

**BIPM comparison BIPM.RI(II)-K1.Sm-153 of  
activity measurements of the radionuclide  $^{153}\text{Sm}$**

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**Abstract**

Since 1998, four national metrology institutes (NMI) have submitted seven samples of known activity of  $^{153}\text{Sm}$  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.Sm-153. The activities ranged from about 125 MBq to 180 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 four NMIs. A graphical presentation is also given.

**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  $^{226}\text{Ra}$  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.Sm-153 key comparison.

## 2. Participants

Four NMIs have submitted seven ampoules for the comparison of  $^{153}\text{Sm}$  activity measurements since 1998. Another NMI also participated but ultimately withdrew its result because of unidentified impurities. The laboratory details are given in Table 1.

**Table 1. Details of the participants in the BIPM.RI(II)-K1.Sm-153**

<b>NMI</b>	<b>Full name</b>	<b>Country</b>	<b>Regional metrology organization</b>	<b>Date of measurement at the BIPM</b>
NIST	National Institute of Standards and Technology	United States	SIM	1998-06-23
PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1999-04-27
NPL	National Physical Laboratory	United Kingdom	EUROMET	1999-07-09
BNM-LNHB	Bureau national de métrologie-Laboratoire national Henri Becquerel	France	EUROMET	2003-09-11

## 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, the PTB, NPL and the BNM-LNHB have provided uncertainty budgets.

The half-life used by the BIPM is 46.284 (4) hours [3].

**Table 2. Standardization methods of the participants for  $^{153}\text{Sm}$** 

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
NIST	Pressurized $4\pi$ ionization chamber* 4P-IC-GR-00-00-00	46.27 (1) h	179 500	98-06-23 12 h UT	0.01	0.75
PTB	LSC and $4\pi(\text{PC})\beta\text{-}\gamma$ coinc. 4P-LS-BP-00-00-00 4P-PC-BP-NA-GR-CO	1.9284 (4) d	125 260 125 430 <sup>†</sup>	99-04-22 0 h UT	0.04 <sup>‡</sup>	0.50
NPL	$4\pi\text{PPC-}\gamma(\text{NaI})$ using DCC 4P-PP-BP-NA-GR-DC	46.285 (4) h	141 270	99-07-02 12 h UT	0.06	0.27
BNM-LNHB	TDCR 4P-LS-MX-00-00-TD	46.2838 (26) h	159 470 156 380 148 950 <sup>†</sup>	03-09-10 12 h UT	0.02	0.17

<sup>†</sup> more than one ampoule submitted

\* calibrated by  $4\pi\beta$  liquid scintillation counting in 1985 4P-LS-BP-00-00-00.

<sup>‡</sup> re-evaluation according to the GUM [4] has resulted in a smaller value than originally submitted.

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.

#### 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  $^{153}\text{Sm}$  arise from seven 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 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].

**Table 3. Details of the solution of  $^{153}\text{Sm}$  submitted**

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 <sup>†</sup>
NIST	SmCl <sub>3</sub> in HCl	1.05	SmCl <sub>3</sub> : 265	1.016	<sup>156</sup> Eu : 0.105 (8) % * <sup>154</sup> Eu : 0.034 (2) %
PTB	SmCl <sub>3</sub> in HCl	0.1	Sm <sup>+++</sup> : 20	0.999	<sup>156</sup> Eu : 9.0 (8) × 10 <sup>-3</sup> % <sup>154</sup> Eu : 3.95 (5) × 10 <sup>-3</sup> % <sup>152</sup> Eu : 9.90 (10) × 10 <sup>-4</sup> % <sup>155</sup> Eu : 5.35 (16) × 10 <sup>-4</sup> % <sup>153</sup> Gd : 1.4 (3) × 10 <sup>-4</sup> %
NPL	SmCl <sub>3</sub> in HCl	1.0	Sm : 20	1.05	<sup>156</sup> Eu : 5.0 (6) × 10 <sup>-3</sup> % <sup>154</sup> Eu : 3.7 (6) × 10 <sup>-3</sup> % <sup>152</sup> Eu : 2.0 (2) × 10 <sup>-3</sup> %
BNM-LNHB	SmCl <sub>3</sub> in HCl	0.1	Sm : 20	1	<sup>156</sup> Eu : 4.9 (5) × 10 <sup>-3</sup> % †† <sup>154</sup> Eu : 2.0 (2) × 10 <sup>-3</sup> % <sup>152</sup> Eu : 1.4 (1) × 10 <sup>-3</sup> % <sup>153</sup> Gd : 2.5 (1.3) × 10 <sup>-4</sup> %

<sup>†</sup> the ratio of the activity of the impurity to the activity of  $^{153}\text{Sm}$  at the reference date

<sup>††</sup> the relative activity of the europium impurities were also measured at the BIPM, giving values in agreement within the standard uncertainties.

\* original NIST value was 0.126 (9) % (see paragraph below).

Following the advice of the CCRI(II), measurements were made at the BIPM concerning the impurities noted by the NIST, giving a different value for the activity of  $^{156}\text{Eu}$ . A discussion of the results and the evaluation of the agreed final activity ratio for this impurity, 0.105 (8) %, are given in [8].

The SIR global impurity corrections are about  $4 \times 10^{-2}$  for all NMIs except the BNM-LNHB where the corrections are 10 times smaller. All the impurity corrections have been updated for the present report, using recent data for the response of the SIR to the impurities.

Measurements repeated at the BIPM after periods of up to six days later produced comparison results in agreement within standard uncertainties for the four laboratories. These measurements confirm the validity of the half-life value used and the evaluation of the impurity corrections.

In principle, the chemical composition of the solutions could have an influence on the SIR measurements owing to the intense x-ray emission from  $^{153}\text{Sm}$ . However, using the efficiency curve of the SIR [9], the contribution of the x-rays to the ionization current is estimated to about 17 %. In consequence, the influence of the chemical composition on the SIR measurements is probably negligible compared to other

sources of uncertainties in this case, although a more detailed study could be performed.

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.

No international or regional comparison for this radionuclide has been held to date so no linking data are identified.

**Table 4. Results of SIR measurements of  $^{153}\text{Sm}$**

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 $u_{c,i} / \text{kBq}$
NIST	3.758 49	179 500	4	576 800	$33 \times 10^{-4}^*$	4700
PTB	3.696 13	125 260	2	571 700	$22 \times 10^{-4}^*$	3100
	3.701 20	125 430		571 700 <sup>†</sup>		3100
NPL	3.557 12	141 270	1	574 700	$42 \times 10^{-4}^*$	2900
BNM-LNHB	3.884 7 (1)	159 470	4	571 740	$10 \times 10^{-4}$	1100
	3.809 5 (1)	156 380		571 760		1100
	3.628 5 (1)	148 950		571 670 <sup>†</sup>		1100

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

\* mainly due to the uncertainty of the activity of the impurities.

#### 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:

- only primary standardized solutions are accepted, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;
- each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);
- 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;
- 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  $^{153}\text{Sm}$  has been identified as 573 700 (1 200) kBq using the results from all four NMIs.

## 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

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

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

$$D_{ij} = D_i - D_j = A_{e_i} - A_{e_j} \quad (3)$$

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. 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  $^{153}\text{Sm}$ , BIPM.RI(II)-K1.Sm-153 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. Other results may be added as and when other NMIs contribute  $^{153}\text{Sm}$  activity measurements to this comparison.

## Acknowledgements

The authors would like to thank the NMIs 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. Table of degrees of equivalence and introductory text for <sup>153</sup>Sm  
Key comparison BIPM.RI(II)-K1.Sm-153**

**MEASURAND :** Equivalent activity of <sup>153</sup>Sm

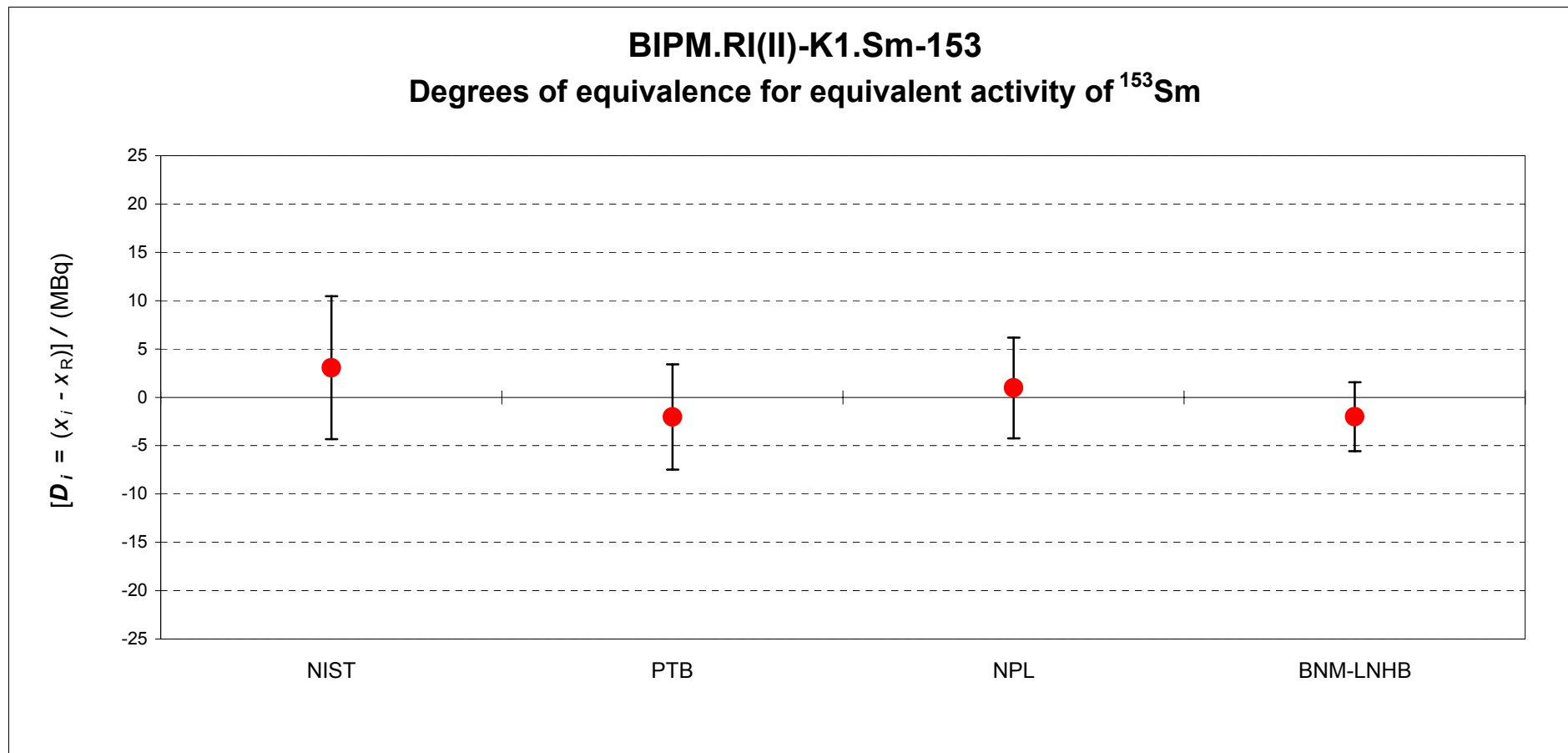
**Key comparison reference value:** the SIR reference value  $x_R$  for this radionuclide is 573.7 MBq, with a standard uncertainty,  $u_R = 1.2$  MBq (see Section 4.1 of the Final Report). the value  $x_i$  is taken as the equivalent activity for laboratory  $i$ .

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms:  $D_i = (x_i - x_R)$  and  $U_i$ , its expanded uncertainty ( $k = 2$ ), both expressed in MBq, and  $U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\sum u_i^2)^{1/2}$  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 numbers:  $D_{ij} = D_i - D_j = (x_i - x_j)$  and  $U_{ij}$ , its expanded uncertainty ( $k = 2$ ), both expressed in MBq. The approximation  $U_{ij}^2 \sim 2^2(u_i^2 + u_j^2)$  is used in the following table.

Lab $i$ ↓			Lab $j$ →							
	$D_i$ $U_i$ / MBq		NIST		PTB		NPL		BNM-LNHB	
	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$	$D_{ij}$	$U_{ij}$
	/ MBq		/ MBq		/ MBq		/ MBq		/ MBq	
NIST	3.1	7.4								
PTB	-2.0	5.4	-5.1	11.3			-3.0	8.5	0.0	6.6
NPL	1.0	5.2	-2.1	11.0	3.0	8.5			3.0	6.2
BNM-LNHB	-2.0	3.6	-5.1	9.7	0.0	6.6	-3.0	6.2		

**Figure 1.** Graph of degrees of equivalence with the KCRV for  $^{153}\text{Sm}$   
(as it appears in Appendix B of the MRA)



**Appendix 1. Uncertainty budgets for the activity of  $^{153}\text{Sm}$  submitted to the SIR****PTB 1999**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
statistics (current measurements of $^{153}\text{Sm}$ 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	–	2.3
dilution factor	–	2.5
radionuclide impurities ( $^{154}\text{Eu}$ , $^{156}\text{Eu}$ )	–	25
half life	–	<0.5
geometry correction	–	1.7
calibration coefficient for $^{153}\text{Sm}$	–	43
<b>Quadratic summation</b>	<b>3.9</b>	<b>50.2</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>50.3</b>	

**NPL 1999**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	4	–
pile up	–	10
timing	–	0.2
impurity : Eu-152	–	1
impurity : Eu-154	–	2
impurity : Eu-156	–	1.5
weighing	–	6
dead time	–	4
resolving time	–	7
background	–	1.6
half-life	–	3
extrapolation	–	12
polynomial fit	–	19
dead time formula	–	0.4
<b>Quadratic summation</b>	<b>4</b>	<b>27</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>27</b>	

**BNM-LNHB 2003**

<b>Relative standard uncertainties</b>	$u_i \times 10^4$ evaluated by method	
	<b>A</b>	<b>B</b>
counting statistics	2	–
weighing	–	5
dilution	–	1.3
dead time	–	1
counting time	–	1
background	–	5
pile-up	–	1
half-life	–	1
decay-scheme parameters	–	10
input parameters and statistical model	–	10
quenching parameter	–	4
radionuclide impurities	–	4.3
<b>Quadratic summation</b>	<b>2</b>	<b>17</b>
<b>Relative combined standard uncertainty, <math>u_c</math></b>	<b>17</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 with efficiency tracing		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, $\gamma$ high efficiency counting		4P-CS-MX-00-00-HE
calibrated IC		4P-IC-GR-00-00-00
internal gas counting		4P-PC-BP-00-00-IG