Update of the BIPM comparison BIPM.RI(II)-K1.F-18 of activity measurements of the radionuclide ^{18}F to include the PTB

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Abstract

Since 2001, six national metrology institutes (NMIs) have submitted six samples of known activity of ^{18}F to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM), the most recent being that of the PTB (Germany). The activities ranged from about 1 MBq to 18 MBq. The key comparison reference value (KCRV) has been recalculated to include the latest value, with the agreement of the CCRI(II). The degrees of equivalence between each equivalent activity measured in the SIR have been recalculated and the results are given in the form of a matrix. A graphical presentation is also given for this key comparison with identifier BIPM.RI(II)-K1.F-18.

1. Introduction

The SIR for activity measurements of γ-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 (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].

From its inception until 31 December 2004, the SIR has measured 872 ampoules to give 634 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.F-18 key comparison.

2. Participants

Six NMIs have submitted six ampoules for the comparison of ^{18}F activity measurements since 2001. The laboratory details are given in Table 1.

Table 1. Details of the participants in the BIPM.RI(II)-K1.F-18

NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	2001-09-21 13 h 10 UT
LNE-LNHB*	Laboratoire national de métrologie et d'essais - Laboratoire national Henri Becquerel	France	EUROMET	2002-04-10 12 h 06 UT
BEV	Bundesamt für Eich- und Vermessungswesen	Austria	EUROMET	2002-11-12 11 h 48 UT
NPL	National Physical Laboratory	United Kingdom	EUROMET	2003-04-29 7 h 46 UT
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	Spain	EUROMET	2004-05-13 10 h 14 UT
PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	2005-04-13 19 h 57 UT

* Previously known as the BNM-LNHB

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 2. The uncertainty budgets for each participant are given in Appendix 1.

The half-life used by the BIPM is 1.8290 (5) hours [3] which is in agreement within the standard uncertainties with 1.8288 (3) hours from Monographie 5 [4]. This difference in half-life value would result in a relative change of less than 3×10^{-4} for the largest time differences between a reference date and the corresponding measurement at the BIPM (for the BEV and the NPL). Consequently, the equivalent activity results in Table 4 have not been modified. Such a short half-life obviously requires a correction for the decay during the SIR measurement.

Table 2. Standardization methods of the participants for ^{18}F

NMI	Method used and acronym (Appendix 3)	Half-life / h	Activity A_i / kBq	Reference date	Relative standard uncertainty × 100 by method of evaluation	
				YY-MM-DD	A	B
IRA	pressurised ionization chamber* 4P-IC-GR-00-00-00	1.829 0 (5)	1 109	01-09-21 14 h 00 UT	0.06	0.37
LNE-LNHB	liquid scintillation using TDCR 4P-LS-PO-00-00-TD	1.829 0 (5)	6 793	02-04-10 12 h 00 UT	0.98	0.21
BEV	pressurised ionization chamber[#] 4P-IC-GR-00-00-00	1.829 5	7 722	02-11-12 05 h 00 UT	0.03	1.01
NPL	$4\pi\beta$(PC)-γ coincidence 4P-PC-BP-NA-GR-CO	1.829 5	2 312	03-04-29 0 h 00 UT	0.08	0.23
CIEMAT	pressurized IC calibrated in April 2004 by $4\pi\beta$(PPC)-γ coincidence [+] 4P-PP-PO-NA-GR-CO and CIEMAT/NIST 4P-LS-PO-00-00-CN	1.828 57 (27)	18 430 [+] 18 540	04-05-13 10 h 00 UT	0.21	0.60 [+] 0.43
PTB	pressurized IC calibrated in April 2005 by $4\pi\beta$(PC)-γ coincidence 4P-PC-BP-NA-GR-CO and CIEMAT/NIST 4P-LS-MX-00-00-CN	1.829 5 (10)	6 146	05-04-13 23 h 00 UT	0.06	0.31

* calibrated in 1997 by $4\pi\gamma$(NaI) counting 4P-NA-GR-00-00-00 and liquid scintillation 4P-LS-PO-00-00-00

[#] secondary standard, calibrated by the NPL

[+] to be used for the KCRV and KCDB.

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 [5] was approved by the 1999 CCRI(II) [6]. The method follows the protocol described in [7] when an NMI makes such a request or when there appear to be discrepancies. As ^{18}F has a very short half-life, provisional measurements were made and impurities were indeed identified in the IRA and the PTB ampoules.

Table 3. Details of the solution of ^{18}F submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity[†]
IRA	F in HCl	0.1	NaCl: 50	0.999 (9)	^{52}Mn: 0.0123 (2) % ^{55}Co: 0.0082 (2) % ^{56}Co: 0.0028 (1) % ^{58}Co: 0.0049 (1) %
LNE-LNHB	FDG* in HCl	0.1	NaCl: 56	0.97 (1)	–
BEV	F in water	–	NaCl: 9400	[‡]	–
NPL	F in water	–	FDG*	1.000	–
CIEMAT	F in water	–	NaCl: 1	1	–
PTB	NaF in water	–	NaF: 55	1.00	^{52}Mn: 2.1 (2) × 10^{-3} % ^{56}Co: 4.1 (1) × 10^{-3} % ^{57}Co: 1.1(2) × 10^{-4} %

[†] the ratio of the activity of the impurity to the activity of ^{18}F at the reference date
[‡] density estimated from [8] : 1.005
* FDG fluoro-deoxy-glucose

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 ^{18}F arise from six 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].

Measurements repeated at the BIPM for up to one half-life produced the same comparison results for the IRA and the LNE-LNHB, and a result in agreement within the SIR uncertainty for the PTB. The BEV, NPL and the CIEMAT ampoules were measured for up to two half-lives and also produced the same comparison results. The impurity corrections for the SIR measurement of the IRA and the PTB ampoules are less than 10^{-3}.

As no submission has been identified as a pilot study, the result of each NMI is eligible for Appendix B of the MRA.

An international comparison held in 2001 for this radionuclide has been evaluated [9] and is linked to the SIR comparison through the measurements made by the linking laboratories.

Table 4. Results of SIR measurements of ^{18}F

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
IRA	3.681 2 (1)	1 109	3	15 312	7×10^{-4}	57
LNE-LNHB	3.499 5 (5)	6 793	4	15 170	5×10^{-4}	150
BEV	3.623 7	7 722	2	15 390	12×10^{-4}	160
NPL	3.774 66	2 312	5	15 281	7×10^{-4}	39
CIEMAT	3.548 3	18 430 18 540	5	15 216* 15 303	6×10^{-4}	97* 73
PTB	3.639 9 (9)	6 146	5	15 316	7×10^{-4}	50

* this CIEMAT value used for the KCRV and KCDB (see Table 2).

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. The key comparison reference value for ^{18}F was previously recorded as 15 245 (32) kBq following the provisions above and in consequence by using the results from the IRA, LNE-LNHB, NPL and the CIEMAT as given in [10]. The KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), and such a modification to include the result of the PTB has been approved by the CCRI(II). Consequently, the new value for the KCRV is 15 259 (29) kBq using the results from the IRA, LNE-LNHB, NPL, CIEMAT 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 [11].

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

$$u_{D_{ij}}^{\ 2} = u_i^{\ 2} + u_j^{\ 2} - 2u(A_{e,i}, A_{e,j}) \qquad (4)$$

and any obvious correlations between the NMIs (such as a traceable calibration) are subtracted 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 appear in Appendix B of the KCDB including those of the linked comparison, CCRI(II)-K3.F-18. 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 comparisons. 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 between the BEV and the NPL into account.

Conclusion

The BIPM ongoing key comparison for ^{18}F, BIPM.RI(II)-K1.F-18 currently comprises six results. These have been analysed with respect to the KCRV determined for this radionuclide which now includes the result of the PTB, and with respect to each other. The matrix of degrees of equivalence, including those for the CCRI(II)-K3.F-18 linked comparison 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 ^{18}F activity measurements to this comparison.

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Table 5. Introductory text and table of degrees of equivalence for ^{18}F

Key comparison BIPM.RI(II)-K1.F-18

MEASURAND : Equivalent activity of ^{18}F

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 15.26 MBq, with a standard uncertainty u_R = 0.03 MBq.
x_R is computed as the mean of the results obtained by primary methods.

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, with n the number of laboratories,
$U_i = 2((1-2/n)u_i^2 + (1/n^2)\Sigma u_i^2)^{1/2}$ when each laboratory has contributed to the reference value (see Final Report).

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 CCRI(II)-K3.F-18 to BIPM.RI(II)-K1.F-18

The value x_i is the equivalent activity for laboratory i participant in CCRI(II)-K3.F-18
having been normalized to the value of the NPL and the BNM-LNHB combined as the link.

The degree of equivalence of laboratory i participant in CCRI(II)-K3. 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.F-18 and one in CCRI(II)-K3.F-18,
or both participants in CCRI(II)-K3.F-18, 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 referring to the link when each laboratory is from the CCRI or one of the linking laboratories
and f is the correlation coefficient.

Linking APMP.RI(II)-K3.F-18 to BIPM.RI(II)-K1.F-18

The value x_i is the equivalent activity for laboratory i participant in APMP.RI(II)-K3.F-18
having been normalized to the value of the NPL and the BNM-LNHB combined as the link.

The degree of equivalence of laboratory i participant in APMP.RI(II)-K3. 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 this laboratory did not contribute to the KCRV.

The degree of equivalence between two laboratories i and j, one participant in BIPM.RI(II)-K1.F-18 or in CCRI(II)-K3.F-18 and one in APMP.RI(II)-K3.F-18,
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 referring to the link when the other laboratory is from the CCRI or is one of the linking laboratories and f is the correlation coefficient.

These statements make it possible to extend the BIPM.RI(II)-K1.F-18 matrices of equivalence
to all participants in the CCRI(II)-K3.F-18 and the APMP.RI(II)-K3.F-18 comparisons.

Table 5 continued. Degrees of equivalence for ^{18}F

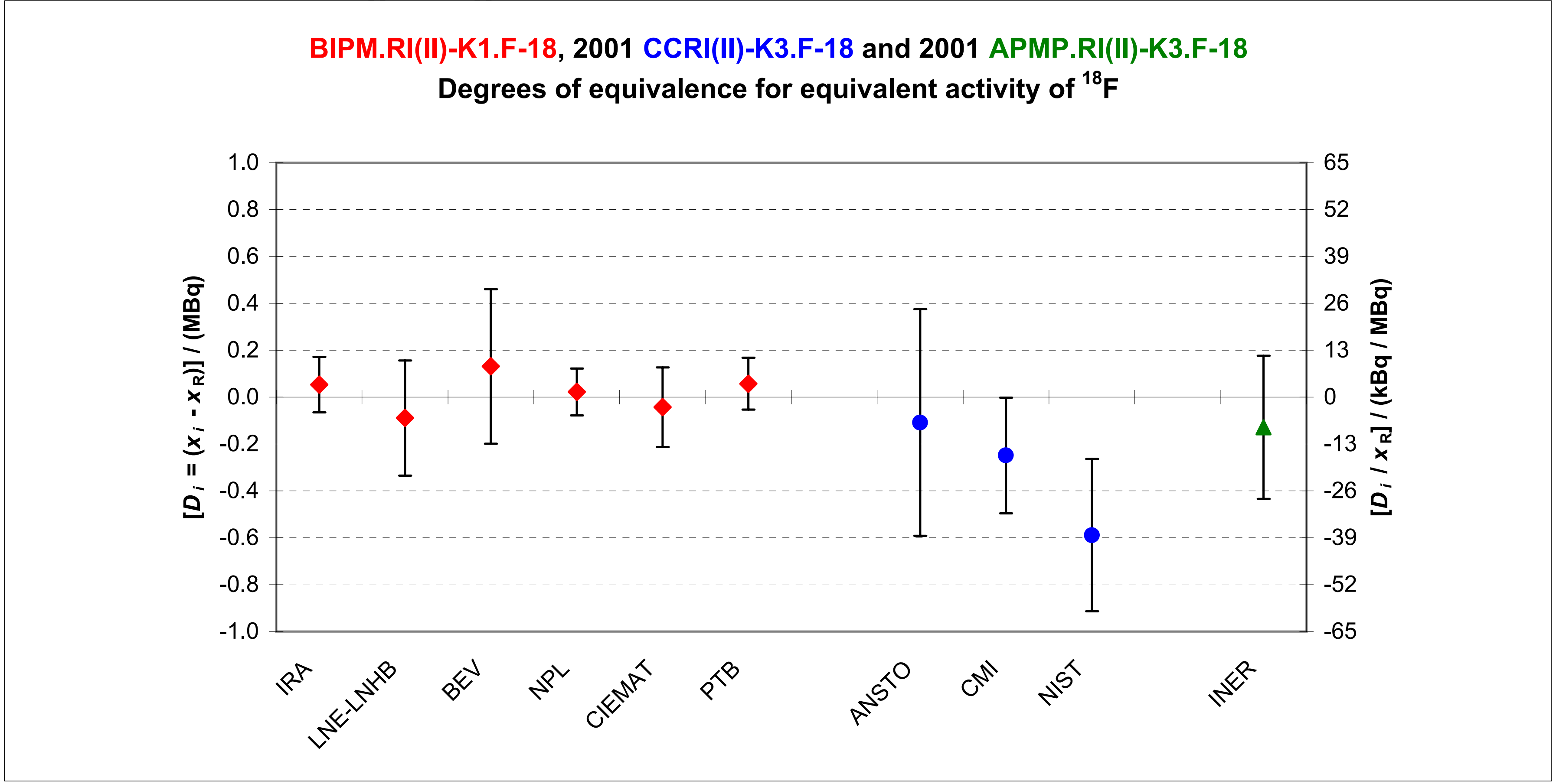
Lab *j* ⇨

Lab *i* ⇩			IRA		LNE-LNHB		BEV		NPL		CIEMAT		PTB	
	D_i / MBq	U_i	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}
IRA	**0.05**	0.12			**0.14**	0.32	**-0.08**	0.34	**0.03**	0.14	**0.10**	0.23	**0.00**	0.15
LNE-LNHB	**-0.09**	0.25	**-0.14**	0.32			**-0.22**	0.44	**-0.11**	0.31	**-0.05**	0.36	**-0.15**	0.32
BEV	**0.13**	0.33	**0.08**	0.34	**0.22**	0.44			**0.11**	0.31	**0.17**	0.37	**0.07**	0.34
NPL	**0.02**	0.10	**-0.03**	0.14	**0.11**	0.31	**-0.11**	0.31			**0.07**	0.21	**-0.04**	0.13
CIEMAT	**-0.04**	0.17	**-0.10**	0.23	**0.05**	0.36	**-0.17**	0.37	**-0.07**	0.21			**-0.10**	0.22
PTB	**0.06**	0.11	**0.00**	0.15	**0.15**	0.32	**-0.07**	0.34	**0.04**	0.13	**0.10**	0.22		
ANSTO	**-0.11**	0.48	**-0.16**	0.49	**-0.02**	0.56	**-0.24**	0.58	**-0.13**	0.47	**-0.07**	0.52	**-0.17**	0.49
CMI	**-0.25**	0.25	**-0.30**	0.27	**-0.16**	0.37	**-0.38**	0.40	**-0.27**	0.23	**-0.21**	0.31	**-0.31**	0.26
NIST	**-0.59**	0.33	**-0.64**	0.34	**-0.50**	0.43	**-0.72**	0.45	**-0.61**	0.31	**-0.55**	0.37	**-0.65**	0.34
INER	**-0.13**	0.31	**-0.18**	0.32	**-0.04**	0.41	**-0.26**	0.44	**-0.15**	0.29	**-0.09**	0.36	**-0.19**	0.32

Lab *i* ⇩			ANSTO		CMI		NIST		INER	
	D_i / MBq	U_i	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}	D_{ij} / MBq	U_{ij}
IRA	**0.05**	0.12	**0.16**	0.49	**0.30**	0.27	**0.64**	0.34	**0.18**	0.32
LNE-LNHB	**-0.09**	0.25	**0.02**	0.56	**0.16**	0.37	**0.50**	0.43	**0.04**	0.41
BEV	**0.13**	0.33	**0.24**	0.58	**0.38**	0.40	**0.72**	0.45	**0.26**	0.44
NPL	**0.02**	0.10	**0.13**	0.47	**0.27**	0.23	**0.61**	0.31	**0.15**	0.29
CIEMAT	**-0.04**	0.17	**0.07**	0.52	**0.21**	0.31	**0.55**	0.37	**0.09**	0.36
PTB	**0.06**	0.11	**0.17**	0.49	**0.31**	0.26	**0.65**	0.34	**0.19**	0.32
ANSTO	**-0.11**	0.48			**0.14**	0.47	**0.48**	0.52	**0.02**	0.50
CMI	**-0.25**	0.25	**-0.14**	0.47			**0.34**	0.31	**-0.12**	0.29
NIST	**-0.59**	0.33	**-0.48**	0.52	**-0.34**	0.31			**-0.46**	0.36
INER	**-0.13**	0.31	**-0.02**	0.50	**0.12**	0.29	**0.46**	0.36		

Figure 1. **Graph of degrees of equivalence with the KCRV for ^{18}F**

(as it appears in Appendix B of the MRA)



N.B. Right-hand axis shows approximate values only

Appendix 1. Uncertainty budgets for the activity of ^{18}F submitted to the SIR

Uncertainty budgets for the IRA:

Evaluation for the 4πγ(NaI) method

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	6	–
weighing	–	5
background	–	2
threshold	–	5
timing	–	2
decay scheme	–	10
efficiency	–	20
Quadratic summation	**6**	**23.6**
Relative combined standard uncertainty, u_c	**24.4**	

Evaluation for the liquid scintillation method

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	15	–
weighing, dilution	–	4
influence of the ^{14}C standard	–	20
effect of radionuclide impurities	–	10
emission probability	–	20
timing	–	60
Quadratic summation	**15**	**67.2**
Relative combined standard uncertainty, u_c	**68.9**	

Evaluation for the reference ionization chamber IG11

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics including decay correction	6	–
combined uncertainty (mean) of the two primary methods above	–	36.8
Quadratic summation	**6**	**36.8**
Relative combined standard uncertainty, u_c	**37.2**	

Uncertainty budget for the LNE-LNHB:

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics, $\nu = 4^{\dagger}$	98	–
weighing	–	5
timing	–	20
half-life	–	3
Quadratic summation	**98**	**21**
Relative combined standard uncertainty, u_c	**100**	

† **number of degrees of freedom**

Uncertainty budget for the BEV:

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	2.7	–
weighing, dilution	–	5
background	–	0.6
half-life	–	5.3
radionuclide impurities	–	–
calibration factor	–	100
ionization chamber	–	10
current measurement	–	10
Quadratic summation	**2.7**	**101.3**
Relative combined standard uncertainty, u_c	**101.3**	

Uncertainty evaluation of the activity of NPL ampoule A 195/03

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting	8	–
counting time	–	1
weighing	–	5
dead-time	–	2
pile-up	–	5
background	–	1
β^+ : EC branching	–	20
half-life	–	8
impurities	–	0.4
extrapolation of efficiency curve	–	5
adsorption	–	0.1
Quadratic summation	**8**	**23**
Total relative combined uncertainty u_c	**25**	

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Uncertainty evaluation of the IC calibration factor using the coincidence method

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting (coincidence measurement)	35	–
IC current measurement	24	–
counting time	–	1
weighing	–	7
dead-time	–	10
resolving time	–	2
Gandy effect	–	1
background	9	–
decay scheme parameters	–	20
half-life	–	2
extrapolation of efficiency curve	–	35
Quadratic summation	**43**	**42**
Total relative combined uncertainty u_c	**60**	

Uncertainty evaluation of the IC calibration factor using the CIEMAT/NIST method

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting (CIEMAT/NIST measurement)	2	–
IC current measurement	24	–
counting time	–	1
weighing	–	5
dead-time	–	2
^{3}H tracer	–	1
quenching	–	2
decay scheme parameters	–	35
half-life	–	3
Quadratic summation	**24**	**36**
Total relative combined uncertainty u_c	**43**	

Uncertainty evaluation of the reference ionization chamber measurements

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
current measurement	21	–
IC calibration factor:	–	
by coincidence		60
by CIEMAT/NIST		43
Quadratic summation	**21**	**60 or 43**
Total relative combined uncertainty u_c	**64 or 48**	

Uncertainty evaluation of the activity of PTB ampoule

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
statistics of current measurements	5	–
radium reference source current	2.5	–
linearity of current measurement	–	5
IC calibration factor	–	30
geometry correction	–	5
adsorption	–	< 0.1
weighing of ampoule	–	2.4
background	–	< 1
half-life	–	4
impurities	–	< 0.1
Quadratic summation	**5.6**	**31.2**
Total relative combined uncertainty u_c	**32**	

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(Tl)	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	CO
bremsstrahlung	BS	anti-coincidence	AC
gamma rays	GR	coincidence counting with efficiency tracing	CT
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 radiations	MX	digital coincidence counting	DC

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(Ge(HP))-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