BIPM comparison BIPM.RI(II)-K1.Fe-59 of activity measurements of the radionuclide ^{59}Fe for the PTB (Germany), LNE-LNHB (France) and the NMIJ (Japan), and the linked APMP.RI(II)-K2.Fe-59 comparison

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Abstract

Three new participations in the BIPM.RI(II)-K1.Fe-59 comparison have been added to the previous results and this has produced a revised value for the key comparison reference value (KCRV), calculated using the power-moderated weighted mean. A link has been made to the APMP.RI(II)-K2.Fe-59 comparison held in 2014 through the NMIJ who participated in both comparisons. Five NMIs used the K1 or K2 comparison to update their degree of equivalence. 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 remaining five NMIs in the BIPM.RI(II)-K1.Fe-59 comparison and the eight other participants in the APMP.RI(II)-K2.Fe-59 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 radioactive solution. For radioactive gases, a different standard ampoule is used. Each NMI completes a 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, A_e, are all given in [1].

From its inception until 31 December 2018, the SIR has measured 1008 ampoules to give 763 independent results for 70 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.Fe-59 key comparison and includes results published previously [3].

In addition, an international comparison was held in 2014 for this radionuclide, APMP.RI(II)-K2.Fe-59 [4]. Seven laboratories took part in this comparison including the NMIJ who participated in the SIR at the same time, enabling to link the APMP.RI(II)-K2 comparison to the BIPM.RI(II)-K1 comparison. The BARC and the KRISS had previously submitted ampoules to the SIR and have updated their results through this APMP.RI(II) comparison.

2. Participants

The PTB, LNE-LNHB and the NMIJ have submitted ampoules for inclusion in this comparison, which replace their earlier SIR submissions. The details of the laboratories that have participated in the BIPM.RI(II)-K1 comparison are given in Table 1a, with the earlier submissions being taken from [3]. In cases where the laboratory has changed its name since the original submission, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB.

The date of measurement in the SIR is also given in Table 1a and is used in the KCDB and all references in this report.

Table 1a. Details of the participants in the BIPM.RI(II)-K1.Fe-59

NMI	Original acronym	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
BKFH	OMH	Országos Mérésügyi Hivatal	Hungary	EURAMET	1976-05-21 1983-07-08 2001-12-20
–	IAEA /RCC†	International Atomic Energy Agency	–	–	1978-03-31
PTB	–	Physikalisch-Technische Bundesanstalt	Germany	EURAMET	1979-03-01 1989-04-12 1995-05-09 2012-04-03

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Table 1a continued. Details of the participants in the BIPM.RI(II)-K1.Fe-59

NMI	Original acronym	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
NIST	NBS	National Institute of Standards and Technology	United States	SIM	1979-04-04 1987-03-18 2001-11-22
NPL	–	National Physical Laboratory	United Kingdom	EURAMET	1979-09-04
ANSTO	AAEC	Australian Nuclear Science and Technology Organisation	Australia	APMP	1980-04-04
CMI-IIR	UVVVR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EURAMET	1984-09-10
LNE-LNHB	LMRI	Laboratoire national de métrologie et d'essais - Laboratoire national Henri Becquerel	France	EURAMET	1989-01-24 2013-09-30
NMIJ	ETL	National Metrology Institute of Japan	Japan	APMP	1997-11-19 2014-07-02
BARC	–	Bhabha Atomic Research Centre	India	APMP	1998-10-13
KRISS	–	Korea Research Institute of Standards and Science	Republic of Korea	APMP	1999-01-05

† the Radiochemical Centre, Amersham, UK

The eight additional NMIs that took part in the APMP.RI(II) international comparison, APMP.RI(II)-K2.Fe-59 in 2014 and are also eligible for the KCDB are shown in Table 1b.

Table 1b. Details of the participants in the 2014 APMP.RI(II)-K2.Fe-59 to be linked to BIPM.RI(II)-K1.Fe-59 comparison

NMI	Full name	Country	Regional metrology organization
BARC	Bhabha Atomic Research Centre	India	APMP
INER	Institute of Nuclear Energy Research	Chinese Taipei	APMP
KRISS	Korea Research Institute of Standards and Science	Korea	APMP
LNMRI/IRD	Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI)/Instituto de Radioproteção e Dosimetria - IRD	Brazil	SIM
NIM	National Institute of Metrology	China	APMP
NMISA	National Metrology Institute of South Africa	South Africa	AFRIMETS
OAP	Office of Atoms for Peace	Thailand	APMP
PTKMR-BATAN	Pusat Teknologi Keselamatan Dan Metrologi Radiasi	Indonesia	APMP

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, the activities submitted, the relative standard uncertainties ($k = 1$) and the half-life used by the participants in the SIR are given in Table 2. The uncertainty budgets for the three new submissions are given in Appendix 1, previous uncertainty budgets are given in the earlier K1 report [3]. The uncertainty budgets for all the participants in the APMP.RI(II)-K2.Fe-59 comparison were published in the final report [4]. The acronyms used for the measurement methods are given in Appendix 2.

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 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. However, no such impurity measurement has needed to be carried out at the BIPM for the three latest submissions.

Table 2. Standardization methods of the SIR participants for ^{59}Fe

NMI	Method used and acronym (see Appendix 2)	Half-life / d	Activity A_i / kBq	Reference date YYYY-MM-DD	Relative standard uncertainty / 10^{-2} by method of evaluation	
					A	B
BKFH	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	44.5(2) [3]	4 412* 4 409	1976-05-15 12 h UT	0.14	0.57
		44.52(2) [9]	3 759	1983-07-15 12 h UT	0.02	0.19
	4πβ(PC)-γ coincidence and anti-coincidence 4P-PC-BP-NA-GR-CO 4P-PC-BP-NA-GR-AC	44.53(3) [10]	4 737	2001-12-15 0 h UT	0.04	0.21
IAEA /RCC[†]	4πβ-γ coincidence 4P-00-BP-00-GR-CO	44.6	1 026* 1 526	1978-02-22 12 h UT	0.07	0.13
PTB	4πβ-γ coincidence 4P-00-BP-00-GR-CO	–	6 050	1979-03-01 0 h UT	0.05	0.20
	Pressurized ionization chamber [a] 4P-IC-GR-00-00-00	–	716.64	1989-04-15 0 h UT	0.05	0.21
	Pressurized ionization chamber [b] 4P-IC-GR-00-00-00	–	6 848	1995-04-01 0 h UT	0.05	0.21
	CIEMAT/NIST and TDCR 4P-LS-MX-00-00-CN 4P-LS-MX-00-00-TD	44.53(3)	7 367.4[d]	2012-03-15 0 h UT	0.04[d]	0.16[d]
NIST	Pressurized ionization chamber [c] 4P-IC-GR-00-00-00	44.51	2 805	1979-01-25 15 h UT	0.02	0.52
	Pressurized ionization chamber [c] 4P-IC-GR-00-00-00		949.7	1987-03-03 17 h UT	0.11	0.30
	Pressurized ionization chamber [c] 4P-IC-GR-00-00-00	44.51(2)	287.4	2001-11-15 12 h UT	0.15	0.34
NPL	Pressurized ionization chamber [b] 4P-IC-GR-00-00-00	44.6	1 550* 1 468	1979-09-01 0 h UT	0.15	0.34
ANSTO	4πβ-γ coincidence 4P-00-BP-00-GR-CO	–	4 024	1980-03-01 0 h UT	0.09	0.36

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Table 2 continued. Standardization methods of the SIR participants for ^{59}Fe

NMI	Method used and acronym (see Appendix 2)	Half-life / d	Activity A_i / kBq	Reference date	Relative standard uncertainty / 10^{-2} by method of evaluation	
LNE-LNHB	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	44.51(2)	231.92* 232.29	1988-12-19 12 h UT	0.06	0.03
	4πβ-γ anti-coinc. 4P-PC-BP-NA-GR-AC and 4P-LS-BP-NA-GR-AC	44.495(8) [8]	971.8 969.7	2013-06-24 12 h UT	0.23 0.16	0.10 0.10
NMIJ	4πβ(PC)-γ(NaI(Tl)) coincidence 4P-PC-BP-NA-GR-CO	–	6 635	1997-11-20 0 h UT	0.14	0.25
		44.495(8) [8]	1 692.6	2014-06-01 0 h UT	0.04	0.14
BARC	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	–	1 276.3	1998-08-13 6 h 30 UT	0.06	0.15
KRISS	4πβ(PPC)-γ coincidence 4P-PP-BP-NA-GR-CO	44.51(2)	3 238	1998-09-01 0 h UT	0.21	0.25

* Two ampoules were submitted
[a] Calibrated using a ^{59}Fe solution standardized by 4π(PC)-γ and 4π(PPC)-γ coincidence
[b] Calibrated by a primary standardization of a ^{59}Fe solution
[c] Calibrated using a ^{59}Fe solution standardized by 4πβ(PC)-γ in 1970
[d] Weighted mean of CIEMAT/NIST and TDCR result [15]; the relative uncertainty of TDCR is adopted for the final result as it is larger than the internal and external relative uncertainties of the weighted mean.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". The recent submissions have added three ampoules for the activity measurements for ^{59}Fe giving rise to 25 ampoules in total. The SIR equivalent activity, A_{ei}, for each ampoule for the previous and new results is given in Table 4a for each NMI, *i*. The relative standard uncertainty arising from the measurements in the SIR is also shown. This uncertainty is additional to that declared by the NMI for the activity measurement shown in Table 2. Although submitted activities are compared with a given source of ^{226}Ra, all the SIR results are normalized to the radium source number 5 [1].

The half-life used by the BIPM has been updated to 44.494(12) d from the DDEP evaluation dating 2014 and all the results in Table 4a have been updated accordingly. The half-life used in the APMP.RI(II)-K2.Fe-59 comparison is 44.495(8) d from the BIPM Monographie 5 [8].

No recent submission has been identified as a pilot study so the result of each NMI is normally eligible for the key comparison database (KCDB) of the CIPM MRA.

An international comparison for this radionuclide, APMP.RI(II)-K2.Fe-59 was held in 2014 [4] and the eight laboratories from this comparison to be added to the matrix of degrees of equivalence are given in Table 1b. The BARC and the KRISS used the APMP comparison to update their degree of equivalence from earlier SIR participation.

The results $(A/m)_i$ of the APMP comparison have been linked to the BIPM.RI(II)-K1.Fe-59 comparison through the measurement in the SIR of one ampoule of the APMP solution standardized by the NMIJ. The link is made using a normalization ratio deduced from the row indicated in Table 4a:

$$A_{\mathrm{e},i} = (A/m)_i \times A_{\mathrm{e,NMIJ}}/(A/m)_{\mathrm{NMIJ}} = (A/m)_i \times 31.0194 \qquad \text{(a)}$$

The details of the links are given in Table 4b. The uncertainties for the APMP.RI(II) comparison results linked to the SIR are comprised of the original uncertainties together with the uncertainty in the link, 5×10^{-4}, given by the uncertainty of the SIR measurement of the NMIJ ampoule.

Table 3. Details of the solution of ^{59}Fe submitted

NMI/ SIR year	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity[a]
BKFH 1976	Fe in HCl	0.1	Fe : 100	–	^{60}Co : 0.012 (3) %
1983			Fe : 140	–	-
2001	$FeCl_3$ in HCl	0.5	$FeCl_3$: 25	–	^{60}Co : 0.020 (2) %
IAEA /RCC 1978	–	–	Fe : 100	–	^{55}Fe : < 0.2 %
PTB 1979	$FeCl_3$ in HCl	0.1	$FeCl_3$: 100	1.000	< 0.01 %
1989			$FeCl_3$: 60	1.00	–
1995					–
2012	$FeCl_3.6H_2O$ in HCl	0.1	$FeCl_3.6H_2O$: 97	1.000	–
NIST 1979	Fe in HCl	1	Fe: 9	1.015 (2)	^{55}Fe : 1×10^{-3} %
1987	$FeCl_3$ in HCl	1.0	$FeCl_3$: 50	1.016	–
2001		0.1	$FeCl_3$: 3500	1.002 (1)	–
NPL 1979	$FeCl_2$ in HCl	0.1	$FeCl_2$: 60	1.001	–
ANSTO 1980	iron chloride in HCl	0.1	–	1.00	< 0.1 %
CMI-IIR 1984	$FeCl_3$ in HCl	0.08	$FeCl_3$: 50	–	< 0.1 %
LNE-LNHB 1989	$FeCl_3$ in HCl	0.1	$FeCl_3$: 5	0.998	–
2013	$FeCl_3.6H_2O$ in HCl	0.1	Fe: 10	1.000	–
NMIJ 1997	$FeCl_3$ in HCl	0.1	$FeCl_3$: 100	1.00	–
2014[b]					–
BARC 1998	$FeCl_3$ in HCl	0.1	$FeCl_3$: 100	1	–
KRISS 1999	$FeCl_3$ in HCl	0.5	$FeCl_3$: 450	1.0073	55Fe : 0.060 (2) %

[a] The ratio of the activity of the impurity to the activity of ^{59}Fe at the reference date.
[b] The same solution as used in the APMP.RI(II)-K2.Fe-59 comparison.

Table 4a. Results of SIR measurements of ^{59}Fe

NMI/ SIR year	Mass of solution /g	Activity submitted A_i / kBq	No. of Ra source used	SIR $A_{e,i}$ / kBq	Relative standard uncertainty from SIR /10^{-4}	Combined uncertainty $u(A_{e,i})$ / kBq
BKFH 1976	3.604 3 3.602 5	4 412 4 409	4	14 579 14 584	4×10^{-4}	86 86
1983	3.604 0	3 759	4	14 639	4×10^{-4}	29
2001	3.641 1	4 737	4	14 685	4×10^{-4}	32
IAEA /RCC 1978	3.627 23[a] 3.609 28	1 026 1 526	2	14 661 14 663	9×10^{-4} 7×10^{-4}	25 24
PTB 1979	3.640 0(1)	6 050	4	14 691	4×10^{-4}	31
1989	3.673 2	716.64	3	14 729	7×10^{-4}	33
1995	3.523 00	6 848	4	14 710	5×10^{-4}	32
2012	3.6282	7 367.4	4	14 609	4×10^{-4}	25
NIST 1979	3.649 45	2 805	3	14 779	7×10^{-4}	78
1987	3.608 4	949.7	3	14 775	7×10^{-4}	49
2001	3.691 2(2)	287.4	1	14 641	17×10^{-4}	60
NPL 1979	3.538 6 3.353 1	1 550 1 468	3	14 688[b] 14 668	5×10^{-4} 6×10^{-4}	55[b] 55
ANSTO 1980	3.581 43	4 024	4	14 548	5×10^{-4}	54
CMI-IIR 1984	3.574 71	19 632	5	14 709	4×10^{-4}	36
LNE-LNHB 1989	3.647 61 3.653 43	231.92 232.29	1	14 603 14 606	16×10^{-4} 14×10^{-4}	26 23
2013	3.6385	970.8[c]	1	14 603	15×10^{-4}	36
NMIJ 1997	3.610 08	6 635	4	14 683	4×10^{-4}	42
2014[‡]	3.602	1 692.6	3	14 576	5×10^{-4}	23
BARC 1998	3.674 9	1 276.3	2	14 511	10×10^{-4}	28

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Table 4a continued. Results of SIR measurements of ^{59}Fe

NMI/ SIR year	Mass of solution /g	Activity submitted A_i / kBq	No. of Ra source used	SIR $A_{e,i}$ / kBq	Relative standard uncertainty from SIR /10^{-4}	Combined uncertainty $u(A_{e,i})$ / kBq
KRISS 1999	3.612 07	3 238	2	14 728	11×10^{-4}	50

‡ Result used to link the APMP.RI(II)-K2 comparison
[a] Mass of solution before dilution 2.426 19 g
[b] The mean of the two A_e values is used with an averaged uncertainty, as attributed to an individual entry [11].
[c] Mean value of the two results obtained using two different methods, as given by the LNE-LNHB (266.8 kBq g^{-1} with a relative uncertainty of 0.2 %)

Table 4b. Results of the 2014 APMP.RI(II) comparison of ^{59}Fe and links to the SIR (see [4] for more detail)

NMI	Measurement method and acronym (see Appendix 2)	Activity* concentration measured $(A/m)_i$ / (kBq g^{-1})	Standard uncertainty u_i / (kBq·g^{-1})	SIR equivalent activity A_{ei} / kBq	Combined standard uncertainty u_i / kBq
BARC	Arithmetic mean of the results obtained by 4P-PC-BP-NA-GR-CO and 4P-LS-BP-NA-GR-CO	472.8	2.1	14 667	66
INER	4P-PC-BP-NA-GR-CO	471.8	1.5	14 635	46
KRISS	4P-LS-BP-NA-GR-CO	471.0	3.0	14 610	92
LNMRI/IRD	Weighted mean of the results obtained by 4P-PC-BP-GH-GR-CO 4P-LS-BP-NA-GR-CO 4P-PC-BP-NA-GR-CO (Gamma window: 1099 keV) and 4P-PC-BP-NA-GR-CO (Gamma window: 1291 keV)	499.8	3.5	15 500	110
NIM	4P-PC-BP-NA-GR-CO	471.7	1.6	14 632	50
NMISA	4P-LS-BP-NA-GR-CO	473.6	1.4	14 691	43
OAP	4P-IC-GR-00-00-00	482	12	14 950	370
PTKMR-BATAN	4P-LS-BP-NA-GR-CO	458.7	3.7	14 230	120

*Referenced to 1 June 2014, 0 h UTC

4.1 The Key Comparison Reference Value

In May 2013 the CCRI(II) decided to calculate the key comparison reference value (KCRV) using the power-moderated weighted mean [12] rather than an unweighted mean, as had been the policy. This type of weighted mean is similar to a Mandel-Paule mean in that the NMIs' uncertainties may be increased until the reduced chi-squared value is one. In addition, it allows for a power smaller than two in the weighting factor. Therefore, all SIR key comparison results can be selected for the KCRV with the following provisions:

a) only results for solutions standardized by primary techniques are accepted, with the exception of radioactive gas standards (for which results from transfer instrument measurements that are directly traceable to a primary measurement in the laboratory may be included);
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) results more than 20 years old are included in the calculation of the KCRV (but are not included in data shown in the KCDB or in the plots in this report as they have expired);
d) possible outliers can be identified on a mathematical basis and excluded from the KCRV using the normalized error test with a test value of 2.5 and using the modified uncertainties;
e) results can also be excluded for technical reasons; and
f) the CCRI(II) is always the final arbiter regarding excluding any data from the calculation of the KCRV.

The data set used for the evaluation of the KCRVs is known as the "KCRV file" and is a 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 made only by the CCRI(II) during one of its biennial meetings as for the case of ^{59}Fe in June 2017, or by consensus through electronic means (i.e., email) as discussed at the CCRI(II) meeting in 2013.

Consequently, the KCRV for ^{59}Fe has been calculated to be 14 639(27) kBq on the basis of the SIR results from the BKFH (2001), PTB (2012), NIST (1987), NPL, ANSTO, CMI-IIR, LNE-LNHB (2013), NMIJ (2014), BARC and the KRISS. The IAEA result has not been used as that is traceable to another laboratory. Although the same primary standardization was used to calibrate the ionization chamber used for NIST samples of 1979 and 1987, the latter sample has smaller uncertainties as it was more thoroughly characterized and so the 1987 value has been used for the KCRV.

The updated KCRV can be compared with the previous KCRV value of 14 662(26) kBq published in 2003 [3] and the value of 14 643(49) kBq obtained using the SIRIC efficiency curve of the SIR [13].

4.2 Degrees of equivalence

Every participant in a comparison is entitled to have one result included in the KCDB as long as the NMI is a signatory or designated institute listed in the CIPM MRA, and the result is valid (i.e., not older than 20 years). Normally, the most recent result is the one included. An NMI may withdraw its result only if all other participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the KCRV [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 result with the KCRV*

The degree of equivalence of the result of a particular NMI, *i*, with the key comparison reference value is expressed as the difference D_i between the values

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

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

$$U_i = 2u(D_i)\,. \qquad (2)$$

When the result of the NMI *i* is included in the KCRV with a weight w_i, then

$$u^2(D_i) = (1\text{-}2w_i)\,u_i^2 + u^2(\mathrm{KCRV})\,. \qquad (3)$$

However, when the result of the NMI *i* is not included in the KCRV, then

$$u^2(D_i) = u_i^2 + u^2(\mathrm{KCRV}). \qquad (4)$$

4.2.2 *Comparison between pairs of NMI results*

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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \qquad (5)$$

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

$$u_{Dij}^{\ 2} = u_i^{\ 2} + u_j^{\ 2} - 2u(A_{ei}, A_{ej}) \qquad (6)$$

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 [14] 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 matrix of all the degrees of equivalence as they will 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. This 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.

Conclusion

The BIPM ongoing key comparison for ^{59}Fe, BIPM.RI(II)-K1.Fe-59 currently comprises five valid results. The key comparison reference value has been re-evaluated using the power-moderated weighted mean to include the PTB, LNE-LNHB and the NMIJ latest results. The SIR results have been analysed with respect to the updated KCRV determined for this radionuclide, providing degrees of equivalence for these five participants.

The results of eight other NMIs that took part in the APMP.RI(II)-K2.Fe-59 comparison in 2014 have been linked to the BIPM ongoing key comparison through one ampoule of the comparison measured in the SIR. These results superseded two earlier results from the BIPM.RI(II)-K1.Fe-59 comparison. The linked results are included in the matrix of degrees of equivalence and shown on the graph. This has enabled the table of degrees of equivalence to include eight additional results.

The degrees of equivalence have been approved by the CCRI(II) and are published in the BIPM key comparison database. Further results may be added when other NMIs contribute ^{59}Fe activity measurements to the ongoing K1 comparison or take part in other linked comparisons.

References

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Table 5. Table of degrees of equivalence and introductory text for ^{59}Fe

Key comparison BIPM.RI(II)-K1.Fe-59

MEASURAND : Equivalent activity of ^{59}Fe

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 14 639 kBq with a standard uncertainty, u_R = 27 kBq (see Section 4.1 of the Final Report). The value x_i is 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 $U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ 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: $D_{ii} = D_i - D_i = (x_i - x_i)$ and U_{ii}, its expanded uncertainty (k = 2), both expressed in MBq. The approximation $U_{ij} \sim 2(u_i^2 + u_j^2)^{1/2}$ may be used in the following table.

Linking APMP.RI(II)-K2.Fe-59 (2014) to BIPM.RI(II)-K1.Fe-59

The value x_i is the equivalent activity for laboratory *i* participant in APMP.RI(II)-K2.Fe-59 having been normalized using the value of the linking laboratories, NMIJ (see Final report).

The degree of equivalence of laboratory *i* participant in APMP.RI(II)-K2.Fe-59 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.

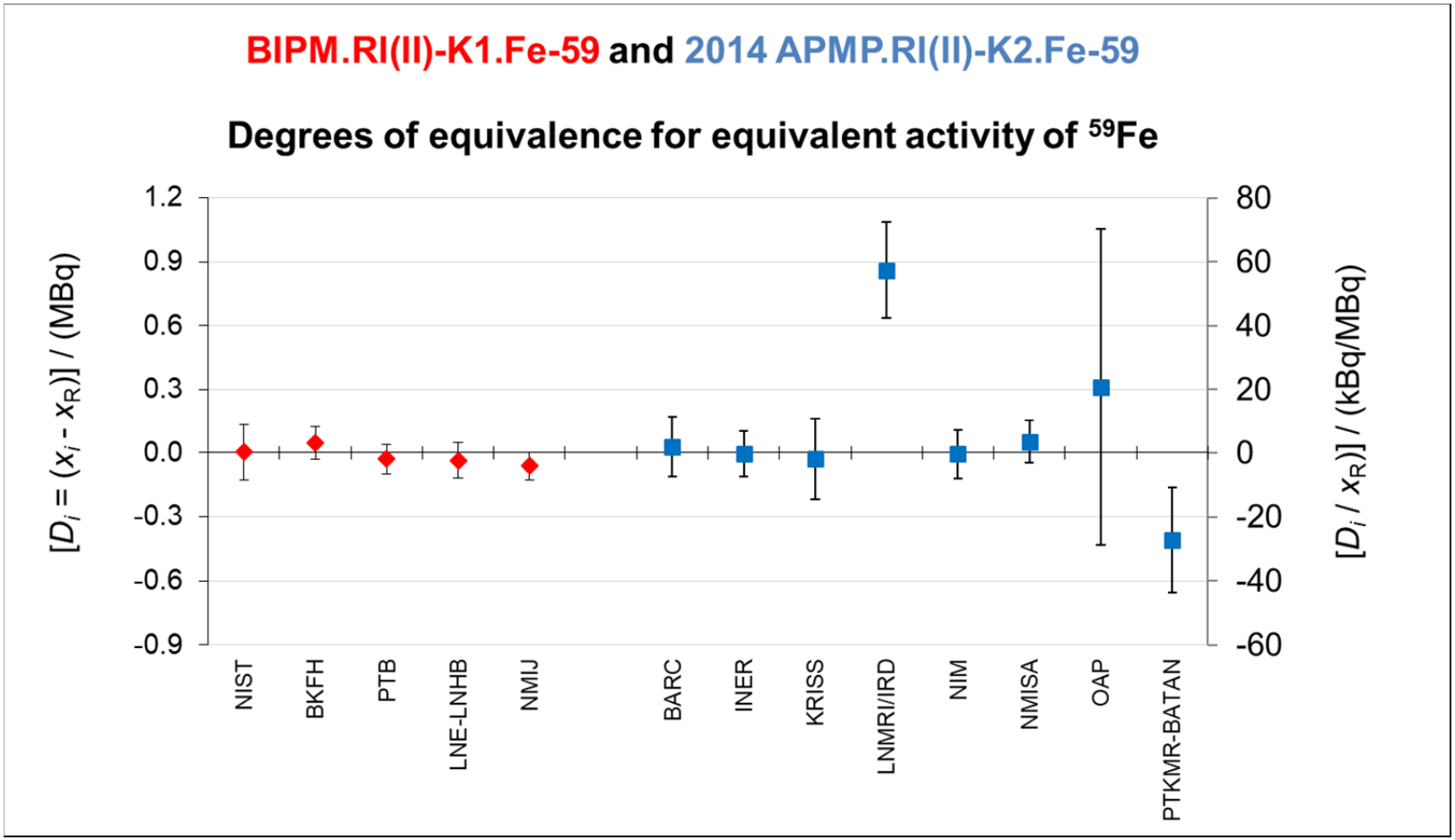
When required, the degree of equivalence between two laboratories *i* and *j*, participants in one or another of the two key comparisons, is given by a pair of terms: $D_{ij} = D_i - D_j$ and U_{ij}, its expanded uncertainty ($k = 2$), both expressed in kBq. Correlations between pairs of laboratories should be taken into account as explained in Section 4.2.2 of the Final report.

These statements make it possible to extend the BIPM.RI(II)-K1.Fe-59 matrices of equivalence to the other participants in the APMP.RI(II)-K2.Fe-59.

Lab *i*

	D_i / MBq	U_i
NIST	**0.00**	0.13
BKFH	**0.046**	0.078
PTB	**-0.030**	0.069
LNE-LNHB	**-0.036**	0.084
NMIJ	**-0.063**	0.067
BARC	**0.03**	0.14
INER	**0.00**	0.11
KRISS	**-0.03**	0.19
LNMRI/IRD	**0.86**	0.23
NIM	**-0.01**	0.11
NMISA	**0.05**	0.10
OAP	**0.31**	0.74
PTKMR-BATAN	**-0.41**	0.25

Figure 1. Graph of degrees of equivalence with the KCRV for ^{59}Fe
(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 ^{59}Fe submitted to the SIR

The PTB has submitted in 2012 detailed uncertainty as follows:

TDCR method

Relative standard uncertainties	**Comments**	$u_{rel,i} \times 10^4$ evaluated by method	
Contributions due to		**A**	**B**
counting statistics	standard deviation of the mean of 10 samples	3	–
weighing		–	6
dilution		–	5
dead time		–	3
background		3	–
counting time		–	1
TDCR value		–	1
input parameters and statistical model	including decay scheme parameters	–	5
ionization quenching and *kB* (model)		–	9
interpolation from calibration curve	included in TDCR value uncertainty		
PMT asymmetry		–	2
decay correction		–	< 5
adsorption		–	5
radionuclide impurities	no impurities detected	–	3
Quadratic summation		**4**	**16**
Relative combined standard uncertainty, u_c		**16**	

CIEMAT/NIST method

Relative standard uncertainties	**Comments**	$u_{rel,i} \times 10^4$ evaluated by method	
Contributions due to		**A**	**B**
counting statistics	standard deviation of the mean of 10 samples	2	–
weighing		–	6
dilution		–	5
dead time		–	10
background		5	–
counting time		–	1
tracer (^{3}H)		–	5
input parameters and statistical model	including decay scheme parameters	–	5
ionization quenching and *kB* (model)		–	7
quenching indicator (SQP(E), tSIE)		–	3
interpolation from calibration curve	included in tracer uncertainty		
decay-scheme parameters	included in "input parameters"		
decay correction		–	< 5
adsorption		–	5
radionuclide impurities	no impurities detected	–	3
Quadratic summation		**4**	**16**
Relative combined standard uncertainty, u_c		**16**	

The LNE-LNHB has submitted in 2013 detailed uncertainty budgets as follows:

4P-PC-BP-NA-GR-AC

Relative standard uncertainties / **Contributions due to**	$u_{rel,i} \times 10^4$	**Method of evaluation**
counting statistics (including extrapolation)	23	A
weighing	10	B
live-time technique	1	B
background	5	A
decay correction	5	B
Relative combined standard uncertainty, u_c	**26**	

4P-LS-BP-NA-GR-AC

Relative standard uncertainties / **Contributions due to**	$u_{rel,i} \times 10^4$	**Method of evaluation**
counting statistics	5	A
weighing	10	B
live-time technique	1	B
background	10	A
decay correction	5	B
extrapolation technique (PNT defocusing/ quenching agent)	12	A
Relative combined standard uncertainty, u_c	**20**	

The NMIJ has submitted in 2014 a detailed uncertainty budget as follows:

Relative standard uncertainties	$u_{rel,i} \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	4	–
weighing	–	8
dead time	–	< 1
background	–	1
counting time	–	2
resolving time	–	3
Gandy effect	–	2
decay correction	–	1
extrapolation of efficiency curve	–	10
Quadratic summation	**4**	**14**
Relative combined standard uncertainty, u_c	**15**	

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
		Cerenkov detector	CD
		calorimeter	CA
		solid plastic scintillator	SP
		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
alpha - particle	AP	selective sampling	SS
mixture of various radiation	MX	high efficiency	HE

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