AP-NA-GR-CO	–	14 911	1977-04-24 13 h UT	0.06	0.29
		158 100	3 475	1978-04-28	0.12	0.32
			5 476	1979-04-09 11 h UT	0.12	0.32
PTB	4πα−γ coincidence 4P-PC-AP-NA-GR-CO	–	15 472	1978-01-01	0.02	0.12
NPL	Pressurized IC 4P-IC-GR-00-00-00 calibrated by 4πα-γ coincidence	–	13 120	1980-10-06	0.12	0.34
	4π(PC)α-γ coinc. 4P-PC-AP-NA-GR-CO 4π(LS)α-γ coinc. with DCC 4P-LS-AP-NA-GR-DC	[9]	10 608 10 659*	2002-12-01	0.08	0.15
PTKMR	4πα−γ coincidence 4P-??-AP-??-GR-CO	–	17 500	1989-11-15 7 h UT	0.59	–
VNIIM	4πα-γ coincidence 4P-PC-AP-NA-GR-CO	[9]	1 888.5	2006-06-01	0.13	0.02

* two ampoules submitted; these values are obtained from the NPL CCRI(II) comparison result using the 4π(PC)α-γ coincidence method only, multiplied by the dilution factor of the solution F_d = 10.0518. The result of the 4π(LS)α-γ digital coincidence measurement is numerically identical.

The standardization methods and the uncertainty budgets used in the CCRI(II) comparison are given in [4]. The standardization methods and the uncertainty budgets used in the COOMET regional comparison are given in [6].

Details regarding the solutions submitted to the SIR are shown in Table 3, including the impurities present, as identified by the laboratory. When given, the standard uncertainties on the evaluations are shown. The BIPM has a standard method available for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [10]. The CCRI(II) agreed in 1999 [11] that this method should be followed according to the protocol described in [12] when an NMI makes such a request or when there appear to be discrepancies. No gamma-emitting impurity was identified at the BIPM in the NPL solution. No impurity measurement was carried out at the BIPM for the VNIIM ^{241}Am submission.

The detail regarding the solution issued for the CCRI(II) international comparison is given in [4] together with the impurity assessments. The presence of ^{241}Pu, a quasi pure beta emitter, was identified by several participants and this may have slightly affected the results of some laboratories, depending on the measurement method used [4]. This impurity has no influence on the SIR measurement. The NPL is the only participant that identified a small amount of ^{237}Np in the CCRI(II) solution. However, this impurity has a negligible effect of the NPL SIR result.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the SIR Master file. The activity measurements for ^{241}Am now arise from 13 ampoules and the SIR equivalent activity A_{ei}, is given in Table 4a for each NMI *i*. The correction of the SIR results for the density of the solution is described in [3]. 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].

As the ionization current produced by the VNIIM ampoule is only about 0.3 pA, the SIR measurement was repeated at the BIPM several times. Consequently, the result in Table 4a is based on the weighted mean and external standard uncertainty of nine separate SIR measurements.

The results of the international CCRI(II)-K2.Am-241 comparison are in publication [4]. The nineteen NMIs, the IRMM and the BIPM to be added to the matrix of degrees of equivalence from this international comparison are those given in Table 1b. The activity of the solution was too low to be measured in the SIR so the results for these laboratories are linked to the BIPM comparison through the measurement of an aliquot of the undiluted mother solution prepared by the NPL, using the following linking equation:

$$A_{ei} = (A/m)_i \times (A_{e,NPL} / (A/m)_{NPL}) = (A/m)_i \times 7.0061 \qquad (1)$$

Table 3. Details of the solutions of ^{241}Am submitted

NMI	Chemical composition	Solvent conc. /(mol dm^{-3})	Carrier: conc. /(µg g^{-1})	Density[1] /(g cm^{-3})	Relative activity of impurity[2]
MKEH 1977	^{241}Am + Eu in HNO_3	3	Eu : 20	1.098	–
1979			Eu : 25	1.098	–
ANSTO	$^{241}Am(NO_3)_3$ in HNO_3	0.1	–	1.00	< 0.1 %
CMI-IIR 1977	^{241}Am + $SmCl_3$ in HNO_3	0.6	$SmCl_3$: 20	1.018	< 0.1 %
1978	^{241}Am + $Sm(NO_3)_3$ in HNO_3	0.1	$Sm(NO_3)_3$: 20	1.002	^{90}Sr : 0.03 %
1979		0.1	$Sm(NO_3)_3$: 20	1.002	β emitters: 0.03 % γ emitters: < 0.001 %
PTB	^{241}Am + $La(NO_3)_3$ in HNO_3	2	$La(NO_3)_3$: 50	1.06	< 0.01 %
NPL 1980	^{241}Am in HNO_3	1	–	1.03	–
2002	^{241}Am in HNO_3 §	0.5	–	1.015	^{237}Np : 9.0 (0.5) × 10^{-4} %
PTKMR	^{241}Am in HCl	0.1	–	1.000	–
VNIIM	^{241}Am in HNO_3 *	2	–	1.065	all Pu < 0.03 % ^{237}Np < 0.01 % #

[1] density value from participant if available, or estimated from the solvent concentration otherwise
[2] the ratio of the activity of the impurity to the activity of ^{241}Am at the reference date.
§ mother (undiluted) solution used in the CCRI(II)-K2.Am-241 comparison.
* solution used in the COOMET.RI(II)-K2.Am-241 comparison.
# a ^{237}Np relative activity of 0.005 % , u = 0.005 % was used to correct the SIR result. The resulting correction factor is 1.0014.

Table 4a. Result of SIR measurements of the ^{241}Am submissions

NMI	Mass of solution m_i / g	Activity submitted A_i / kBq	N° of Ra source used	SIR A_{ei} /MBq		Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / MBq
				Original value	Corrected for density		
MKEH 1977	3.604 0 3.601 7	8 116 8 111	1	2069.4 2070.4	2061.1 2062.1	17×10^{-4} 17×10^{-4}	7.8
1979	3.603 2	14 831	1	2053.8	2045.6	17×10^{-4}	4.2
ANSTO	3.623 2	4 362	1	2046.7	2046.7	19×10^{-4}	7.3
CMI-IIR 1977	3.599 95	14 910	1	2036.9	2035.4	17×10^{-4}	6.9
1978	3.722 07	3 475	1	2050.7	2050.7	19×10^{-4}	8.0
1979	3.595 39	5 476	1	2052.9	2052.9	17×10^{-4}	7.7
PTB	3.780 3 (1)	15 472	1	2063.7	2058.7	18×10^{-4}	4.5
NPL 1980	3.670 1	13 120	1	2053.7	2051.2	20×10^{-4}	8.4
2002	3.594 79 3.611 90	10 608 10 659	1	2057.4 2058.8	2056.1# 2057.5	17×10^{-4} 17×10^{-4}	5.0# 4.9
PTKMR	3.607	17 500	1	2067	2067	21×10^{-4}	13
VNIIM	3.856	1 888.5	1	2057.9	2052.6 *	35×10^{-4}	7.7 *

the mean equivalent activity value is 2056.9 (4.7) MBq.
* this result is used to make the link for the COOMET.RI(II) key comparison.

The results for the four laboratories already in the SIR and the seventeen other laboratories, all of which participated in the international comparison, are given in Table 4b. The uncertainties for the results from the international comparison linked to the SIR are comprised of the original uncertainties (uncertainty budgets are in [4]) together with the uncertainty in the link, 1.5×10^{-3}, given by the combined uncertainty of the SIR measurement of the NPL mother solution of the CCRI(II)-K2.Am-241 comparison.

The results for the CMI-IIR, NPL and the PTB that are linked through the international comparison agree within a standard uncertainty with their original SIR equivalent activity values. The difference in the linked and 1979 results for the MKEH is outside the standard uncertainties and the new linked value is closer to the KCRV. In addition it should be noted that the correction factor for density applied to all the ^{241}Am results is based on a study of A. Rytz in 1978 [3] in which he used ampoules filled with ^{241}Am solutions around 3.7 cm^3, while the 1979 ampoule of the MKEH contains only about 3.28 cm^3 of solution. Consequently, it was agreed in May 2007 that the MKEH 1979 SIR result would not be used in the KCRV.

Table 4b. Results of the 2003 international comparison measurements of ^{241}Am linked to the SIR

NMI	Activity * concentration $(A/m)_i$ / (kBq g^{-1})	Relative standard uncertainty × 100	SIR A_{ei} / MBq	Combined uncertainty $u_c(A_{ei})$ / MBq
BARC	294.97	0.34	2066.6	7.7
BEV	295.92[a]	0.47	2073	10
BIPM	294.03[b]	0.18	2060.0	4.8
CIEMAT	294.07[b]	0.24	2060.3	5.8
CMI-IIR	293.76	0.17	2058.1	4.7
CNEA	292.53	0.25	2049.5	6.0
IFIN-HH	296.9[c]	0.36	2080.1	8.1
ININ	294.2[d]	2.4	2061	50
IRMM	293.86[b]	0.05	2058.8	3.3
KRISS	294.2	0.27	2061.2	6.4
LNE-LNHB	293.77[e]	0.08	2058.2	3.5
LNMRI	296.25[b]	0.11	2075.6	3.9
MKEH	293.87	0.17	2058.9	4.7
NIST	293.31	0.18	2055.0	4.8
NMIJ	293.96	0.25	2059.5	6.0
NMISA	294.90	0.09	2066.1	3.6
NPL	293.58[c]	0.17	2056.9[f]	4.7
PTB	293.35[b]	0.18	2055.2	4.8
RC	293.71[b]	0.15	2057.8	4.4
SMU	296.6[g]	1.2	2078	25
VNIIM	293.46[b]	0.09	2056.0[h]	3.6

* referenced to 2002-12-01, 0h UTC

[a] result traceable to NPL

[b] weighted mean of results obtained using several methods.

[c] result of the 4π(PC)α-γ coincidence measurement

[d] result traceable to LNE-LNHB

[e] result of the 4π(PC)α-γ anticoincidence measurement.

[f] the mean equivalent activity value and standard uncertainty, 2056.9 (4.7) MBq, which is the same result as in Table 4a, is used to make the link for the CCRI(II) key comparison

[g] result traceable to PTB

[h] result superseded in the KCDB by the SIR result in Table 4a.

As the CCRI comparison is more recent for the CMI-IIR, MKEH and the PTB, the earlier SIR results have been superseded by the values in Table 4b for the results presented in the KCDB.

The VNIIM result linked through the CCRI(II) comparison agrees within a standard uncertainty with its recent SIR equivalent activity value given in Table 4a.

The results of the regional COOMET.RI(II)-K2.Am-241 comparison have been published [6]. The two laboratories to be added to the matrix of degrees of equivalence from this publication are those given in Table 1c together with the pilot laboratory. The results $(A/m)_i$ for these laboratories are linked to the SIR through the measurement in the SIR of an ampoule of the same solution standardized by the VNIIM. The link is made using a normalization ratio deduced from the line indicated in Table 4a:

$$A_{ei} = (A/m)_i \times (A_{e,\mathrm{VNIIM}} / (A/m)_{\mathrm{VNIIM}}) = (A/m)_i \times 4.1910 \qquad (2)$$

The results of the links are given in Table 4c. The uncertainties for the regional comparison linked to the SIR are comprised of the original uncertainties together with the uncertainty in the link, 3.5×10^{-3}, given by the uncertainty of the SIR measurement of the VNIIM solution of the COOMET.RI(II)-K2.Am-241.

Table 4c. Results of the 2006 COOMET regional comparison of ^{241}Am 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} / MBq	Combined standard uncertainty $u_{c\,i}$ / MBq
			A	B		
BelGIM	HPGe γ-spectrometry traceable to the VNIIM UA-GH-GR-00-00-00	491.5	0.44	1.0*	2060	24
CENTIS -DMR	Weighted mean of LS counting 4P-LS-AP-00-00-00 and a calibrated HPGe spectrometer traceable to the OMH and CIEMAT UA-GH-GR-00-00-00	487.4	0.55		2043	13
VNIIM	4πα-γ coincidence 4P-PC-AP-NA-GR-CO	489.8	0.13		2052.6#	7.7

*referenced to 2006-06-01 0:00 h UT
same result as in Table 4a
* revised in 2011 to match the details in the COOMET report, *Metrologia Technical Supplement,s* 2007, **44**, 06001

No recent submission has been identified as a pilot study so the results of each NMI are normally eligible for Appendix B of the MRA. The result from the PTKMR is not included as this is not yet a designated laboratory of the Puslit KIM-LIPI, Indonesia.

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 used for the evaluation of the KCRVs is known as the KCRV file and is the reduced data set from the SIR master-file. Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are only made by the CCRI(II), normally during one of its biennial meetings.

Consequently, the KCRV for ^{241}Am, as approved by the CCRI(II) in May 2007, has changed from 2053.7 (3.3) MBq [3] to the new value of 2055.8 (2.8) MBq using the results in Table 4a from the ANSTO, CMI-IIR (1979), PTB, NPL (2002), PTKMR and the VNIIM.

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.

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

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

$$D_i = A_{ei} - \text{KCRV} \tag{2}$$

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 over 20 years ago 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 2003 CCRI(II)-K2.Am-241 international comparison, linked through the SIR measurement of the undiluted mother solution of the comparison, are given as the extension of the matrix in Table 5 and as the second set of values in Figure 1. The correlation associated with the distribution of the same solution in the international comparison has been ignored in the analysis as the overall uncertainties are quite large. The correlations coming from the link to the SIR through the NPL and the traceability, of the BEV to the NPL, of the SMU to the PTB and of the ININ to the LNE-LNHB, have been taken into account.

The results of the 2006 COOMET.RI(II)-K2.Am-241 regional comparison, linked through the SIR measurement of the COOMET comparison solution submitted by the VNIIM, are given as the further extension of the matrix in Table 5 and as the third set of values in Figure 1. The correlation associated with the distribution of the same solution in the regional comparison has been ignored in the analysis as the overall uncertainties are quite large. The correlations coming from the link to the SIR through the VNIIM and the traceability of the BelGIM to the VNIIM have been taken into account.

Conclusion

The BIPM ongoing key comparison for ^{241}Am, BIPM.RI(II)-K1.Am-241 now comprises just three results in the KCDB as three previous entries have been superseded by the CCRI international comparison. The latest submissions from the NPL and the VNIIM have been analysed with respect to the new KCRV determined for this radionuclide, and with respect to every other previous submission. The results for the NPL and the VNIIM in this ongoing comparison agree with the KCRV within a standard uncertainty. 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 the 2003 international comparison, CCRI(II)-K2.Am-241 have been linked to the BIPM ongoing comparison through an ampoule of the undiluted mother solution of the comparison measured in the SIR. This has enabled three NMIs to update their earlier SIR results and fourteen NMIs, the IRMM and the BIPM to have results published for ^{241}Am, as this radionuclide was last compared internationally in 1963.

The results of two other NMIs that took part in the COOMET.RI(II)-K2.Am-241 comparison in 2006 have been linked to the BIPM ongoing comparison through an ampoule of the comparison solution measured in the SIR.

All these linked results are included in the matrix of degrees of equivalence approved by the CCRI(II). Other results may be added as and when other NMIs contribute ^{241}Am activity measurements to this comparison or take part in other linked comparisons.

Acknowledgements

The authors would like to thank all the NMIs for their participation in the international comparison and in the regional comparison, Mr Sammy Courte for the recent SIR measurements and Dr P.J. Allisy-Roberts of the BIPM for editorial assistance.

References

[1] Ratel G., The Système International de Référence and its application in key comparisons, *Metrologia*, 2007, **44**(4), S7-S16

[2] MRA: *Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes*, International Committee for Weights and Measures, 1999, 45 pp. http://www.bipm.org/pdf/mra.pdf.

[3] Ratel G., Michotte C., BIPM comparison BIPM.RI(II)-K1.Am-241 of the activity measurements of the radionuclide ^{241}Am *Metrologia*, 2003, **40**, *Tech. Suppl.*, 06001

[4] Ratel G., International comparison of activity measurements of a solution of ^{241}Am, *Metrologia*, **XX**, *Tech. Suppl.*, 0600XX, in preparation.

[5] Rytz A., Rapport sur la comparaison internationale de ^{241}Am, juillet 1963, *Rapport BIPM* 22 Décembre 1964.

[6] Kharitonov I.A., Zanevsky A.V., Milevski V., Ivaniukovich A., Oropesa P., Moreno Y., Measurement of the activity concentration of the radionuclide Am-241 in a solution, *Metrologia*, 2007, **44**, *Tech. Suppl.*, 06001.

[7] Martin M.J. and Blichert-Toft P.H., Radioactive atoms. Auger-electron, α-, β-, γ-, and x-ray data, *Nuclear Data Tables* **A8 (1-2)**, 1970, 145.

[8] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Helmer R., Nichols A., Schönfeld E., Dersch R., Table of radionuclides, *BIPM Monographie* 5, 2004, volume 2.

[9] IAEA-TECDOC-619, X-ray and gamma-ray standards for detector calibration, (1991), Vienna, IAEA.

[10] Michotte C., Efficiency calibration of the Ge(Li) detector of the BIPM for SIR-type ampoules, *Rapport BIPM*-1999/03, 15 pp.

[11] *Comité Consultatif pour les Étalons de Mesures des Rayonnements Ionisants* 16th meeting (1999), 2001, CCRI(II) 81-82.

[12] Michotte C., Protocol on the use of the calibrated spectrometer of the BIPM for the measurement of impurities in ampoules submitted to the SIR, *CCRI(II)/01-01*, 2001, 2 pp.

[13] Ratel G., Evaluation of the uncertainty of the degree of equivalence, 2005, *Metrologia* **42**, 140-144..

Table 5. Table of degrees of equivalence and introductory text for ^{241}Am

Key comparison BIPM.RI(II)-K1.Am-241

MEASURAND : **Equivalent activity of ^{241}Am**

Key comparison reference value: the SIR reference value x_R for this radionuclide is 2055.8 MBq, with a standard uncertainty, u_R = 2.8 MBq (see section 4.1 of the Report).

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
$U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\Sigma u_i^2)^{1/2}$ 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 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.Am-241 to BIPM.RI(II)-K1.Am-241

The value x_i is the equivalent activity for laboratory i participant in CCRI(II)-K2.Am-241 having been normalized using the SIR measurement of the NPL undiluted solution of the CCRI(II)-K2 comparison.

The degree of equivalence of laboratory i participant in CCRI(II)-K2. 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 as none of these laboratories contributed to the KCRV.

The degree of equivalence between two laboratories i and j, one participant in BIPM.RI(II)-K1.Am-241 and one in CCRI(II)-K2.Am-241, or both participants in CCRI(II)-K2.Am-241, is given by a pair of terms expressed in MBq: $D_{ij} = D_i - D_j$ and U_{ij}, its expanded uncertainty (k = 2), approximated by $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_l^2)^{1/2}$ with l referring to the link when each laboratory is from the CCRI or is the linking laboratory and f is the correlation coefficient.

Linking COOMET.RI(II)-K2.Am-241 to BIPM.RI(II)-K1.Am-241

The value x_i is the equivalent activity for laboratory i participant in COOMET.RI(II)-K2.Am-241 having been normalized using the SIR measurement of the VNIIM solution of the COOMET.RI(II)-K2 comparison.

The degree of equivalence of laboratory i participant in COOMET.RI(II)-K2. 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 as these laboratories did not contribute to the KCRV.

The degree of equivalence between two laboratories i and j, one participant in BIPM.RI(II)-K1.Am-241 or in CCRI(II)-K2.Am-241 and one in COOMET.RI(II)-K2.Am-241, is given by a pair of terms expressed in MBq: $D_{ij} = D_i - D_j$ and U_{ij}, its expanded uncertainty (k = 2), approximated by $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_l^2)^{1/2}$ with l referring to the link when the other laboratory is from the COOMET or is the linking laboratory and f is the correlation coefficient.

These statements make it possible to extend the BIPM.RI(II)-K1.Am-241 matrices of equivalence to all participants in the CCRI(II)-K2.Am-241 and the COOMET.RI(II)-K2.Am-241 comparisons.

Table 5 continued

Lab *j* ⇒

Lab *i* ⇓

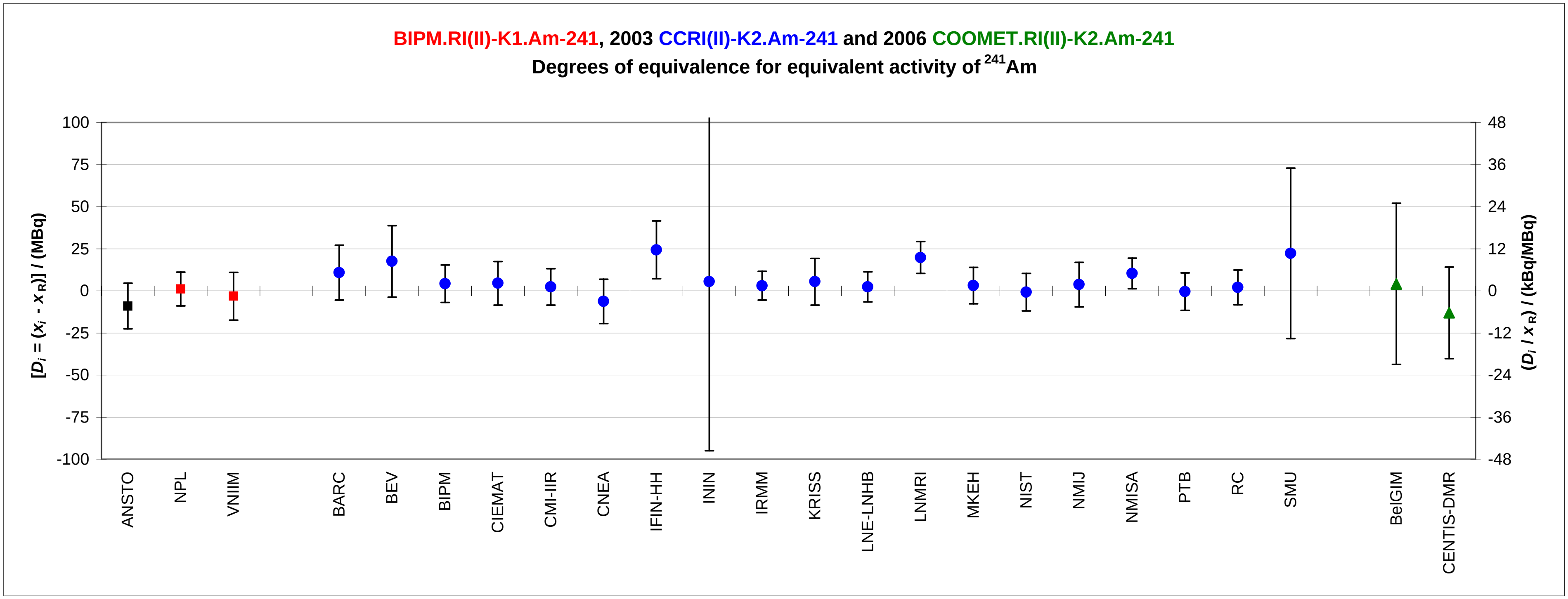
Lab *i*	D_i	U_i	ANSTO		NPL		VNIIM		BARC		BEV		BIPM		CIEMAT		CMI-IIR		CNEA		IFIN-HH		ININ		IRMM	
	/ MBq		D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq
ANSTO	-9	14			-10	17	-6	21	-20	21	-27	25	-13	18	-14	19	-11	17	-3	19	-33	22	-15	101	-12	16
NPL	1	10	10	17			4	18	-10	16	-16	19	-3	10	-3	12	-1	10	7	12	-23	17	-4	100	-2	7
VNIIM	-3	14	6	21	-4	18			-14	22	-21	26	-7	18	-8	19	-6	18	3	19	-28	22	-9	102	-6	17
BARC	11	16	20	21	10	16	14	22			-7	24	7	16	6	17	8	16	17	17	-14	21	5	101	8	14
BEV	17	21	27	25	16	19	21	26	7	24			13	21	13	22	15	21	24	22	-7	25	12	102	14	20
BIPM	4	11	13	18	3	10	7	18	-7	16	-13	21			0	12	2	10	11	13	-20	17	-1	100	1	8
CIEMAT	5	13	14	19	3	12	8	19	-6	17	-13	22	0	12			2	12	11	14	-20	18	-1	101	1	10
CMI-IIR	2	11	11	17	1	10	6	18	-8	16	-15	21	-2	10	-2	12			9	12	-22	17	-3	100	-1	7
CNEA	-6	13	3	19	-7	12	-3	19	-17	17	-24	22	-11	13	-11	14	-9	12			-31	18	-12	101	-9	10
IFIN-HH	24	17	33	22	23	17	28	22	14	21	7	25	20	17	20	18	22	17	31	18			19	101	21	15
ININ	5	101	15	101	4	100	9	102	-5	101	-12	102	1	100	1	101	3	100	12	101	-19	101			2	100
IRMM	3	9	12	16	2	7	6	17	-8	14	-14	20	-1	8	-1	10	1	7	9	10	-21	15	-2	100		
KRISS	5	14	14	19	4	13	9	20	-5	18	-12	22	1	13	1	15	3	13	12	15	-19	19	0	101	2	11
LNE-LNHB	2	9	11	16	1	8	6	17	-8	14	-15	20	-2	8	-2	10	0	8	9	11	-22	15	-3	100	-1	4
LNMRI	20	10	29	17	19	8	23	17	9	15	2	20	16	9	15	11	17	8	26	11	-5	16	14	100	17	5
MKEH	3	11	12	17	2	10	6	18	-8	16	-14	21	-1	10	-1	12	1	10	9	12	-21	17	-2	100	0	7
NIST	-1	11	8	17	-2	10	2	18	-12	16	-18	21	-5	10	-5	12	-3	10	5	13	-25	17	-6	100	-4	8
NMIJ	4	13	13	19	3	13	7	20	-7	17	-14	22	0	13	-1	14	1	12	10	15	-21	18	-2	101	1	11
NMISA	10	9	19	16	9	8	13	17	0	15	-7	20	6	8	6	11	8	8	17	11	-14	15	5	100	7	4
PTB	-1	11	9	17	-2	10	3	18	-11	16	-18	21	-5	10	-5	12	-3	10	6	13	-25	17	-6	100	-4	8
RC	2	10	11	17	1	9	5	18	-9	15	-15	20	-2	10	-3	12	0	9	8	12	-22	16	-4	100	-1	7
SMU	22	51	31	52	21	50	25	53	11	52	5	54	18	50	18	51	20	50	29	51	-2	52	17	112	19	50
BelGIM	4	48	13	50	3	48	7	45	-7	50	-13	52	0	49	0	49	2	48	10	49	-20	50	-1	111	1	48
CENTIS-DMR	-13	27	-4	30	-14	28	-10	23	-24	31	-31	34	-17	28	-18	29	-15	28	-7	29	-37	31	-19	104	-16	27

Lab *j* ⇒

Lab *i* ⇓

Lab *i*	D_i	U_i	KRISS		LNE-LNHB		LNMRI		MKEH		NIST		NMIJ		NMISA		PTB		RC		SMU		BelGIM		CENTIS-DMR	
	/ MBq		D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq	D_{ij}	U_{ij} / MBq
ANSTO	-9	14	-14	19	-11	16	-29	17	-12	17	-8	17	-13	19	-19	16	-9	17	-11	17	-31	52	-13	50	4	30
NPL	1	10	-4	13	-1	8	-19	8	-2	10	2	10	-3	13	-9	8	2	10	-1	9	-21	50	-3	48	14	28
VNIIM	-3	14	-9	20	-6	17	-23	17	-6	18	-2	18	-7	20	-13	17	-3	18	-5	18	-25	53	-7	45	10	23
BARC	11	16	5	18	8	14	-9	15	8	16	12	16	7	17	0	15	11	16	9	15	-11	52	7	50	24	31
BEV	17	21	12	22	15	20	-2	20	14	21	18	21	14	22	7	20	18	21	15	20	-5	54	13	52	31	34
BIPM	4	11	-1	13	2	8	-16	9	1	10	5	10	0	13	-6	8	5	10	2	10	-18	50	0	49	17	28
CIEMAT	5	13	-1	15	2	10	-15	11	1	12	5	12	1	14	-6	11	5	12	3	12	-18	51	0	49	18	29
CMI-IIR	2	11	-3	13	0	8	-17	8	-1	10	3	10	-1	12	-8	8	3	10	0	9	-20	50	-2	48	15	28
CNEA	-6	13	-12	15	-9	11	-26	11	-9	12	-5	13	-10	15	-17	11	-6	13	-8	12	-29	51	-10	49	7	29
IFIN-HH	24	17	19	19	22	15	5	16	21	17	25	17	21	18	14	15	25	17	22	16	2	52	20	50	37	31
ININ	5	101	0	101	3	100	-14	100	2	100	6	100	2	101	-5	100	6	100	4	100	-17	112	1	111	19	104
IRMM	3	9	-2	11	1	4	-17	5	0	7	4	8	-1	11	-7	4	4	8	1	7	-19	50	-1	48	16	27
KRISS	5	14			3	12	-14	12	2	13	6	13	2	15	-5	12	6	13	3	13	-17	51	1	49	19	30
LNE-LNHB	2	9	-3	12			-17	6	-1	8	3	8	-1	11	-8	5	3	8	0	7	-20	50	-2	48	15	28
LNMRI	20	10	14	12	17	6			17	8	21	9	16	11	9	6	20	9	18	8	-2	50	16	48	33	28
MKEH	3	11	-2	13	1	8	-17	8			4	10	-1	12	-7	8	4	10	1	9	-19	50	-1	48	16	28
NIST	-1	11	-6	13	-3	8	-21	9	-4	10			-5	13	-11	8	0	10	-3	10	-23	50	-5	49	12	28
NMIJ	4	13	-2	15	1	11	-16	11	1	12	5	13			-7	11	4	13	2	12	-18	51	0	49	17	29
NMISA	10	9	5	12	8	5	-9	6	7	8	11	8	7	11			11	8	8	7	-12	50	6	48	23	28
PTB	-1	11	-6	13	-3	8	-20	9	-4	10	0	10	-4	13	-11	8			-3	10	-23	49	-5	49	13	28
RC	2	10	-3	13	0	7	-18	8	-1	9	3	10	-2	12	-8	7	3	10			-20	50	-2	48	15	28
SMU	22	51	17	51	20	50	2	50	19	50	23	50	18	51	12	50	23	49	20	50			18	69	35	57
BelGIM	4	48	-1	49	2	48	-16	48	1	48	5	49	0	49	-6	48	5	49	2	48	-18	69			17	51
CENTIS-DMR	-13	27	-19	30	-15	28	-33	28	-16	28	-12	28	-17	29	-23	28	-13	28	-15	28	-35	57	-17	51		

Figure 1. Graph of degrees of equivalence with the KCRV for ^{241}Am
(as it appears in Appendix B of the MRA)



N.B. The right hand scale is indicative only.

Appendix 1. Uncertainty budgets for the activity of ^{241}Am submitted to the SIR

Uncertainty budgets for the NPL measurement of 2002

4πα(PC)-γ coincidence method

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	6	–
weighing	–	5
adsorption	–	3
dead time	–	1
pile-up	–	< 1
resolving time	–	0.5
Gandy effect	–	< 0.1
counting time	–	< 1
extrapolation of efficiency curve	–	13
decay correction	–	3
background	5	–
impurities	–	0.1
Quadratic summation	**7.8**	**14.6**
Relative combined standard uncertainty, u_c	**17**	

Uncertainty budget for the VNIIM measurement of 2006

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	0.6	–
weighing	9	–
dead time	–	0.2
resolving time	1.2	–
counting time	–	0.01
decay correction	–	0.001
background	9	–
impurities	–	1.5
Quadratic summation	**12.8**	**1.5**
Relative combined standard uncertainty, u_c	**13**	

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 radiations	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(Ge(HP))-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