Update of the BIPM comparison BIPM.RI(II)-K1.Ho-166m activity measurements of the radionuclide ^{166m}Ho for the PTB (Germany), with linked results for the EURAMET.RI(II)-K2.Ho-166m comparison

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Abstract

The PTB submitted directly to the SIR one ampoule of ^{166m}Ho prepared from a dilution of the solution distributed to four laboratories participating in the EURAMET.RI(II)-K2.Ho-166m comparison in 2013. No attempt to evaluate a new KCRV has been made and the usual procedure was applied to link the results of the comparison to the usual SIR entries through the PTB. One NMI used this K2 comparison to update its previous degree of equivalence older than 20 years. The degrees of equivalence between each equivalent activity measured in the International Reference System (SIR) and the KCRV have been calculated and the results are given in the form of a table for the four remaining laboratories in the BIPM.RI(II)-K1.Ho166m comparison, the six laboratories in the regional comparison APMP.RI(II)-K2.Ho166m and the four laboratories in the regional EURAMET.RI(II)-K2.Ho166m comparison. A graphical presentation is also given.

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 with 3.6 g of the radionuclide in liquid form. For radioactive gases, a different standard ampoule is used. Each 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 ²²⁶Ra 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].

From its inception until 31 December 2013, the SIR has measured 973 ampoules to give 728 independent results for 67 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 standardizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the CIPM Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the

BIPM.RI(II)-K1.Ho-166m key comparison and the previous results were published in 2003 [3] and 2009 [8].

In addition, a APMP comparison for this radionuclide, APMP.RI(II)-K2.Ho-166m, was held in 2000 with the NMIJ as the pilot laboratory [3-4]. Although eleven laboratories took part in this comparison, only six NMIs in addition to the NMIJ are eligible to be linked to the BIPM key comparison through the NMIJ to the new key comparison reference value (KCRV). Recently, in 2013, another regional comparison EURAMET.RI(II)-K2.Ho-166m, was organized with the PTB as coordinating laboratory. Five laboratories took part in this comparison including the PTB who participated in the SIR at the same time, enabling to link the EURAMET.RI(II)-K2 comparison to the BIPM.RI(II)-K1 comparison. The LNE-LNHB uses the comparison to replace their previous result which had been submitted 24 years earlier. In addition, the EURAMET comparison allowed three further laboratories, the CIEMAT, the CMI and the IRMM, linked to the BIPM key comparison through the PTB, to receive their first entry for this radionuclide.

2. Participants

Five NMIs have submitted six ampoules to the SIR for the comparison of ^{166m}Ho 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.

Original acronym	NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
LMRI	LNE- LNHB	Laboratoire national de métrologie et d'essais-Laboratoire national Henri Becquerel	France	EURAMET	1989-11-17
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1999-03-01
-	IRA	Institut de Radiophysique Appliquée	Switzerland	EURAMET	2006-12-22
_	NPL	National Physical Laboratory	United Kingdom	EURAMET	2009-03-02
_	PTB	Physikalisch- Technische Bundesanstalt	Germany	EURAMET	2013-04-03

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

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

The three additional NMIs that took part in the EURAMET regional comparison EURAMET.RI(II)-K2.Ho-166m in 2013 are also eligible and are indicated in Table 1c. The LNE-LNHB used the EURAMET comparison to update their entry in the KCDB.

NMI	Full name	Country
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	Laboratorio Nacional de Metrologia das Radiaçoes Ionizantes	Brazil
NIM	National Institute of Metrology	China
OAP	Office of Atoms for Peace	Thailand

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

Table 1c. Details of the participants in the EURAMET.RI(II)-K2.Ho-166m carried out in 2013 to be linked to BIPM.RI(II)-K1.Ho-166m

NMI	Full name	Country
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	Spain
СМІ	Czech Metrology Institute	Czech Republic
IRMM	EC-JRC Institute for Reference Materials and Measurements	European Union
LNE- LNHB	Laboratoire national de métrologie et d'essais-Laboratoire national Henri Becquerel	France

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 uncertainty budget provided by the PTB is given in Appendix 1 attached to this report.

The half-life used by the BIPM is 1200 (200) years [5]. The value in *Monographie BIPM*-5 Volume 2 is 1200 (180) years [6] but the small difference in the uncertainty would not affect any of the SIR measurement results. The participants in the EURAMET.RI(II)-K2.Ho-166m comparison used, besides the above recommended value, the half-life value 1132.6(3.9) a, as reported by Nedjadi et al. [7]. The latter value was also used by the PTB for the present SIR submission.

NMI	Method used and acronym (see Appendix 2)	Half-life / a	Activity A _i /kBq	Reference date	Relative standard uncertainty / 10 ⁻² by method of evaluation	
				YY-MM-DD	А	В
LNE- LNHB	4πγ(NaI(Tl)) 4P-NA-GR-00-00-00	1200 (200) [5]	110.7 110.9 [#]	1989-10-21 12 h UT	0.13	0.13
NMIJ	$4\pi\beta$ - γ coincidence 4P-PC-BP-NA-GR-CO	1200	471	1999-03-01 0 h UT	0.31	0.15
IRA	$4\pi\beta$ -γ coincidence 4P-PC-BP-NA-GR-CO	1200 (180) [6]	161.4	2006-02-01 0 h UT	0.10	0.19
NPL	$4\pi\beta$ (PPC)- γ coincidence [*] 4P-PP-BP-NA-GR-CO	1200 (200) [5]	69.06	2009-02-15 12 h UT	0.07	0.25
	$4\pi\beta(LS)-\gamma$ coincidence 4P-LS-BP-NA-GR-CO		68.91		0.07	0.25
РТВ	4π(LS) CIEMAT/NIST 4P-LS-MX-00-00-CN 4π(LS) TDCR 4P-LS-MX-00-00-TD	1132.6 (3.9) [7]	134.49**	2013-01-01 0 h UT	0.06 ^{****} 0.07	0.21 ^{***} 0.23

Table 2.	Standardization	methods	of the Sl	IR partici	pants for ^{166m} Ho
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[#] two ampoules submitted

* method selected by the NPL for the KCDB

result obtained as the weighted mean of the results determined with the method 4P-LS-MX-00-CN and the method 4P-LS-MX-00-TD

relative uncertainty of TDCR method adopted by the PTB

Details of the standardization methods used in the APMP.RI(II)-K2.Ho-166m comparison may be obtained from [4]. The uncertainty budgets for the five laboratories, the CNEA, INER, KRISS, LNMRI and the NIM are given in [3]. The OAP uncertainty budget, being designated at a later stage can be found in [8].

Details of the standardization methods used in the EURAMET.RI(II)-K2.Ho-166m comparison may be obtained from [9] as well as the uncertainty budgets for the four linked laboratories, the CIEMAT, the LNE-LNHB, the CMI and the IRMM.

Details regarding the solutions 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. The BIPM standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer is described in [10]. The CCRI(II) agreed in 1999 [11] that this method should be followed according to the protocol described in [12] when an NMI makes such a request or when there appear to be discrepancies. For the IRA solution, trace impurities of ^{152, 154, 155}Eu had been removed by ion-exchange chromatography and the repeated results from the SIR as well as the BIPM impurity check are consistent with no impurity being present. Further details are indicated in section 4.

Details of the solution issued for the APMP comparison are given in [4] but are identical to the NMIJ ampoule in Table 3.

The solution sent to the SIR by the PTB in 2013 was prepared by diluting suitable aliquots of the original solution distributed to the participants in the EURAMET.RI(II)-K2.Ho-166m comparison. The dilution factor used was 3.2093(16); its value was determined by means of a calibrated balance traceable to the National German mass standard. Further details concerning the solution used in the EURAMET.RI(II)-K2.Ho-166m comparison are provided in [9]. Impurity checks carried out on the solution by the PTB using γ -ray spectrometry detected no γ -ray emitting impurity. These results were confirmed by the CMI and the IRMM. No impurity determination was undertaken at the BIPM.

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. $/(\mu g g^{-1})$	Density /(g cm ⁻³)	Relative activity of impurity ¹
LNE- LNHB 1989	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 ² 1999	HoCl ₃ in HCl	0.1	HoCl ₃ : 7700	1.01	¹⁵² Eu : 0.15 (5) % ¹⁵⁴ Eu : 0.60 (10) %
IRA 2006	HoCl ₃ in HCl	0.15	HoCl ₃ : 2300	_	_
NPL 2009	HoCl ₃ in HCl	1	HoCl ₃ : 100	1	¹⁵² Eu : 0.122 (12) % ¹⁵⁴ Eu : 0.239 (24) %
PTB 2013	HoCl ₃ in HCl	1	Unknown ³ : ~ 3900	~1.02	_

¹ the ratio of the activity of the impurity to the activity of ^{166m}Ho at the reference date

² the same solution as used in the APMP comparison.

³ mainly rare earth salts.

4. **Results**

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". The activity measurements for ^{166m}Ho arise from six ampoules, including the recent PTB submission 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 ²²⁶Ra, all the SIR results are normalized to the radium source number 5 [1].

The IRA ampoule was measured ten months after the reference date, again three months later and re-measured once more, two years later. All three SIR results agree to within 5×10^{-4} which is within the SIR uncertainty.

The SIR correction for impurities is 2.6×10^{-3} for the NPL ampoule.

The results of the APMP.RI(II)-K2.Ho-166m comparison have been published [3, 4]. The six laboratories added to the matrix of degrees of equivalence from this publication are those given in Table 1b. The results for these six NMIs are all linked to the SIR through the result of the NMIJ using the simple ratio

$$A_{ei} = \left[\left(A/m \right)_i / \left(A/m \right)_{\text{NMIJ}} \right] \times A_{e\text{NMIJ}} = \left(A/m \right)_i \times 77.08 \text{ as shown in Table 4b.}$$

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 results of the EURAMET.RI(II)-K2.Ho-166m comparison are to be published [9]. Five laboratories have been added to the matrix of degrees of equivalence from this publication and are given in Table 1b. The results of four NMIs are linked to the SIR through the SIR result of the PTB using the simple ratio $A_{ei} = \left[(A/m)_i / (A/m)_{\text{PTB}} \right] \times A_{e\text{PTB}} = (A/m)_i \times 83.07$ as shown in Table 4c.

The uncertainties for the EURAMET.RI(II)-K2.Ho-166m comparison linked to the SIR are comprised of the original NMI uncertainties (given in Table 4c) together with the uncertainty in the link, 17.7×10^{-4} , given by the uncertainty in the SIR measurement of the PTB ampoule of this EURAMET.RI(II)-K2 comparison and finally the uncertainty in the dilution factor 0.05 %.

NMI	Mass of	Activity	N° of	SIR	Relative	Combined
	solution	submitted	Ra	A_e / kBq	uncertainty	uncertainty
	<i>m</i> _i / g	A_i / kBq	source		from SIR	$u_{c,i}$ / kBq
			used			
LNE-	3.667 74	110.7	1	9995	19×10^{-4}	26
LNHB	3.674 32	110.9	1	10 017	$15 imes 10^{-4}$	24
1989				10 015 ^a	18×10^{-4}	26
				$10\ 012^{a,b}$		26
NMIJ	3.612 12	471	2	10 051	11×10^{-4}	36
1999						
IRA	3.536 2 (1)	161.4	1	9913	14×10^{-4}	26
2006						
NPL	3.598 29	69.06 ^c	1	9932 ^c	15×10^{-4}	30
2009		68.91		9909		30
PTB 2013	3.61719	134.49 ^d	1	9912	17.7×10^{-4}	28

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

^a second measurement on 26/02/03 of the same ampoules, with an SIR impurity correction 10 times lower (see [3])

^b using the mean of the two A_e values with an averaged uncertainty, as attributed to an individual entry [13]

^c result from the method 4P-PP-BP-NA-GR-CO designated for the KCDB

^d result obtained as the weighted mean of the results determined with the method 4P-LS-MX-00-CN and the method 4P-LS-MX-00-TD

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

NMI	Measurement method	Date reported to pilot laboratory	Measured activity concentration $(A/m)_i / (kBq g^{-1})$ $(u_{rel})_i$	Equivalent SIR activity A _{ei} /kBq	Combined standard uncertaint y 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 %)	9905	250
INER	$4\pi\beta-\gamma$ coincidence	13-Jul-99	130.5 (0.45 %)	10 059	47
KRISS	$4\pi\beta-\gamma$ coincidence	17-Jan-00	128.8 (0.2 %)	9928	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
OAP	Pressurized IC [#]	30-Jun-00	130.0 (1.3 %)	10 020	130

* SIR measured value, see Table 4a [#] using the IC response curve traceable to the NIST and the NMIJ

NMI	Measurement method	Date reported to pilot laboratory	Measured activity ^a concentration $(A/m)_i / (kBq g^{-1})$ $(u_{rel})_i$	Equivalent SIR activity A _{ei} /kBq	Combined standard uncertainty $u_{c,i}$ / kBq
PTB	4P-LS-MX-00-00-CN 4P-LS-MX-00-00-TD	2013-Feb-22	119.32 ^b (0.21 %)	9912	28
CIEMAT	4P-LS-BP-00-00-CN 4P-NA-GR-00-00-00	2013-Jun-25	119.19 ^c (0.23 %)	9901	30
LNE- LNHB	4P-PC-BP-NA-GR-AC	2013-Jun-26	119.05 ^d (0.27 %)	9890	32
СМІ	4P-PC-BP-NA-GR-CO	2013-Jul-11	119.7 (0.64 %)	9944	67
IRMM	4P-LS-MX-00-00-CN 4P-LS-MX-00-00-TD 4P-CS-PE-00-00-HE 4P-NA-PH-00-00-HE ^e 4P-NA-PH-00-00-HE ^e	2013-Aug-23	119.29 ^f (0.08 %)	9910	20

Table 4c.Results	of EURAMET measurements	s of ^{166m} Ho and links to the SIR
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^a at reference date 1 January 2013 0 h UTC

^b weighted mean of the results obtained with both methods. Uncertainty conservatively taken as that of TDCR method

^c unweighted mean of the results obtained with both methods

^d two other methods have been used giving results in good agreement. The result obtained with the anticoincidence method was taken as the final laboratory result

^e these two methods used well-type NaI(Tl) detectors of different sizes

^f result obtained using the power-moderated mean using a power of 1.4 in the relative weights [14]

4.1 <u>The key comparison reference value</u>

The reduced data set used for the evaluation of the KCRVs is known as the "KCRV file" and is the reduced data set from the SIR master-file. Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are only made by the CCRI(II), normally during one of its biennial meetings, or by consensus through electronic means (e.g., email) as discussed and accepted at the CCRI(II) meeting in 2013.

The KCRV for ^{166m}Ho was evaluated in 2009 as 9978 (33) kBq, the arithmetic mean of the results from the LNE-LNHB, NMIJ, IRA and the NPL[8]. The new entry of the PTB is in close agreement with the values previously reported by the IRA, the NPL and with the first value reported by the LNE-LNHB in 1989 as shown in Table 4 a. This seems to support a lower value of the KCRV and suggest that a new evaluation of it should be undertaken. On this occasion, the new formalism of the power-

moderated mean developed by S. Pommé [14] and accepted by the CCRI(II) will be used.

4.2 <u>Degrees of equivalence</u>

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 CIPM MRA. Normally, the most recent result is the one included. An 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

$$D_i = A_{e_i} - \text{KCRV} \tag{1}$$

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

$$U_i = 2u_{D_i},\tag{2}$$

taking correlations into account as appropriate [15].

4.2.2 Comparison of any two NMIs with each other

The degree of equivalence between any pair of NMIs, i and j, is expressed as the difference D_{ij} in their results

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

and the expanded uncertainty of this difference $U_{ij} = 2u(D_{ij})$, where

$$u_{Dij}^{2} = u_{i}^{2} + u_{j}^{2} - 2u(A_{ei}, A_{ej})$$
⁽⁴⁾

where any obvious correlations between the NMIs (such as a traceable calibration, or correlations normally coming from the SIR or from the linking factor in the case of linked comparison) are subtracted using the covariance $u(A_{ei}, A_{ej})$ (see [16] for more detail). However, the CCRI decided in 2011 that these "pair-wise degrees of equivalence" no longer need to be published as long as the methodology is explained.

Table 5 shows the table of the degrees of equivalence with the KCRV as they appear in 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 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. The graphical representation indicates in part the degree of equivalence between the NMIs but obviously does not take into account the correlations between the different NMIs. It should be noted that the final data in this paper, while correct at the time of publication, will become out-of-date as NMIs make new comparisons. The formal results under the CIPM MRA [2] are those available in the KCDB.

5. Conclusion

The BIPM ongoing key comparison for ^{166m}Ho, BIPM.RI(II)-K1.Ho-166m currently comprises five results. The KCRV for this radionuclide has not been yet re-evaluated and the results have been analysed with respect to the actual KCRV. The results of the EURAMET-K2.Ho-166m comparison held in 2013 have been linked to the BIPM comparison through the mutual participation of the PTB. As a consequence five new results for ^{166m}Ho have been registered and this has enabled the table of degrees of equivalence to include fourteen results in total.

Other results may be added as and when other NMIs contribute ^{166m}Ho activity measurements to the SIR comparison or take part in other linked regional comparisons.

Acknowledgements

The authors would like to thank the NMIs for their participation in this comparison.

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Table 5. Introductory text and table of degrees of equivalence for ^{166m}Ho Key comparison BIPM.RI(II)-K1.Ho-166m

MEASURAND : Equivalent activity of ^{166m}Ho

Key comparison reference value: the SIR reference value for this radionuclide is $x_{R=}9.978$ MBq with a standard uncertainty, $u_{R} = 0.033$ MBq. x_{i} is the equivalent activity for laboratory *i* and x_{R} 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_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 .

When required, 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 $U_{ii} \sim 2(u_i^2 + u_i^2)^{1/2}$ may be used.

Linking APMP.RI(II)-K2.Ho-166m (2000) 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.

When required, 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 $U_{ii} = 2(u_i^2 + u_j^2 - 2fu_i^2)^{1/2}$ with *I* being the linking laboratory when each laboratory is from the APMP and *f* is the correlation coefficient.

Linking EURAMET.RI(II)-K2.Ho-166m (2013) to BIPM.RI(II)-K1.Ho-166m

The value x_i is the equivalent activity for laboratory *i* participant in EURAMET.RI(II)-K2.Ho-166m having been normalized to the value of the PTB as the linking laboratory.

The degree of equivalence of laboratory *i* participant in EURAMET.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.

When required, the degree of equivalence between two laboratories *i* and *j*, one participant in BIPM.RI(II)-K1.Ho-166m and one in EURAMET.RI(II)-K2.Ho-166m,

or both participants in EURAMET.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 *I* being the linking laboratory when each laboratory is from the EURAMET 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 EURAMET.RI(II)-K2.Ho-166m.

Table 5.continued

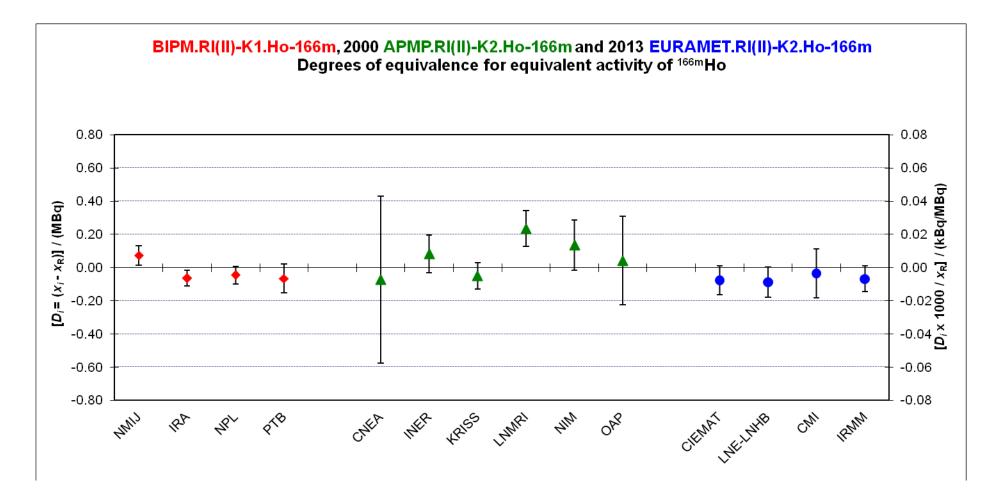
Lab *i* ↓

	Di	U _i	
	/ MBq		
LNE-LNHB	0.04	0.05	
NMIJ	0.07	0.06	
IRA	-0.06	0.05	
NPL	-0.05	0.05	
РТВ	-0.07	0.09	
CNEA	-0.07	0.50	
INER	0.08	0.11	
KRISS	-0.05	0.08	
LNMRI	0.24	0.11	
ΝΙΜ	0.14	0.15	
ΟΑΡ	0.04	0.27	
CIEMAT	-0.08	0.09	

CIEMAI	-0.08	0.09
LNE-LNHB	-0.09	0.09
СМІ	-0.03	0.15
IRMM	-0.07	0.08
	0101	0.00

The first four laboratories are those used for evaluating the KCRV. The second value of LNE-LNHB supersedes the previous one for evaluating the degree of equivalence.

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



Appendix 1. Uncertainty budget for the activity determination of ^{166m}Ho as reported by the PTB – Method (4P-LS-MX-00-00-TD)

Relative standard uncertainties	$u_i \times 10^4$	
	evaluated	by method
Contributions due to	Α	В
counting statistics	5 ^a	_
(in)homogeneity	_	15 ^b
weighing	_	8
dead time	_	3
background	3	
counting time	_	1
adsorption	_	5
impurities	_	3^{c}
TDCR value and fit		1^d
model		5
ionization quenching and kB (model)		3
decay-scheme parameters		6
decay correction		< 1 ^e
quenching indicator		3
dilution		5
PMT asymmetry		2
Quadratic summation	6	21
Relative combined standard uncertainty, <i>u_c</i>	22	

^a standard deviation of the mean of three samples ^b estimation due to a slight discrepancy of one sample ^c no impurity detected ^d includes contribution due to interpolation from calibration curve ^e the half-life value $T_{1/2} = 1132.6(3.9)$ a was used

Relative standard uncertainties	tive standard uncertainties $u_i \times 10^4$	
	evaluated	by method
Contributions due to	Α	В
counting statistics	5^{f}	—
(in)homogeneity	-	15 ^g
weighing	-	8
dead time	-	10
background	5	
counting time	_	1
adsorption	_	5
impurities	_	3 ^h
tracer (³ H)		2^{i}
model		5
ionization quenching and kB (model)		3
decay-scheme parameters		6
decay correction		< 1 ^j
quenching indicator ($SPQ(E)$, ^t SIE)		3
dilution		5
PMT asymmetry		2
Quadratic summation	7	23
Relative combined standard uncertainty, <i>u_c</i>	2	24

Uncertainty budget for the activity determination of $^{166m}{\rm Ho}$ as reported by the PTB – Method (4P-LS-MX-00-00-CN)

^f standard deviation of the mean of three samples ^g estimation due to a slight discrepancy of one sample ^h no impurity detected ⁱ includes interpolation from calibration curve ^j the half-life value $T_{1/2} = 1132.6(3.9)$ a was used

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	Nal(TI)	NA
		Ge(HP)	GH
		Ge(Li)	GL
		Si(Li)	SL
		CsI(TI)	CS
		ionization chamber	IC
		grid ionization chamber	GC
		bolometer	BO
		calorimeter	CA
		PIPS detector	PS
Radiation	acronym	Mode	acronym
positron	PO	efficiency tracing	ET
beta particle	BP	internal gas counting	IG
Auger electron	AE	CIEMAT/NIST CN	
conversion electron	CE	sum counting	SC
mixed electrons	ME	coincidence	СО
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with efficiency tracing	СТ
X - rays	XR	anti-coincidence counting with efficiency tracing	AT
photons (x + γ)	PH	triple-to-double coincidence ratio counting	TD
photons + electrons	PE	selective sampling	SS
alpha - particle	AP	high efficiency	HE
mixture of various radiation	MX	digital coincidence counting	DC

Examples method	acronym
$4\pi(PC)\beta$ - γ -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 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-HE
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG