

BIPM comparison BIPM.RI(II)-K1.Ho-166m
of activity measurements of the radionuclide $^{166}\text{Ho}^m$
and links for the 2000 regional comparison APMP.RI(II)-K2.Ho-166m

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Abstract

Since 1989, two national metrology institutes have submitted three samples of known activity of $^{166}\text{Ho}^m$ to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures. The activities were between about 100 kBq and 500 kBq. 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 these two NMIs. 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.Ho-166m. The results of an APMP regional comparison, comparison identifier APMP.RI(II)-K2.Ho-166m, completed in 2000 for this radionuclide have been linked to the SIR results through that of the NMIJ. This has enabled five other NMIs to have degrees of equivalence in the KCDB.

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) that was set up under the Mutual Recognition

Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Ho-166m key comparison.

In addition, an APMP comparison for this radionuclide, APMP.RI(II)-K2.Ho-166m, was held in 2000 with the NMIJ as the pilot laboratory. Although eleven laboratories took part in this comparison, only five NMIs in addition to the NMIJ are eligible to be linked to the BIPM key comparison.

2. Participants

Two NMIs have submitted three ampoules to the SIR for the comparison of $^{166}\text{Ho}^m$ activity measurements since 1989. 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 the latter are used in the KCDB.

Table 1a. Details of the participants in the BIPM.RI(II)-K1.Ho-166m

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	1989-11-17
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1999-03-01

The five eligible NMIs that took part in the APMP regional comparison, APMP.RI(II)-K2.Ho-166m in 2000 are shown in Table 1b.

Table 1b. Details of the participants in the APMP.RI(II)-K2.Ho-166m of 2000

NMI	Full name	Country
CNEA	Comision Nacional de Energia Atomica	Argentina
INER	Institute of Nuclear Energy Research	Chinese Taipei
KRISS	Korea Research Institute of Standards and Science	Korea
LNMRI	Laboratorio Nacional de Metrologia das Radiações Ionizantes	Brazil
NIM	National Institute of Metrology	China

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. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. Consequently, the uncertainty budget provided by the NMIJ is given in Appendix 1 attached to this report.

The half-life used by the BIPM is 1200 (200) years [3].

Details of the standardization methods used in the APMP comparison may be obtained from [4].

Table 2. Standardization methods of the SIR participants for $^{166}\text{Ho}^m$

NMI	Method used and acronym (see Appendix 3)	Half-life / a	Activity A_i / kBq	Reference date yyyy-mm-dd	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
BNM-LNHB	$4\pi\gamma(\text{NaI}(\text{Tl}))$ 4P-NA-GR-00-00-00	1200 (200) [3]	110.7 110.9	1989-10-21 2 h UT	0.13	0.13
NMIJ	$4\pi\beta\text{-}\gamma$ coincidence 4P-PC-BP-NA-GR-CO	1200	471	1999-03-01 0 h UT	0.31	0.15

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 for the APMP comparison are given in [4] and indicated in Table 3.

Table 3. Details of the solution of $^{166}\text{Ho}^m$ submitted to the SIR

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. / (μg g ⁻¹)	Density / (g cm ⁻³)	Relative activity of impurity *
BNM-LNHB	Ho in HCl	1	–	0.998	¹³⁴ Cs : 1.19 (5) % ¹⁶⁰ Tb : 0.18 (1) % ¹⁵⁴ Eu : 0.21 (5) % ¹⁵² Eu : 0.32 (2) %
NMIJ [†]	HoCl ₃ in HCl	0.1	HoCl ₃ : 7700	1.01	¹⁵² Eu : 0.15 (5) % ¹⁵⁴ Eu : 0.60 (10) %

* the ratio of the activity of the impurity to the activity of $^{166}\text{Ho}^m$ at the reference date

† the same solution as used in the APMP comparison.

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 $^{166}\text{Ho}^m$ arise from three ampoules and the SIR equivalent activity for each ampoule, A_{ei} , is given in Table 4a for each NMI, i . The dates of measurement in the SIR are given in Table 1. The relative standard uncertainties arising from the measurements in the SIR are also shown. 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 corrections for impurity are generally small, with a maximum value of 1.018 for BNM-LNHB (1989). The BNM-LNHB ampoules were measured again in the SIR after 13 years, at which time the impurity correction is ten times lower because of the decay. The results of these later measurements are also given in Table 4a and show a better coherence between the two ampoules than the original SIR measurements. Consequently, the results of the second measurements are more robust and are used for both the KCRV and the KCDB.

The NMIJ ampoule was measured five weeks before the reference date, at the reference date and again four years later. All three results agree to within 6×10^{-4} which is within the SIR uncertainty.

The results of the APMP comparison have been published [4]. The five laboratories to be added to the matrix of degrees of equivalence from this publication are those given in Table 1b. The results for these five NMIs are all linked to the SIR through the result of the NMIJ using the simple ratio from $A_{ei} = [(A/m)_i / (A/m)_{\text{NMIJ}}] \times A_{\text{eNMIJ}}$ as shown in Table 4b.

Table 4a. Results of SIR measurements of $^{166}\text{Ho}^m$

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	3.667 74	110.7	1	9 995	19×10^{-4}	26
	3.674 32	110.9	1	10 017	15×10^{-4}	24
				10 015*	18×10^{-4}	26
				10 012†		26
NMIJ	3.612 12	471	2	10 051	11×10^{-4}	36

* second measurement on 26/02/03 of the same ampoules (see text)

† the mean of the two A_e values is used with an averaged uncertainty, as attributed to an individual entry [8].

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, 11×10^{-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 five laboratories, the CNEA, INER, KRIS, LNMRI and the NIM are given in Appendix 1.

Table 4b. Results of APMP measurements of $^{166}\text{Ho}^m$ and links to the SIR

NMI	Measurement method	Date reported to pilot laboratory	Activity concentration measured $(A/m)_i$ / (kBq g ⁻¹) $(u_{rel})_i$	Equivalent SIR activity A_{ei} / kBq	Combined standard uncertainty $u_{c,i}$ / kBq
NMIJ	$4\pi\beta-\gamma$ coincidence	01-Mar-99	130.4 (0.35 %)	10 051*	36
CNEA	$4\pi\beta-\gamma$ coincidence	03-Jun-99	128.5 (2.5 %)	9 905	250
INER	$4\pi\beta-\gamma$ coincidence	13-Jul-99	130.5 (0.45 %)	10 059	47
KRIS	$4\pi\beta-\gamma$ coincidence	17-Jan-00	128.8 (0.2 %)	9 928	23
LNMRI	$4\pi\beta-\gamma$ coincidence	30-Jun-00	132.5 (0.4 %)	10 213	42
NIM	Ge spectrometry	23-Jun-00	131.2 (0.66 %)	10 113	68

* SIR measured value, see Table 4a

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 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 $^{166}\text{Ho}^m$ has been identified as 10 033 (19) kBq using the results from the BNM-LNHB and the NMIJ.

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

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

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 manner 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 known correlations into account where possible.

The results of the APMP comparison have been linked to those of the SIR through the NMIJ. The degrees of equivalence to 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.

5. Conclusion

The BIPM ongoing key comparison for $^{166}\text{Ho}^m$, BIPM.RI(II)-K1.Ho-166m currently comprises two 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 five other NMIs in the APMP key comparison for $^{166}\text{Ho}^m$ have been linked to the BIPM ongoing key comparison through the common participant, 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 $^{166}\text{Ho}^m$ activity measurements to the SIR comparison or take part in other linked regional comparisons.

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Table 5. Introductory text and table of degrees of equivalence for $^{166}\text{Ho}^m$

Key comparison BIPM.RI(II)-K1.Ho-166m

MEASURAND : Equivalent activity of $^{166}\text{Ho}^m$

Key comparison reference value: the SIR reference value for this radionuclide is $x_R = 10.033$ MBq with a standard uncertainty, $u_R = 0.019$ MBq.

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 MBq, 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 MBq.

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

Linking APMP.RI(II)-K2.Ho-166m to BIPM.RI(II)-K1.Ho-166m

The value x_i is the equivalent activity for laboratory i participant in APMP.RI(II)-K2.Ho-166m 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 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.

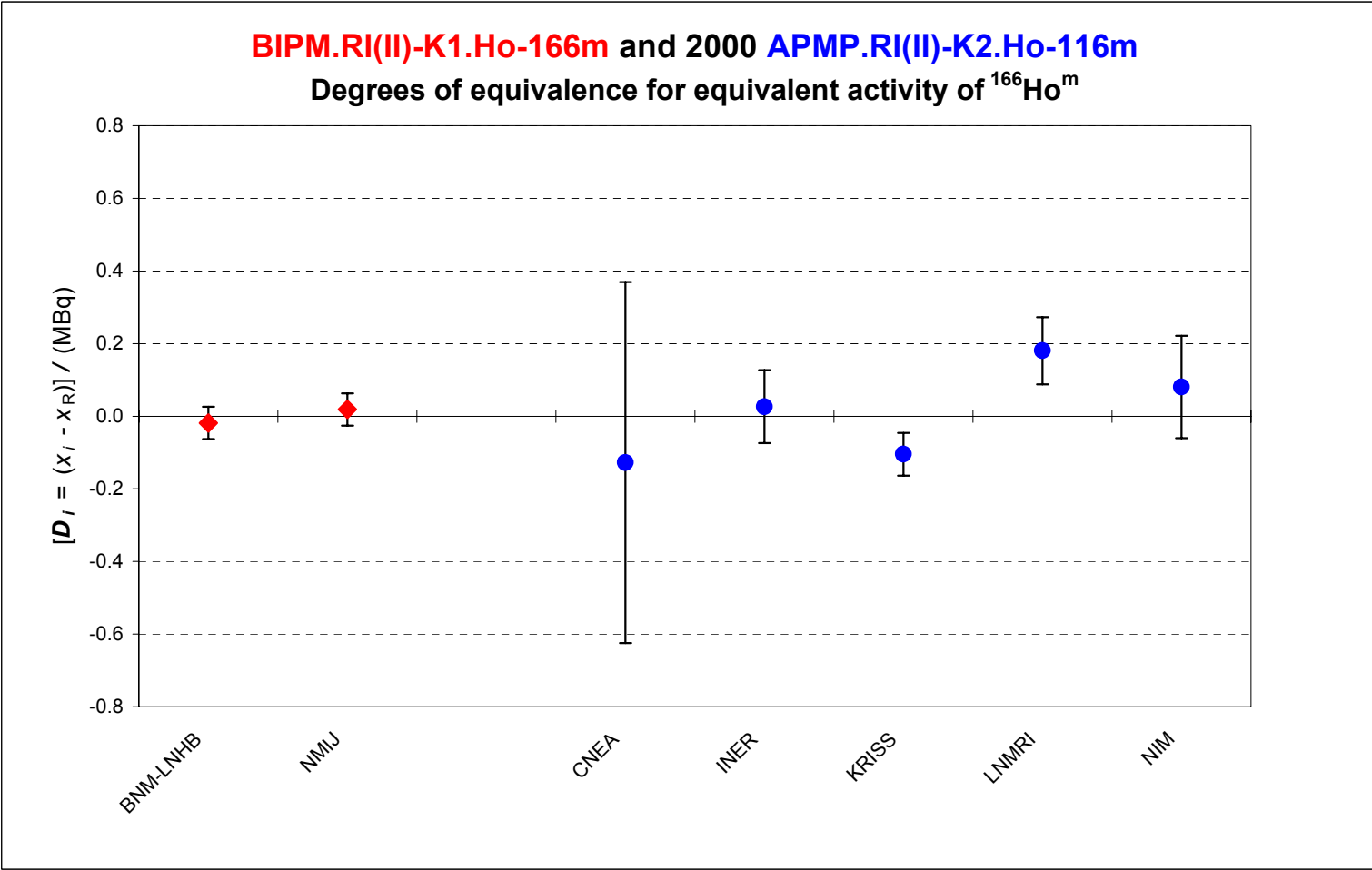
The degree of equivalence between two laboratories i and j , one participant in BIPM.RI(II)-K1.Ho-166m and one in APMP.RI(II)-K2.Ho-166m, or both participants in APMP.RI(II)-K2.Ho-166m, is given by a pair of terms expressed in MBq: $D_{ij} = D_i - D_j$ and U_{ij} , its expanded uncertainty ($k = 2$), approximated by $U_{ij} = 2(u_i^2 + u_j^2 - 2fu_i^2)^{1/2}$ with l being the linking laboratory when each laboratory is from the APMP and f is the correlation coefficient.

These statements make it possible to extend the BIPM.RI(II)-K1.Ho-166m matrices of equivalence to all participants in APMP.RI(II)-K2.Ho-166m.

Table 5 continued. Degrees of equivalence for $^{166}\text{Ho}^m$

Lab <i>i</i> ↓		Lab <i>j</i> →													
		BNM-LNHB		NMIJ		CNEA		INER		KRISS		LNMRI		NIM	
		D_i	U_i	D_{ij}	U_{ij}	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		/ MBq		/ MBq	
BNM-LNHB		-0.02	0.04			0.11	0.50	-0.04	0.11	0.09	0.07	-0.20	0.10	-0.10	0.15
NMIJ		0.02	0.04	0.04	0.09	0.15	0.50	-0.01	0.11	0.12	0.08	-0.16	0.11	-0.06	0.15
CNEA		-0.13	0.50	-0.11	0.50			-0.15	0.50	-0.02	0.50	-0.31	0.50	-0.21	0.51
INER		0.03	0.10	0.04	0.11	0.15	0.50			0.13	0.10	-0.15	0.12	-0.05	0.16
KRISS		-0.10	0.06	-0.09	0.07	0.02	0.50	-0.13	0.10			-0.29	0.09	-0.18	0.14
LNMRI		0.18	0.09	0.20	0.10	0.31	0.50	0.15	0.12	0.29	0.09			0.10	0.16
NIM		0.08	0.14	0.10	0.15	0.21	0.51	0.05	0.16	0.18	0.14	-0.10	0.16		

Figure 1. Graph of degrees of equivalence with the KCRV for $^{166}\text{Ho}^m$
(as it appears in Appendix B of the MRA)



Appendix 1. Uncertainty budgets for the activity of $^{166}\text{Ho}^m$ **Uncertainty budgets submitted to the SIR****NMIJ Uncertainty budget (2000)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	20	–
weighing	–	2
dead time	–	3
background	4	–
coincidence resolving time	–	3
half-life	–	1
linear extrapolation ($4\pi\beta\gamma$ coincidence)	23	–
radionuclide impurities (^{152}Eu and ^{154}Eu)	–	15
Quadratic summation	31	16
Relative combined standard uncertainty, u_c	35	

Budgets submitted for the APMP comparison (2000)**Uncertainty evaluation of the CNEA for the activity of Ho-166m**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	7	–
weighing	–	5
dead time	–	4
background	25	–
timing	–	20
half-life	–	<1
extrapolation of efficiency curve (coincidence method)	250	–
Quadratic summation	251	21
Relative combined standard uncertainty, u_c	252	

INER Uncertainty budget

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	34	–
weighing	–	22
dead time and resolving time	–	8
background	–	5
time base	–	1
half-life	–	1
extrapolation	16	–
Quadratic summation	38	24
Relative combined standard uncertainty, u_c	45	

KRISS Uncertainty budget

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	14	–
weighing	–	3
dead time	–	2
background	–	6
timing	–	3
half-life	–	2
extrapolation	–	11
Quadratic summation	14	14
Relative combined standard uncertainty, u_c	19	

LNMRI Uncertainty budget

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics (20×10^{-4} included in the fitting)	*	–
weighing	–	5
dead time	–	14
resolving time	–	8
delay mismatch	–	6
pile up	–	–
background	–	1
timing	–	–
half-life	–	–
adsorption	–	–
extrapolation	36	–
Quadratic summation	36*	18
Relative combined standard uncertainty, u_c	40	

NIM Uncertainty budget

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	8.7	–
weighing	–	2
pile up	–	7
timing	–	1
half-life	–	–
peak analysis method	–	7
gamma ray probability	–	36
full energy peak efficiency	–	54
coincidence summing	–	4
Quadratic summation	8.7	66
Relative combined standard uncertainty, u_c	66	

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, is 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 2. Acronyms used to identify different measurement methods

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

Geometry	acronym	Detector	acronym
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop counter	PP
2π	2P	liquid scintillation counting	LS
undefined solid angle	UA	NaI(Tl)	NA
		Ge(HP)	GH
		Ge-Li	GL
		Si-Li	SL
		CsI	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

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