

**BIPM comparison BIPM.RI(II)-K1.Y-88 of**  
**activity measurements of the radionuclide <sup>88</sup>Y**  
**and links for the 2000 regional comparison APMP.RI(II)-K2.Y-88**

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## **Abstract**

Since 1976, twelve national metrology institutes (NMI) and two other laboratories have submitted thirty-five samples of known activity of <sup>88</sup>Y 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.Y-88. The activities ranged from about 400 kBq to 29 MBq. The degrees of equivalence between each equivalent activity measured in the SIR and the key comparison reference value (KCRV) have been calculated and the results are given in the form of a matrix for ten NMIs. A graphical presentation is also given. The results of an APMP regional comparison, comparison identifier APMP.RI(II)-K2.Y-88, completed in 2000 for this radionuclide have been linked to the SIR results through that of the NMIJ. This has enabled six other NMIs to have degrees of equivalence in the KCDB and two others to supersede their earlier result in the SIR.

## **1. Introduction**

The SIR for activity measurements of  $\gamma$ -ray-emitting radionuclides was established in 1976. Each NMI may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid form. For radioactive gases, a different standard ampoule is used. 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 <sup>226</sup>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 2003, the SIR has measured 849 ampoules to give 615 independent results for 62 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key

comparison database (KCDB) of the Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Y-88 key comparison.

In addition, an APMP comparison for this radionuclide, APMP.RI(II)-K2.Y-88, was held in 2000 with the NMIJ as the pilot laboratory. Although fourteen laboratories took part in this comparison, only eight NMIs in addition to the NMIJ are currently eligible to be linked to the BIPM key comparison.

## 2. Participants

Twelve NMIs and two other laboratories have submitted thirty-five ampoules for the comparison of  $^{88}\text{Y}$  activity measurements since 1976. One other ampoule was submitted but as it was not well sealed, the result has not been recorded. One participant ultimately withdrew an earlier result. The laboratory details are given in Table 1a. In cases where the laboratory has changed its name since the original submission, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB. The AECL was an invited participant in various SIR comparisons, as in the early years, J.G.V. Taylor of the AECL was a personal member of the predecessor to the CCRI(II).

**Table 1a. Details of the participants in the BIPM.RI(II)-K1.Y-88**

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1976-11-19 2000-03-15
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1977-03-25 1978-04-17 1986-06-17 1999-11-17
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1977-05-17
ASMW*	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1977-06-13 1980-06-13 1988-11-09 1999-12-01

\* another laboratory in the country

Table 1a continued overleaf.

**Table 1a continued. Details of the participants in the BIPM.RI(II)-K1.Y-88**

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
–	OMH	Országos Mérésügyi Hivatal	Hungary	EUROMET	1977-09-23 1981-12-04 1985-10-29 1993-04-09
AAEC	ANSTO	Australian Nuclear Science and Technology Organisation	Australia	APMP	1977-09-29
IAEA	–	International Atomic Energy Agency	–	–	1978-05-22
IER	IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	1979-10-05 2001-06-13
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1980-10-24 2002-06-19
AECL	–	Atomic Energy of Canada Ltd	Canada	–	1986-11-27
LMRI LPRI	BNM-LNHB	Bureau national de métrologie-Laboratoire national Henri Becquerel	France	EUROMET	1987-02-05 and 1987-02-13 1994-04-18
–	VNIIM	D.I. Mendeleev Institute for Metrology	Russian Federation	COOMET	1993-01-05
–	LNMRI	Laboratorio Nacional de Metrologia das Radiações Ionizantes	Brazil	SIM	1999-02-09
–	BEV	Bundesamt für Eich- und Vermessungswesen	Austria	EUROMET	2001-09-26

The eight eligible NMIs and two other institutes that took part in the APMP regional comparison, APMP.RI(II)-K2.Y-88 in 2000 are shown in Table 1b.

**Table 1b. Details of the participants linked from the APMP.RI(II)-K2.Y-88 comparison of 2000**

<b>NMI</b>	<b>Full name</b>	<b>Country</b>
ANSTO <sup>+</sup>	Australian Nuclear Science and Technology Organisation	Australia
BARC	Bhabha Atomic Research Centre	India
CNEA	Comisión Nacional de Energía Atómica	Argentina
INER	Institute of Nuclear Energy Research	Chinese Taipei
KRISS	Korea Research Institute of Standards and Science	Korea
LNMRI <sup>+</sup>	Laboratorio Nacional de Metrologia das Radiações Ionizantes	Brazil
NIM	National Institute of Metrology	China
OAP*	Office of Atoms for Peace	Thailand
MINT**	Malaysian Institute for Nuclear Technology Research	Malaysia
P3KRBiN ***	Pusat Penelitian & Pengembangan Keselamatan Radiasi & Biomedika Nuklir	Indonesia

<sup>+</sup> updating entries in the BIPM(II)-K1.Y-88 comparison

\* designated by the National Institute of Metrology, Thailand (NIMT)

\*\* not currently designated by the National Metrology Laboratory of the Standards and Industrial Research Institute of Malaysia (NML-SIRIM)

\*\*\* not currently designated by the Research Center for Calibration, Instrumentation and Metrology – Indonesian Institute of Sciences (Puslit KIM-LIPI).

### 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. The list of acronyms used to summarize the methods is given in Appendix 3. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. When submitted by the NMIs, the uncertainty budgets are given in Appendix 1 attached to this report, as is the case for the CMI-IIR, PTB, NMIJ, IRA, BEV and the NIST.

The half-life used by the BIPM is 106.60 (4) d [3]. This is in agreement with 106.630 (25) d, the value recommended by the IAEA [4]. The SIR data could be revised using the IAEA half-life. However, the updated degrees of equivalence would not differ significantly as most of the SIR measurements were performed within two months following the reference date. In the extreme case of four months, for the IRA (1979) ampoule, the relative change in  $A_e$  is about  $2 \times 10^{-4}$ .

Details of the standardization methods used in the APMP comparison may be obtained from [5].

**Table 2. Standardization methods of the participants for  $^{88}\text{Y}$**

NMI	Method used and acronym (see Appendix 3)	Half-life	Activity $A_i$ / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
NMIJ	4 $\pi$ x- $\gamma$ coincidence 4P-PC-MX-NA-GR-CO	–	926.8 934.7 <sup>†</sup>	76-11-01 12 h UT	0.07	0.56
	4 $\pi$ (e,x)- $\gamma$ coinc. 4P-PC-MX-NA-GR-CO	106.65 d	1 656.5	00-04-01 0 h UT	0.22	0.19
CMI-IIR	4 $\pi$ (e,x)- $\gamma$ coincidence 4P-PP-MX-NA-GR-CO	–	28 640	77-03-04 13 h UT	0.10	1.42
		107 d	3 618	78-03-06 11 h UT	0.20	0.63
		106.6 d	16 950	86-05-14 12 h UT	0.05	0.23
	4 $\pi$ e <sub>x</sub> - $\gamma$ coincidence 4P-PC-AE-NA-GR-CO	106.6 d	5 168	99-11-15 12 h UT	0.10	0.36 <sup>††</sup>
NPL	Pressurized 4 $\pi$ ionization chamber 4P-IC-GR-00-00-00*	–	1 911.7 1 779.0 <sup>†</sup>	77-06-01 0 h UT	0.03	0.35
PTB	4 $\pi$ PC- $\gamma$ coincidence 4P-PC-MX-NA-GR-CO	–	2 939.4 2 914.1 <sup>†</sup>	77-05-01 0 h UT	0.02	0.05
		–	10 004 9 944 <sup>†</sup>	80-04-20 12 h UT	0.06	0.07
	Pressurized 4 $\pi$ ionization chamber 4P-IC-GR-00-00-00 **	–	3 431.5	88-10-01 0 h UT	0.04	0.21
	4 $\pi$ PC(EC)- $\gamma$ coinc. 4P-PC-MX-NA-GR-CO	[4]	5 383.8	99-11-01 0 h UT	0.06	0.10

continued overleaf.

**Table 2 continued. Standardization methods of the participants for  $^{88}\text{Y}$** 

NMI	Method used and acronym (see Appendix 3)	Half-life	Activity $A_i$ / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
OMH	4 $\pi$ (e,x)- $\gamma$ coincidence 4P-PP-MX-NA-GR-CO	[3]	1 355.8 1 355.0 <sup>†</sup>	77-10-01 12 h UT	0.21	0.61
			2 233	81-10-01 12 h UT	0.05	0.52
		106.612 (14) d [6]	3 196	85-11-01 12 h UT	0.03	0.29
		[4]	1 000.4	93-04-01 12 h UT	0.05	0.25
ANSTO	4 $\pi$ (e,x)- $\gamma$ coinc. 4P-PC-MX-NA-GR-CO	—	472.6	77-09-01 0 h UT	0.18	0.35
IAEA	Pressurized 4 $\pi$ ionization chamber 4P-IC-GR-00-00-00 calibrated by the CMI-IIR 1978 measurement	—	3 638	78-03-06 11 h UT	0.20	0.63
IRA	4 $\pi$ PC(e,x)- $\gamma$ coinc. 4P-PC-MX-NA-GR-CO	—	5 092 5 098 <sup>†</sup>	79-06-01 0 h UT	0.03	0.41
	4 $\pi$ (e,x)- $\gamma$ coinc. 4P-PC-MX-NA-GR-CO and 4 $\pi\gamma$ counting 4P-NA -GR-00-00-HE	2559.60 (96) h	861.3	01-07-01 12 h UT	0.05	0.22
NIST	4 $\pi$ (e+x)- $\gamma$ anti-coinc. 4P-PP-MX-NA-GR-AC	—	904.46	80-09-01 17 h UT	0.03	0.05
	Pressurized 4 $\pi$ ionization chamber <sup>†</sup> 4P-IC-GR-00-00-00	106.65 (4) d	1 187.5	02-06-01 17 h UT	0.05	0.20
AECL	4 $\pi$ (PC)- $\gamma$ coincidence 4P-PC-MX-NA-GR-CO	—	601.0 1 008.2 <sup>†</sup>	86-09-01 17 h UT	0.07	0.29

continued overleaf.

**Table 2 continued. Standardization methods of the participants for <sup>88</sup>Y**

NMI	Method used and acronym (see Appendix 3)	Half-life / d	Activity $A_i$ / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
BNM-LNHB	4 $\pi$ x- $\gamma$ coincidence 4P-PP-XR-NA-GR-CO	–	456.75	86-12-23 12 h UT	0.10	0.02
			459.07 <sup>†</sup>			
			2 822	94-02-25 12 h UT	0.12	0.30
VNIIM	x <sub>K</sub> (NaI(Tl))- $\gamma$ coincidence UA-NA-XR-NA-GR-CO	–	3 763	92-10-09 12 h UT	0.18	0.28
LNMRI	4 $\pi$ PC $\beta$ - $\gamma$ (NaI(Tl)) 4P-PC-BP-NA-GR-CO	106.62 (2) d	400.8	98-12-01 0 h UT	0.16	0.38
BEV	Pressurized 4 $\pi$ ionization chamber calibrated by the NPL 4P-IC-GR-00-00-00	106.630 d	1 230.2	01-09-01 0 h UT	0.06	0.37

<sup>†</sup> two ampoules submitted

\* calibrated using a primary standard of <sup>88</sup>Y in 1977 using 4P-PC-MX-NA-GR-CO

\*\* calibrated by 4 $\pi$ PC- $\gamma$  (absorber) 4P-PC-MX-NA-GR-CO and 4 $\pi$ PPC- $\gamma$  (discriminator) 4P-PP-MX-NA-GR-CO

<sup>+</sup> calibrated in 1980 by 4 $\pi$ (e+x)- $\gamma$  anti-coincidence 4P-PP-MX-NA-GR-AC

<sup>††</sup> original registered value of 0.6 % was not a quadratic sum of the components.

Details regarding the solution 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. Recently the BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [7]. The CCRI(II) agreed in 1999 [8] that this method should be followed according to the protocol described in [9] when an NMI makes such a request or when there appear to be discrepancies.

Details of the solution issued for the APMP comparison are given in [5] and indicated in Table 3.

**Table 3. Details of the solutions of <sup>88</sup>Y submitted to the SIR**

NMI	Chemical composition	Solvent conc. / (mol dm <sup>-3</sup> )	Carrier: conc. / (μg g <sup>-1</sup> )	Density / (g cm <sup>-3</sup> )	Relative activity of impurity *
NMIJ	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 50	–	–
	YCl <sub>3</sub> in HCl †	0.1	YCl <sub>3</sub> : 50	1.01	–
CMI-IIR	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 20	1	< 0.1 %
		0.08	YCl <sub>3</sub> : 20	–	< 0.2 %
		1	YCl <sub>3</sub> : 20	–	< 0.1 %
NPL	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 50	1.001	–
PTB	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 40	–	–
	Y(NO <sub>3</sub> ) <sub>3</sub> in HNO <sub>3</sub>	0.1	Y(NO <sub>3</sub> ) <sub>3</sub> : 20	1.0015	< 0.05 %
	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 40	1.00	–
YCl <sub>3</sub> : 20			0.999	–	
OMH	Y in HCl	0.1	Y: 25	–	< 0.04 %
	YCl <sub>3</sub> in HCl	0.1	Y: 25	–	–
	Y in HCl	0.1	Y: 25	–	<sup>57</sup> Co : 0.03 (1) % ††
ANSTO	YCl <sub>3</sub> in HCl	1.0	< 100	1.00	–
IAEA	YCl <sub>3</sub> in HCl	0.08	YCl <sub>3</sub> : 20	–	< 0.2 %
IRA	Y <sup>+++</sup> in HCl	0.1	Y <sup>+++</sup> : 7.4	–	–
			Y <sup>+++</sup> : 20	0.999 (6)	–
NIST	Y <sup>+++</sup> in HCl	0.5	Y <sup>+++</sup> : 15	1.01	–
	YCl <sub>3</sub> in HCl	0.4	YCl <sub>3</sub> : 70	1.005 (1)	–
AECL	<sup>88</sup> YCl <sub>3</sub> in HCl	0.1	–	1.00	–
BNM-LNHB	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 10	0.999	< 0.01 %
	YCl <sub>3</sub> .H <sub>2</sub> O in HCl	1	YCl <sub>3</sub> .H <sub>2</sub> O: 100	1.016	–
VNIIM	YCl <sub>3</sub> in HCl	0.1	Y : 20	1.001	< 0.1 %
LNMRI	La <sub>2</sub> O <sub>3</sub> in HCl	0.1	La <sub>2</sub> O <sub>3</sub> : 150	–	–
BEV	YCl <sub>3</sub> in HCl	0.1	YCl <sub>3</sub> : 40	1.0	–

\* the ratio of the activity of the impurity to the activity of <sup>88</sup>Y at the reference date

† the same solution as used in the APMP comparison

†† negligible influence on the SIR measurement.

#### 4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The activity measurements for  $^{88}\text{Y}$  arise from thirty-five 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 and are used in the KCDB and all references in this report.

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

The NMIJ ampoule was measured two weeks before the reference date, at the reference date and again three weeks later. All three results agree to within  $6 \times 10^{-4}$ , the SIR combined standard uncertainty.

Five NMIs (the PTB, IRA, NMIJ, OMH and the CMI) that have sent different ampoules over about 13 years, demonstrate consistency within statistical standard uncertainties between  $2 \times 10^{-3}$  and  $5 \times 10^{-3}$ .

One earlier result was withdrawn and does not appear here. No recent submission has been identified as a pilot study so the most recent result of each NMI is normally eligible for Appendix B of the MRA. However, the IAEA no longer undertakes the metrology of activity and the AECL is not a designated laboratory of the NRC, Canada, therefore none of these results is included in the KCDB.

**Table 4a. Results of SIR measurements of  $^{88}\text{Y}$**

NMI	Mass of solution $m_i / \text{g}$	Activity submitted $A_i / \text{kBq}$	N° of Ra source used	SIR $A_e / \text{kBq}$	Relative uncertainty from SIR	Combined uncertainty $u_{c,i} / \text{kBq}$
NMIJ	3.607 43	926.8	3	6931	$5 \times 10^{-4}$	40
	3.638 36	934.7		6935		40
	3.646 26	1 656.5	4	6903	$4 \times 10^{-4}$	21
CMI-IIR	3.704 7	28 640	5	7150	$3 \times 10^{-4}$	100
	3.610 55	3 618	4	7043	$4 \times 10^{-4}$	47
	3.626 4	16 950	5	6882	$3 \times 10^{-4}$	17
	3.598 9	5 168	5	6865	$3 \times 10^{-4}$	26
NPL	3.666 5	1 911.7	4	6910	$4 \times 10^{-4}$	24
	3.412 0	1 779.0		6901 <sup>†</sup>		24

continued overleaf.

**Table 4a continued. Results of SIR measurements of <sup>88</sup>Y**

NMI	Mass of solution $m_i$ / g	Activity submitted $A_i$ / kBq	N° of Ra source used	SIR $A_e$ / kBq	Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / kBq
PTB	3.664 7 (1)	2 939.4	4	6868.2	$4 \times 10^{-4}$	4.7
	3.633 1 (1)	2 914.1		6868.4		4.7
	3.622 17	10 004	5	6904	$4 \times 10^{-4}$	7
	3.600 31	9 944		6905		7
	3.656 0	3 431.5	4	6871	$4 \times 10^{-4}$	15
	3.567 54	5 383.8	5	6877	$3 \times 10^{-4}$	8
OMH	3.603 8	1 355.8	4	6894	$4 \times 10^{-4}$	45
	3.601 8	1 355.0		6894		45
	3.600 6	2 233	4	6932	$4 \times 10^{-4}$	36
	3.606 7	3 196	5	6858	$3 \times 10^{-4}$	20
	3.595 8	1 000.4	3	6865	$5 \times 10^{-4}$	18
ANSTO	3.574 3	472.6	3	6924 <sup>†</sup>	$6 \times 10^{-4}$	28
IAEA	3.616 02	3 638	4	7043	$4 \times 10^{-4}$	47
IRA	3.598 4	5 092	4	6876	$5 \times 10^{-4}$	28
	3.602 4	5 098		6876		28
	3.570 1 (1)	861.3	3	6893	$5 \times 10^{-4}$	16
NIST	3.613 8	904.46	3	6889	$6 \times 10^{-4}$	5
	3.578 2 (2)	1 187.5	3	6913	$6 \times 10^{-4}$	15
AECL	0.285 91*	601.0	3	6873	$7 \times 10^{-4}$	21
	0.479 67*	1 008.2		6878 <sup>†</sup>	$8 \times 10^{-4}$	21
BNM-LNHB	3.600 99	456.75	3	6817	$7 \times 10^{-4}$	8
	3.619 26	459.07		6812	$6 \times 10^{-4}$	8
	3.651 2	2 822	4	6882	$4 \times 10^{-4}$	22
VNIIM	3.464 7	3 763	4	6912	$5 \times 10^{-4}$	23
LNMRI	3.461 37	400.8	2	6907 <sup>†</sup>	$8 \times 10^{-4}$	29
BEV	3.595 9	1 230.2	3	6895	$6 \times 10^{-4}$	27

<sup>†</sup> the mean of the two  $A_e$  values shown for the same measurement date is used with an averaged uncertainty, as attributed to an individual entry [10]

\* mass of solution before dilution

<sup>†</sup> superseded in the KCDB by the more recent APMP comparison result in Table 4b.

The results of the APMP comparison have been published [5]. However as the P3KRBiN is not currently a designated institute of Indonesia and the MINT is not currently a designated institute of Malaysia, neither of these results can be included in the KCDB. The eight laboratories to be added to the matrix of degrees of equivalence

from this publication are those indicated in Table 1b. The results for these eight NMIs are all linked to the SIR through the result of the NMIJ using the simple ratio from

$$A_{ei} = [(A/m)_i / (A/m)_{\text{NMIJ}}] \times A_{e\text{NMIJ}}$$

as shown in Table 4b.

**Table 4b. Results of APMP measurements of  $^{88}\text{Y}$  and links to the SIR**

NMI	Measurement method	Date reported to the pilot laboratory	Activity concentration measured <sup>†</sup> $(A/m)_i / (\text{kBq g}^{-1})$ $(u_{\text{rel}})_i$	Equivalent SIR activity $A_{ei} / \text{kBq}$	Combined standard uncertainty $u_{c,i} / \text{kBq}$
ANSTO	Ionization chamber <sup>†</sup>	23-Jun-00	453.1 (0.45 %)	6885	31
BARC	Ionization chamber <sup>++</sup>	21-Jul-00	455.7 (1.53 %)	6924	106
CNEA	Ge spectrometry	05-Oct-00	453.7 (1.3 %)	6894	86
INER	4 $\pi$ PC $\beta$ - $\gamma$ coincidence	26-Jun-00	456.5 (0.36 %)	6936	25
KRISS	4 $\pi$ PPC $\beta$ - $\gamma$ coincidence	19-Jul-00	454.9 (0.29 %)	6912	20
LNMRI	Ionization chamber <sup>†</sup>	04-Jul-00	455.3 (0.47 %)	6918	33
NIM	Ge spectrometry	23-Jun-00	454.3 (0.64 %)	6903	44
OAP	Ionization chamber traceable to NMIJ	07-Sep-00	453.3 (1.5 %)	6888	103
MINT	Ge spectrometer traceable to NIST	30-Jun-00	479.6 (0.8 %)	7287 <sup>*</sup>	58
P3KRBiN	Ionization chamber traceable to PTB	14-Jun-00	450.4 (1.0 %)	6844 <sup>*</sup>	65

<sup>†</sup> at reference date 1 April 2000 0h UT

<sup>+</sup> calibrated by primary measurements of  $^{88}\text{Y}$  solution in 1977 for the ANSTO and in April 2000 for the LNMRI

<sup>++</sup> calibrated by extrapolation from other primary measurements in 1982 and verified against more recent primary standardizations

<sup>\*</sup> not included in the KCDB (see Table 1b).

The uncertainties for the APMP comparison linked to the SIR are comprised of the original NMI uncertainties (given in Table 4b) together with the uncertainty in the

link,  $4 \times 10^{-4}$ , given by the uncertainty in the SIR measurement of the NMIJ ampoule of this APMP.RI(II)-K2 comparison. The uncertainty budgets for the eight other laboratories, the ANSTO, BARC, CNEA, INER, KRISS, LNMRI, NIM and the OAP are also given in Appendix 1.

The results for the ANSTO and the LNMRI from the APMP comparison agree within the combined standard uncertainty with their original SIR equivalent activity values. However, as the APMP comparison result is more recent for these NMIs, the earlier SIR results have been superseded by the values in Table 4b for the results presented in the KCDB.

#### 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, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;
- 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 mother-file. Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are only made by the CCRI(II), normally during one of its biennial meetings.

Consequently, the KCRV for  $^{88}\text{Y}$  has been identified as 6893 (5) kBq using the most recent results from the NPL, ASMW, AECL, VNIIM, OMH, BNM-LNHB, CMI-IIR, PTB, NMIJ, IRA and the results from the NIST (1980), the LNMRI (1999) and the ANSTO (1977).

#### 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

$$D_i = A_{ei} - \text{KCRV} \quad (1)$$

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

$$U_i = 2u_{D_i}, \quad (2)$$

taking correlations into account as appropriate (see Appendix 2).

#### 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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \quad (3)$$

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

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

and any obvious correlations in the standard uncertainties for a given component,  $u_{\text{corr},k}$ , between the NMIs (such as a traceable calibration) are subtracted using an appropriate correlation coefficient,  $f_k$ , 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 (identified as  $x_R$  in the KCDB), is shown in Figure 1 where the value shown as a black square predates 1983. This figure 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 APMP comparison have been linked to those of the SIR through the NMIJ. The degrees of equivalence with the KCRV and between the pairs of NMIs are shown as the extension of the matrix in Table 5. The correlations associated with having a linking laboratory have been taken into account but the correlations associated with the distribution of the same solution have been ignored in the analysis as the overall uncertainties are quite large.

## Conclusion

The BIPM ongoing key comparison for  $^{88}\text{Y}$ , BIPM.RI(II)-K1.Y-88 currently comprises ten results. These have been analysed with respect to the KCRV determined for this radionuclide, and with respect to each other. The matrix of degrees of equivalence has been approved by the CCRI(II) and is published in the BIPM key comparison database.

The results of eight other NMIs in the APMP key comparison for  $^{88}\text{Y}$  have been linked to the BIPM ongoing key comparison through the common participant and pilot laboratory for this comparison, the NMIJ. 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  $^{88}\text{Y}$  activity measurements to the SIR comparison or take part in other linked regional comparisons.

## Acknowledgements

The authors would like to thank the NMIs and J.G.V. Taylor and R.H. Martin of the AECL for their participation in this comparison, Mr Christian Colas of the BIPM for his dedicated work in maintaining the SIR since its inception and for the thousands of measurements he has made over the years, and Dr P.J. Allisy-Roberts of the BIPM for editorial assistance.

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**Table 5. Introductory text and table of degrees of equivalence for <sup>88</sup>Y**

**Key comparison BIPM.RI(II)-K1.Y-88**

**MEASURAND :** Equivalent activity of <sup>88</sup>Y

**Key comparison reference value: the SIR reference value for this radionuclide is  $x_R = 6893$  kBq with a standard uncertainty,  $u_R = 5$  kBq.**

**$x_R$  is the mean of the SIR results (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 kBq, and with  $n$  the number of laboratories

$U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\sum u_j^2)^{1/2}$  when each laboratory has contributed to the calculation of  $x_R$ .

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 with correlations included between the BEV and the NPL

**Linking APMP.RI(II)-K2.Y-88 to BIPM.RI(II)-K1.Y-88**

**The value  $x_i$  is the equivalent activity for laboratory  $i$  participant in APMP.RI(II)-K2.Y-88 having been normalized to the value of the NMIJ as the linking laboratory.**

The degree of equivalence of laboratory  $i$  participant in APMP.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 kBq.

The approximation  $U_i = 2(u_i^2 + u_R^2)^{1/2}$  is used in the following table as none of these  $D_i$  contributed to the KCRV.

The degree of equivalence between two laboratories  $i$  and  $j$ , one participant in BIPM.RI(II)-K1.Y-88 and one in APMP.RI(II)-K2.Y-88, or both participants in APMP.RI(II)-K2.Y-88, is given by a pair of terms expressed in kBq:  $D_{ij} = D_i - D_j$  and  $U_{ij}$ , its expanded uncertainty ( $k = 2$ ), approximated by  $U_{ij} \sim 2(u_i^2 + u_j^2 - 2fu_i^2)^{1/2}$  with  $l$  being the linking laboratory when each laboratory is from the APMP and  $f$  is the correlation coefficient, with correlations included between the OAP and the NMIJ.

These statements make it possible to extend the BIPM.RI(II)-K1.Y-88 matrices of equivalence to the participants in APMP.RI(II)-K2.Y-88.

Table 5 continued. Degrees of equivalence for <sup>88</sup>Y

Lab j →

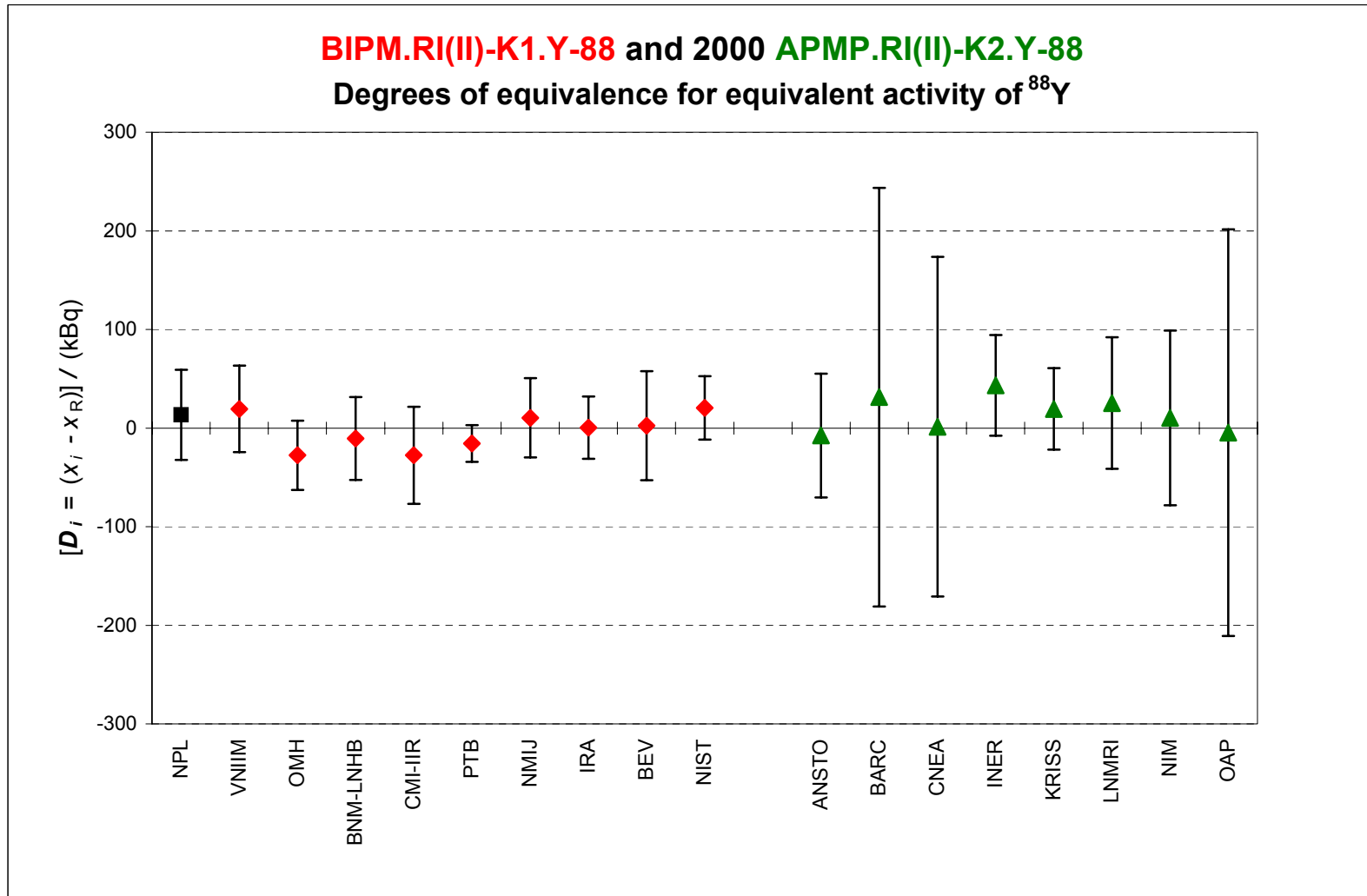
Lab i ↓	$D_i$ $U_i$ / kBq		NPL		VNIIM		OMH		BNM-LNHB		CMI-IIR		PTB		NMIJ		IRA		BEV		NIST	
	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$
NPL	13	46																				
VNIIM	19	44	6	66			47	58	30	64	47	69	35	49	9	62	19	56	17	71	-1	55
OMH	-28	35	-41	60	-47	58			-17	57	0	63	-12	39	-38	55	-28	48	-30	65	-48	47
BNM-LNHB	-11	42	-24	65	-30	64	17	57			17	68	5	47	-21	61	-11	54	-13	70	-31	53
CMI-IIR	-28	49	-41	71	-47	69	0	63	-17	68			-12	54	-38	67	-28	61	-30	75	-48	60
PTB	-16	19	-29	51	-35	49	12	39	-5	47	12	54			-26	45	-16	36	-18	56	-36	34
NMIJ	10	40	-3	64	-9	62	38	55	21	61	38	67	26	45			10	53	8	68	-10	52
IRA	0	32	-13	58	-19	56	28	48	11	54	28	61	16	36	-10	53			-2	63	-20	44
BEV	2	55	-11	33	-17	71	30	65	13	70	30	75	18	56	-8	68	2	63			-18	62
NIST	20	32	7	57	1	55	48	47	31	53	48	60	36	34	10	52	20	44	18	62		

ANSTO	-8	63	-21	78	-27	77	20	72	3	76	20	81	8	64	-18	74	-8	70	-10	82	-28	69
BARC	31	212	18	217	12	217	59	215	42	217	59	218	47	213	21	216	31	214	29	219	11	214
CNEA	1	172	-12	179	-18	178	29	176	12	178	29	180	17	173	-9	177	1	175	-1	180	-19	175
INER	43	51	30	69	24	68	71	62	54	67	71	72	59	52	33	65	43	59	41	74	23	58
KRISS	19	41	6	62	0	61	47	54	30	59	47	66	35	43	9	57	19	51	17	67	-1	50
LNMRI	25	67	12	82	6	80	53	75	36	79	53	84	41	68	15	78	25	73	23	85	5	72
NIM	10	89	-3	100	-9	99	38	95	21	98	38	102	26	89	0	97	10	94	8	103	-10	93
OAP	-5	206	-18	212	-24	211	23	209	6	211	23	212	11	207	-15	193	-5	208	-7	213	-25	208

Lab i ↓	$D_i$ $U_i$ / kBq		ANSTO		BARC		CNEA		INER		KRISS		LNMRI		NIM		OAP	
	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$
NPL	13	46	21	78	-18	217	12	179	-30	69	-6	62	-12	82	3	100	18	212
VNIIM	19	44	27	77	-12	217	18	178	-24	68	0	61	-6	80	9	99	24	211
OMH	-28	35	-20	72	-59	215	-29	176	-71	62	-47	54	-53	75	-38	95	-23	209
BNM-LNHB	-11	42	-3	76	-42	217	-12	178	-54	67	-30	59	-36	79	-21	98	-6	211
CMI-IIR	-28	49	-20	81	-59	218	-29	180	-71	72	-47	66	-53	84	-38	102	-23	212
PTB	-16	19	-8	64	-47	213	-17	173	-59	52	-35	43	-41	68	-26	89	-11	207
NMIJ	10	40	18	74	-21	216	9	177	-33	65	-9	57	-15	78	0	97	15	193
IRA	0	32	8	70	-31	214	-1	175	-43	59	-19	51	-25	73	-10	94	5	208
BEV	2	55	10	82	-29	219	1	180	-41	74	-17	67	-23	85	-8	103	7	213
NIST	20	32	28	69	-11	214	19	175	-23	58	1	50	-5	72	10	93	25	208

ANSTO	-8	63			-39	221	-9	183	-51	79	-27	73	-33	90	-18	107	-3	215
BARC	31	212	39	221			30	273	-12	218	12	216	6	222	21	229	36	295
CNEA	1	172	9	183	-30	273			-42	179	-18	176	-24	184	-9	193	6	268
INER	43	51	51	79	12	218	42	179			24	64	18	82	33	101	48	212
KRISS	19	41	27	73	-12	216	18	176	-24	64			-6	77	9	96	24	210
LNMRI	25	67	33	90	-6	222	24	184	-18	82	6	77			15	110	30	216
NIM	10	89	18	107	-21	229	9	193	-33	101	-9	96	-15	110			15	224
OAP	-5	206	3	215	-36	295	-6	268	-48	212	-24	210	-30	216	-15	224		

**Figure 1. Graph of degrees of equivalence with the KCRV for  $^{88}\text{Y}$**   
 (as it appears in Appendix B of the MRA)



**Appendix 1. Uncertainty budgets for the activity of <sup>88</sup>Y**

Uncertainty budgets submitted to the SIR by the CMI-IIR, PTB, NMIJ, IRA, BEV and the NIST.

**CMI-IIR (1999)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
<b>Contributions due to</b>		
counting statistics	10	–
weighing	–	1
dead time	–	1
background	–	5
pile-up	–	15
resolving time	–	5
Gandy effect	–	1
counting time	–	1
adsorption	–	1
radionuclide impurities	–	10
extrapolation of efficiency curve	–	30
<b>Quadratic summation</b>	<b>10</b>	<b>36</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>37</b>	

**PTB (1999)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
<b>Contributions due to</b>		
statistics (current measurements of <sup>88</sup> Y solution in BIPM type ampoule; number of repetitions $n \geq 15$ )	3	–
linearity of current measurement	–	5
radium reference source current (number of repetitions $n \geq 15$ )	2.5	–
background (relative to source current)	–	<0.5
weighing of ampoule	–	2.4
dilution factor (no dilution performed)	–	0
half-life	–	<0.5
radionuclide impurities	–	0
geometry correction	–	3
calibration coefficient for <sup>88</sup> Y	–	12
<b>Quadratic summation</b>	<b>3.9</b>	<b>13.6</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>14.2</b>	

**NMIJ (2000)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	10	–
weighing	–	5
adsorption	–	5
dead time	–	5
pile-up	–	10
timing	–	5
background	5	–
half-life	–	5
decay scheme	–	5
extrapolation	19	–
small contribution of e <sup>+</sup>	–	10
radionuclide impurities	–	5
<b>Quadratic summation</b>	<b>22</b>	<b>19</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>29</b>	

**IRA 4P-PC-MX-NA-GR-CO (2001)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
background	–	19
half-life	–	2
dilution factor	–	0.2
weighing of the sources	–	21
dead time correction	–	0.1
timing	–	0.2
extrapolation of efficiency function (includes pile up uncertainty)	10	–
<b>Quadratic summation</b>	<b>10</b>	<b>28</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>30</b>	

**IRA 4P-NA -GR-00-00-HE (2001)**

<b>Relative standard uncertainties</b>	$u_i \times 10^4$ evaluated by method	
<b>Contributions due to</b>	<b>A</b>	<b>B</b>
background	–	6.3
energy cut off	–	24
dead time correction	–	0.2
timing	–	0.2
half-life	–	1.5
dilution factor	–	0.2
weighing of the sources	–	21.1
efficiency calculation	4	–
counting statistics	1.1	–
<b>Quadratic summation</b>	<b>4.1</b>	<b>32.6</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>33</b>	

The mean value of the results for the two methods is taken as the final value together with the arithmetic mean of the uncertainties for the two methods,  $\frac{1}{2}(u_1^2 + u_2^2)^{0.5}$ .

**BEV (2001)**

<b>Relative standard uncertainties</b>	$u_i \times 10^4$ evaluated by method	
<b>Contributions due to</b>	<b>A</b>	<b>B</b>
counting statistics	6	–
weighing	–	5
background	–	9
half-life	–	2
chamber calibration factor	–	33
ionization chamber	–	10
current measurement	–	10
radionuclide impurities	–	<1
<b>Quadratic summation</b>	<b>6</b>	<b>37</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>38</b>	

**NIST (2002)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
<b>Contributions due to</b>		
PIC A* net response per gram of BIPM-02-88 measured relative to RRS10	1	–
PIC A net response for RRS10 measured relative to RRS50	4	–
PIC A net response per Bq of $^{88}\text{Y}$ in solution, measured relative to RRS50	3	–
Activity used to calibrate PIC A net response per Bq of $^{88}\text{Y}$ in solution	–	15
PIC A charge collection	–	5
Gravimetric measurements	–	5
Half-life of $^{88}\text{Y}$	0.1	–
Half-life of $^{226}\text{Ra}$	0.4	
Live time	–	5
Source positioning	–	5
Photon-emitting impurities	–	8
<b>Quadratic summation</b>	<b>5</b>	<b>20</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>20</b>	

\* PIC A, pressurized ionization chamber A

**Uncertainty budgets for the APMP 2000 comparison.****ANSTO Uncertainty budget**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
calibration factor	–	39.4
IC current measurement	6.3	–
IC stability correction	–	20.5
background measurement	0.03	–
sample mass	–	0.1
Y-88 half life	–	2.7
<b>Quadratic summation</b>	<b>6.3</b>	<b>44.5</b>
<b>Total relative combined uncertainty <math>u_c</math></b>	<b>45</b>	

**BARC Uncertainty budget**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
Counting statistics	10	–
calibration factor	–	135
source positioning	–	20
source volume	–	10
collection efficiency	–	20
electrometer non-linearity	–	10
wall thickness of source container	–	10
current from radium source	50	–
background	40	–
<b>Quadratic summation</b>	<b>65</b>	<b>139</b>
<b>Total relative combined uncertainty <math>u_c</math></b>	<b>153</b>	

**CNEA Uncertainty budget**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
<b>Contributions due to</b>		
counting statistics	7	–
weighing	–	5
dead time	–	4
background	25	–
timing	–	20
half life	–	<1
extrapolation of efficiency curve	120	–
<b>Quadratic summation</b>	<b>123</b>	<b>21</b>
<b>Total relative combined uncertainty <math>u_c</math></b>	<b>125</b>	

**INER Uncertainty budget**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
<b>Contributions due to</b>		
counting	11	–
weighing	–	20
dead time and resolving time	–	9
background	5	–
timing	–	1
linear extrapolation	25	–
Half-life	–	2
Decay scheme	–	5
<b>Quadratic summation</b>	<b>28<sup>†</sup></b>	<b>22</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>36</b>	

<sup>†</sup> effective degrees of freedom, 33

**KRISS Uncertainty budget**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
<b>Contributions due to</b>		
counting statistics ( $v^\dagger = 8$ )	14	–
weighing	–	4
dead time	–	2
background	–	9
timing	–	0.5
half-life	–	1
linear extrapolation	–	23
<b>Quadratic summation</b>	<b>14</b>	<b>25</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>29</b>	

<sup>†</sup> degrees of freedom

**LNMRI Uncertainty budget**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
<b>Contributions due to</b>		
counting statistics	13*	–
weighing	–	10
dead time	–	29
resolving time	–	13
delay mismatch	–	7
pile up	–	–
background	–	9
timing	–	–
half-life	–	<0.01
adsorption	–	–
extrapolation	31	–
<b>Quadratic summation</b>	<b>31</b>	<b>35</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>47</b>	

\* included in the extrapolation fitting

**NIM Uncertainty budget**

<b>Relative standard uncertainties</b>	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
counting statistics ( $v^\dagger = 5$ )	5.8	–
weighing	–	2
peak analysis method	–	10
pile up	–	7
timing	–	1
coincidence summing	–	3
full energy peak efficiency	–	54
$\gamma$ -ray probability	–	32
half-life	–	0.2
<b>Quadratic summation</b>	<b>5.8</b>	<b>64</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>64</b>	

<sup>†</sup> degrees of freedom

**OAP Uncertainty budget**

<b>Relative standard uncertainties</b>	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
ionization chamber current	24	–
stability of ionization chamber system	27	–
calculated response	–	100
background instability	–	40
half-life	–	4
<b>Quadratic summation</b>	<b>36</b>	<b>108</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>114</b>	

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

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

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

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

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

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

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

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

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

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

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

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

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

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

Then the total combined uncertainty becomes

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

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

or, after recombination

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

similar to (A-11).

### Appendix 3. Acronyms used to identify different measurement methods

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

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

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