BIPM comparison BIPM.RI(II)-K1.I-123 of activity measurements of the radionuclide ^{123}I and links for the 1983 EUROMET.RI(II)-K2.I-123 comparison

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Abstract

Since 1981, three national metrology institutes (NMI) and two other laboratories have submitted nine samples of known activity of ^{123}I to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures. The activities ranged from about 1.8 MBq to 84 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 three NMIs and an international laboratory. A graphical presentation is also given. The results of this comparison have been approved by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), comparison identifier BIPM.RI(II)-K1.I-123. The results of a EUROMET comparison, identifier EUROMET.RI(II)-K2.I-123 held in 1983 that have been approved for provisional equivalence for this radionuclide, have been linked to the SIR results. This has enabled two other NMIs to have their results in the key comparison database.

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 (or gaseous) form. The NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of ^{226}Ra using pressurized ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity are all given in [1].

Since its inception until 31 December 2002, the SIR has measured 835 ampoules to give 606 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.I-123 key comparison.

In addition, an international comparison was held in 1983 for this radionuclide, EUROMET.RI(II)-K2.I-123 [3], piloted by the IRMM and this comparison has been given the status of having provisional equivalence in the KCDB. Although five laboratories took part in this comparison, two are SIR participants and two others are eligible to be linked to the BIPM key comparison through this EUROMET comparison.

2. Participants

Three NMIs and two other laboratories have submitted nine ampoules for the comparison of ^{123}I activity measurements since 1981. 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 two eligible participants that took part in the EUROMET.RI(II)-K2.I-123 comparison in 1983 are given in Table 1b.

Table 1a. Details of the participants in the BIPM.RI(II)-K1.I-123

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
LMRI	BNM-LNHB	Bureau national de métrologie-Laboratoire national Henri Becquerel	France	EUROMET	1981-04-09 1983-05-31
NIRH	–	National Institute of Radiation Hygiene	Denmark	EUROMET	1983-03-11
IRMM (CBNM)	–	Institute for Reference Materials and Measurements	European Union	EUROMET	1983-05-31
IER	IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	1985-06-27
	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1985-10-25

Table 1b. Details of the participants in the EUROMET.RI(II)-K2.I-123

NMI	Full name	Country	Regional metrology organization
NPL	National Physical Laboratory	United Kingdom	EUROMET
SCK-CEN	Studiecentrum voor Kernenergie - Centre d'étude de l'Energie Nucléaire	Belgium	EUROMET

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. Consequently, no uncertainty budgets were provided for this comparison.

The half-life used by the BIPM is 13.22 (4) h and this agrees with the recent evaluation of 13.21 (3) h in [4].

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 [5]. The CCRI(II) agreed in 1999 [6] that this method should be followed according to the protocol described in [7] when an NMI makes such a request or when there appear to be discrepancies.

Details of the solution issued and the standardization methods used in the EUROMET comparison are given in [3].

Table 2. Standardization methods of the participants for ^{123}I

NMI	Method used and acronym (see Appendix 3)	Half-life / h	Activity A_i / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
BNM-LNHB	4πγ 4P-NA-GR-00-00-00	–	4 114 4 117 4 120 †	81-04-09 12 h UT	0.09	0.26
	Pressurized IC 4P-IC-GR-00-00-00 calibrated by 4π(NaI(Tl))γ 4P-NA-GR-00-00-00	13.21 [8]	1 803 *††	83-06-01 0 h UT	0.28 ††	
NIRH	Pressurized IC (type IG11) 4P-IC-GR-00-00-00	–	84 390	83-03-09 9 h UT	0.7	0.9
IRMM †	4π(e_A,x)-γ coinc. 4P-PC-MX-NA-GR-CO and HPGe γ-spectrometer [3] UA-GH-GR-00-00-00	13.21 [8]	1 868 * 1 853 *	83-06-01 0 h UT	0.32	0.44
IRA	Pressurized IC 4P-IC-GR-00-00-00 calib. in 1985 by 4π(PC)x_e–γ coinc. 4P-PC-BP-NA-GR-CO and 4π(NaI(Tl))γ 4P-NA-GR-00-00-00	–	20 509 **	85-06-27 10 h UT	0.47 **	
PTB	Pressurized IC 4P-IC-GR-00-00-00 calib. in 1985 by 4π(PC)-γ and 4P-PC-MX-NA-GR-CO 4π(PPC)-γ coinc 4P-PP-MX-NA-GR-CO	–	29 520	85-10-22 23 h UT	0.05	0.49

† several ampoules submitted

†† value published in [3], that differs slightly from the earlier value registered in the SIR (1795 kBq, relative standard uncertainty of 0.46×10^{-2}) based on earlier decay scheme parameters

* the three ampoules measured by the BNM-LNHB and the IRMM for the EUROMET.RI(II)-K2.I-123 and measured in the SIR are used to make the link for the EUROMET key comparison

** the original value registered in the SIR (20 536 kBq, relative standard uncertainty of 0.38×10^{-2}), was based on the IC calibration using both these primary methods and two other secondary methods.

Table 3. Details of the solution of ^{123}I submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity†
BNM-LNHB	NaI + $Na_2S_2O_3$ + NaCl 0.5 % in water	–	NaI : 50 $Na_2S_2O_3$: 50	0.998	^{125}I : 2.96 (5) %
	KI + $Na_2S_2O_3$ in NaOH *	5×10^{-4}	KI : 33 $Na_2S_2O_3$: 16	–	^{125}I : 1.21 (5) %
NIRH	Hippurate (from IRE‡)	–	–	–	^{125}I : 0.6 (1) %
IRMM	KI + $Na_2S_2O_3$ in NaOH *	5×10^{-4}	KI : 33 $Na_2S_2O_3$: 16	–	^{125}I : 1.28 (8) %
IRA	KI + Na_2SO_3 in diluted LiOH	8×10^{-4}	KI : 50 Na_2SO_3 : 20	1.000	^{125}I : 6.9 (4) % ^{121}Te : 0.032 (3) % ^{123}Te^{m} : $1.5\ (2) \times 10^{-3}$ % ^{126}I : $5\ (2) \times 10^{-4}$ %
PTB	$Na_2S_2O_3$ in NaOH	0.02	$Na_2S_2O_3$: 40	1	–

† the ratio of the activity of the impurity to the activity of ^{123}I at the reference date
* same solution as for the EUROMET comparison
‡ Institut national des radioéléments (IRE) in Belgium that used to supply this material.

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 ^{123}I arise from nine ampoules and the SIR equivalent activity for each ampoule, A_{ei}, is given in Table 4 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. 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].

The ^{125}I impurity has a negligible influence on the registered SIR measurements. The ampoule of IRA that contains several impurities has the highest impurity correction (1.0016).

Measurements repeated at the BIPM after periods of up to 1 or 2 days later produced comparison results in agreement within the SIR uncertainty for the IRMM and for the BNM-LNHB (1981 and 1983). These measurements confirm the validity of the half-life value used.

When the measurements at the BIPM are repeated after periods of up to 4 days later for the NIRH and IRA ampoules, the impurity corrections reach values of 1.021 and 1.19 respectively. These repeated measurements show a decreasing trend for both laboratories, although the results still agree within one and two SIR uncertainties respectively.

One early result was withdrawn and is not included in this report. 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 NIRH no longer undertakes the metrology of activity so their results are not included in the KCDB.

Table 4a. Results of SIR measurements of ^{123}I

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
BNM-LNHB	3.608 60	4 114	2	119 990	17×10^{-4}	390
	3.611 81	4 117		120 050		390
	3.613 78	4 120		119 840		390
	3.603 57 (2)	1 803	2	120 020 *	27×10^{-4}	470
NIRH	3.627 9	84 390	3	118 200	88×10^{-4} †	1700
IRMM	3.729 9	1 868	2	120 114 *	27×10^{-4}	730
	3.700 2	1 853		120 131 *††		730
IRA	3.587 3 (1)	20 509	4	119 450 **	8×10^{-4} **	570
PTB	3.629 07	29 520	1	121 400 **	99×10^{-4} †**	1300

* result used to link the EUROMET.RI(II)-K2.I-123 comparison
** revised data analysis of the SIR that is more appropriate to short lived radionuclides
† the uncertainty on the ^{123}I half life produces a large SIR uncertainty because of the delay of more than two days between the SIR measurement and the reference date
†† the mean of the A_e values is used with an averaged uncertainty, as attributed to an individual entry [9].

The results of the international comparison EUROMET(II)-K2.I-123 have been published [3]. The two laboratories to be added to the matrix of degrees of equivalence from this previous publication are those given in Table 1b. The results $(A/m)_i$ for these laboratories are linked to the SIR through the measurement in the SIR of the three ampoules standardized by the BNM and the IRMM and measured in the SIR at the same time as the EUROMET comparison. The link is made using a ratio deduced from these three lines of Table 4a:

$$A_{ei} = (A/m)_i \times \tfrac{1}{3} \sum_{L=1}^{3} (A_{e,\mathrm{NMI},L} / (A/m)_{\mathrm{NMI},L}) = (A/m)_i \times 239.86 \qquad \text{(a)}$$

The results of the links for the two other laboratories are given in Table 4b. The uncertainties for the international comparison linked to the SIR are comprised of the original uncertainties together with the uncertainty in the link, 2.7×10^{-3}, given by the uncertainty of the SIR measurement of the EUROMET ampoules.

Table 4b. Results of 1983 EUROMET.RI(II) measurements of ^{123}I linked to the SIR

NMI	Activity * concentration $(A/m)_i$ / (kBq g^{-1})	Relative standard uncertainty × 100	Linked SIR A_{ei} / kBq	Combined uncertainty $u_c(A_{ei})$ / kBq
NPL	502.3	0.24	120 480	440
SCK-CEN	495.2	0.28	118 780	460

* referenced to 1983-06-01, 0 h UT, as given in [3].

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 ^{123}I has been identified as 120 250 (410) kBq using the results from the BNM-LNHB (1983), IRMM, IRA and the PTB.

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

$$D_i = A_{ei} - \mathrm{KCRV} \qquad (1)$$

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

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

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

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

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

$$u_{Dij}{}^2 = u_i{}^2 + u_j{}^2 - \sum_k \left(f_k u_{\mathrm{corr},k}\right)_i^2 - \sum_k \left(f_k u_{\mathrm{corr},k}\right)_j^2 \qquad (4)$$

and any obvious correlations in the standard uncertainties for a given component, $u_{\mathrm{corr},k,}$, 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. The additional matrix cells show the two results from the 1983 EUROMET comparison linked to those of the SIR and given in Table 4b. 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. This 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.

Conclusion

The BIPM ongoing key comparison for ^{123}I, BIPM.RI(II)-K1.I-123 currently comprises four 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 two other NMIs that took part in the EUROMET.RI(II)-K2.I-123 comparison in 1983 have been linked to the BIPM ongoing key comparison through three ampoules of the comparison measured in the SIR. These linked results are included in the matrix of degrees of equivalence approved by the CCRI(II).

Other results may be added as and when other NMIs contribute ^{123}I activity measurements to this comparison or take part in other linked comparisons.

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Table 5. Introductory text and table of degrees of equivalence for ^{123}I

Key comparison BIPM.RI(II)-K1.I-123

MEASURAND : Equivalent activity of ^{123}I

Key comparison reference value: the SIR reference value for this radionuclide is $x_R = 120.25$ MBq with a standard uncertainty, $u_R = 0.41$ MBq.
x_R is the mean of the four latest SIR results.

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 with n the number of laboratories $U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\Sigma u_j^2)^{1/2}$ 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 MBq.

The approximation $U_{ij} \sim 2(u_i^2 + u_j^2)^{1/2}$ is used in the following table.

Linking EUROMET.RI(II)-K2.I-123 to BIPM.RI(II)-K1.I-123

The value x_i is the equivalent activity for laboratory i participant in EUROMET.RI(II)-K2.I-123 having been normalized to the value of the BNM-LNHB and IRMM as linking laboratories.

The degree of equivalence of laboratory i participant in EUROMET.RI(II)-K2.I-123 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.I-123 and one in EUROMET.RI(II)-K2.I-123, or both participants in EUROMET.RI(II)-K2.I-123, 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 being the linking laboratory when each laboratory is from the EUROMET and f is the correlation coefficient.

These statements make it possible to extend the BIPM.RI(II)-K1.I-123 matrices of equivalence to all participants in EUROMET.RI(II)-K2.I-123.

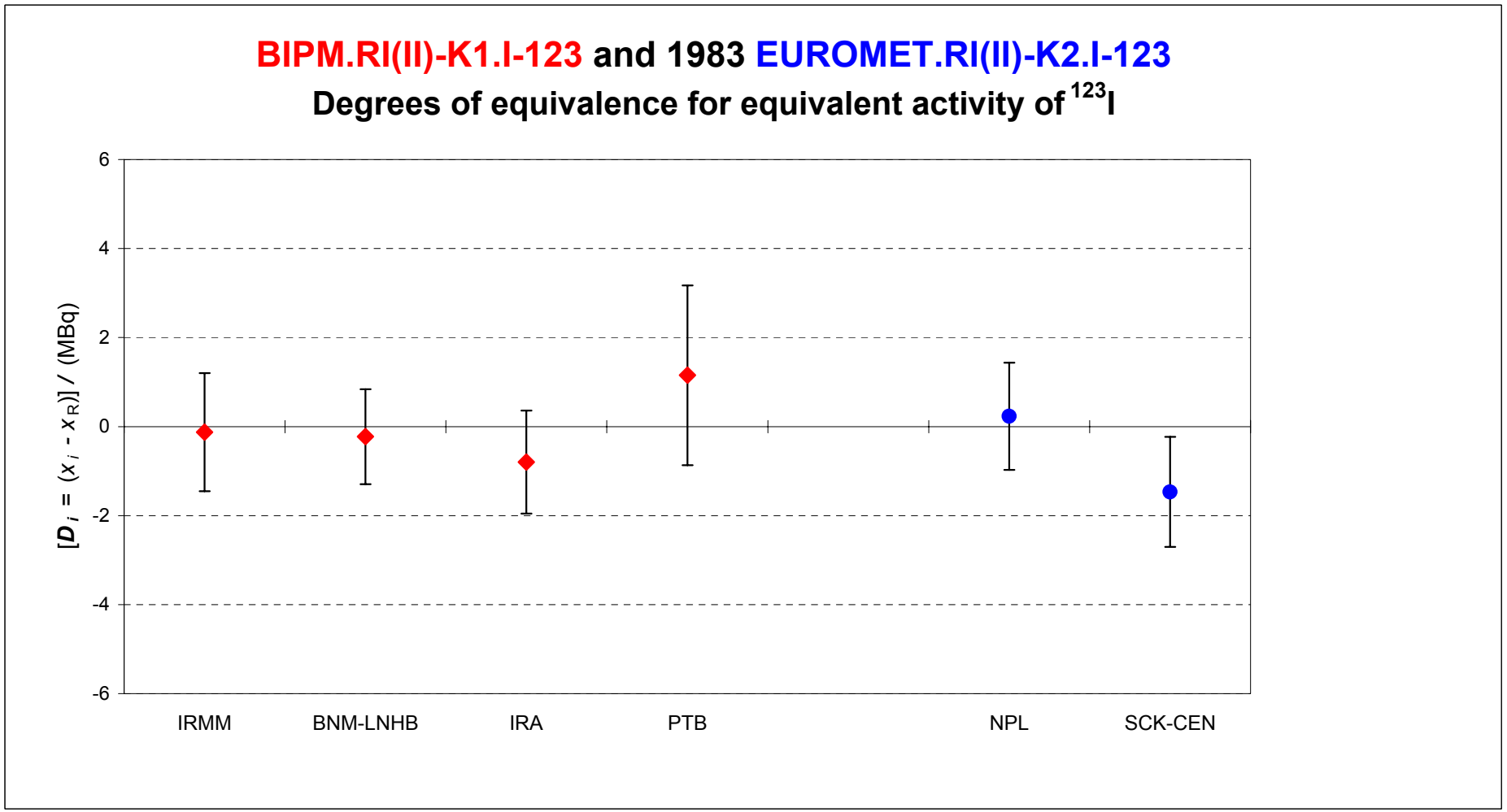
Table 5 continued. Degrees of equivalence for ^{123}I

Lab *j* ⟹

Lab *i* ⇩

	D_i / MBq	U_i / MBq	IRMM D_{ij} / MBq	IRMM U_{ij} / MBq	BNM-LNHB D_{ij} / MBq	BNM-LNHB U_{ij} / MBq	IRA D_{ij} / MBq	IRA U_{ij} / MBq	PTB D_{ij} / MBq	PTB U_{ij} / MBq	NPL D_{ij} / MBq	NPL U_{ij} / MBq	SCK-CEN D_{ij} / MBq	SCK-CEN U_{ij} / MBq
IRMM	**-0.1**	1.3			**0.1**	1.7	**0.7**	1.9	**-1.3**	3.0	**-0.4**	1.4	**1.3**	1.5
BNM-LNHB	**-0.2**	1.1	**-0.1**	1.7			**0.6**	1.5	**-1.4**	2.8	**-0.5**	0.9	**1.2**	0.9
IRA	**-0.8**	1.2	**-0.7**	1.9	**-0.6**	1.5			**-2.0**	2.8	**-1.0**	1.4	**0.7**	1.5
PTB	**1.2**	2.0	**1.3**	3.0	**1.4**	2.8	**2.0**	2.8			**0.9**	2.7	**2.6**	2.8
NPL	**0.2**	1.2	**0.4**	1.4	**0.5**	0.9	**1.0**	1.4	**-0.9**	2.7			**1.7**	0.9
SCK-CEN	**-1.5**	1.2	**-1.3**	1.5	**-1.2**	0.9	**-0.7**	1.5	**-2.6**	2.8	**-1.7**	0.9		

Figure 1. Graph of degrees of equivalence with the KCRV for ^{123}I
(as it appears in Appendix B of the MRA)



Appendix 1. Uncertainty budgets for the activity of ^{123}I submitted to the SIR

No uncertainty budgets have been submitted for the SIR results.

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 for example 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 $u_i = u_{Aei}$ its uncertainty.

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

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

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

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

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.

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

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

Then the total combined uncertainty becomes

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

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

or, after recombination

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

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

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

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

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

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

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

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.

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

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

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

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

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

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**
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	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
bremsstrahlung	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

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-00-00-00
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG