Activity measurements of the radionuclides ^{18}F, ^{64}Cu, ^{99m}Tc and ^{11}C for the NRC, Canada, in the ongoing comparisons BIPM.RI(II)-K4 series and KCRV update in the corresponding BIPM.RI(II)-K1 comparison

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Abstract

In 2017, comparisons of activity measurements of ^{18}F, ^{64}Cu, ^{99m}Tc and ^{11}C using the Transfer Instrument of the International Reference System (SIRTI) took place at the National Research Council of Canada (NRC, Canada). Ampoules containing ^{18}F, ^{64}Cu, ^{99m}Tc and ^{11}C solutions were measured in the SIRTI for about 3 half-lives (10 half-lives for ^{11}C). The NRC standardized the activity in the ampoules by 4π(PC)β–γ anti-coincidence measurements (CIEMAT/NIST liquid scintillation counting for ^{11}C). The comparisons, identifiers BIPM.RI(II)-K4.F-18, BIPM.RI(II)-K4.Cu-64 and BIPM.RI(II)-K4.Tc-99m are linked to the corresponding BIPM.RI(II)-K1.F-18, BIPM.RI(II)-K1.Cu-64 and BIPM.RI(II)-K1.Tc-99m comparisons. The BIPM.RI(II)-K1 ^{18}F and ^{99m}Tc key comparison reference values have been updated to include the latest BIPM.RI(II)-K4 linked results and degrees of equivalence for those three comparisons have been evaluated.

1. Introduction

Radionuclides with half-life often much less than a day are essential for nuclear medicine and particularly for medical imaging. The use of nuclear medicine is increasing with the accessibility of these radionuclides which are consequently of great interest to the National Metrology Institutes (NMIs) in terms of standardization and SI traceability. However, sending ampoules of short-lived radioactive material to the Bureau International des Poids et Mesures (BIPM) for measurement in the International Reference System (SIR) [1] is only practicable for the NMIs that are based in Europe and near to the BIPM. Consequently, to extend the utility of the SIR and enable other NMIs to participate, a transfer instrument (SIRTI) has been developed at the BIPM with the support of the Consultative Committee for Ionizing Radiation CCRI(II) Transfer Instrument Working Group [2].

The BIPM ongoing K4 comparisons of activity measurements of ^{18}F (half-life $T_{1/2}$ = 1.8288(3) h [3])[1], ^{64}Cu ($T_{1/2}$ = 12.7004(20) h [4]), ^{99m}Tc ($T_{1/2}$ = 6.0067(10) h [3]), and of ^{11}C ($T_{1/2}$ =

[1] Hereafter, the last digits of the standard uncertainties are given in parenthesis.

20.361(23) min [3]) are based on the SIRTI, a well-type NaI(Tl) crystal calibrated against the SIR, which is moved to each participating laboratory. The stability of the system is monitored using a ^{94}Nb reference source (half-life of 20 300(1 600) years [5]) from the Joint Research Centre of the European Commission (JRC, Geel), which also contains the ^{93m}Nb isotope. The ^{18}F, ^{64}Cu, ^{99m}Tc or ^{11}C count rate above a low-energy threshold, defined by the ^{93m}Nb x-ray peak at 16.6 keV, is measured relative to the ^{94}Nb count rate above the same threshold. Once the threshold is set, a brass liner is placed in the well to suppress the ^{93m}Nb contribution to the ^{94}Nb stability measurements. It should be noted that the uncertainty associated with the ^{94}Nb decay correction is negligible. The ^{99m}Tc SIR ampoule is placed in the detector well in a brass liner; for the ^{18}F, ^{64}Cu and ^{11}C SIR ampoules, a PVC (polyvinyl chloride) liner is used instead to stop the β^+ particles while minimizing the production of bremsstrahlung. No extrapolation to zero energy is carried out as all the measurements are made with the same threshold setting. The live-time technique using the MTR2 module from the Laboratoire National d’Essais – Laboratoire National Henri Becquerel, France (LNE-LNHB) [6] is used to correct for dead-time losses, taking into account the width of the oscillator pulses. The standard uncertainty associated with the live-time correction, due to the effect of the finite frequency of the oscillator, is negligible.

Similarly, to the SIR, a SIRTI equivalent activity, A_E, is deduced from the ^{18}F, ^{64}Cu, ^{99m}Tc or ^{11}C and the ^{94}Nb counting results and the ^{18}F, ^{64}Cu, ^{99m}Tc or ^{11}C activity measured by the NMI. A_E is inversely proportional to the detection efficiency, i.e.. A_E is the activity of the source measured by the participant divided by the ^{18}F, ^{64}Cu, ^{99m}Tc or ^{11}C count rate in the SIRTI relative to the ^{94}Nb count rate. The possible presence of impurity in the solution should be accounted for using γ-ray spectrometry measurements carried out by the NMI.

The present ^{18}F, ^{64}Cu and ^{99m}Tc K4 comparisons are linked to the corresponding BIPM.RI(II)-K1 comparisons through the calibration of the SIRTI against the SIR at the BIPM and, consequently, the degrees of equivalence with the K1 key comparison reference value (KCRV) can be evaluated. The K4 ^{18}F and ^{99m}Tc comparison results based on primary measurements carried out by the NMI, or ionization chamber measurements traceable to primary ^{18}F and ^{99m}Tc measurements are eligible for inclusion in the KCRV, with some restrictions.

The ^{11}C K4 comparison is not linked to the BIPM.RI(II)-K1.C-11 comparison because of the very short ^{11}C half-life that enables only one NMI to send ampoules to the BIPM for participation in the K1 comparison. The NRC was the first NMI to officially participate in the BIPM.RI(II)-K4.C-11 comparison so no KCRV and no degrees of equivalence could be evaluated. However, the measurement details have already been published [30].

The protocol [7] and previous comparison results for the BIPM.RI(II)-K4 comparisons are available in the key comparison database of the CIPM (International Committee on Weights and Measures) Mutual Recognition Arrangement [8]. Publications concerning the details of the SIRTI and its calibration against the SIR can be found elsewhere [9, 10].

2. Participants

As detailed in the protocol, participation in the BIPM.RI(II)-K4 comparisons mainly concerns member states that are located geographically far from the BIPM (except for ^{11}C) and that have developed a primary measurement method for the radionuclide of concern. However, at the time of the comparison, the NMI may decide for convenience to use a secondary method, for example a calibrated ionization chamber. In this case, the traceability of the calibration needs to be clearly identified.

The present comparisons took place at the National Research Council of Canada (NRC), Ottawa, Canada, in June 2017, who used the 4π(PC)β–γ anti-coincidence measurements for the activity standardizations (CIEMAT/NIST liquid scintillation counting for ^{11}C). NRC was the first NMI to carry out comparisons of four radionuclides during a single two-week period.

3. The SIRTI at the NRC

The reproducibility and stability of the SIRTI at the NRC were checked by measuring the count rate produced by the reference ^{94}Nb source No. 3, the threshold position (defined by the ^{93m}Nb x-ray peak), the background count rate, and the frequency of the oscillator No. 1 for the live-time correction and the room temperature as shown in Figure 1a. The plots shown in the Figure represent the differences from the values indicated in the figure caption, using the appropriate units, as given, for each quantity measured.

The SIRTI background measured at the NRC was relatively high but was stable. The temperature and oscillator frequency were very stable, too. The ^{94}Nb measurement results show a slow increasing trend (~3 x 10^{-4} change in relative count rate in 8 days) that seems anti-correlated with the threshold position. However, as the comparison result is proportional to a ratio of the radionuclide compared and the ^{94}Nb reference source, these slow fluctuations should not impact the comparison results. Nevertheless, this small effect is taken into account in the uncertainty evaluation, except for ^{99m}Tc that is insensitive to this level of threshold fluctuations [9].

The mean ^{94}Nb No. 3 count rate, corrected for live-time, background, and decay, measured at the NRC, was 7631.55(41) s^{-1} and was used to normalize the ^{99m}Tc count rates. This mean value agrees within two standard uncertainties with the weighted mean of the ^{94}Nb No. 3 count rate since the set-up of the system in March 2007, 7632.30(23) s^{-1}. Finally, on the return of the SIRTI to the BIPM after the NRC comparison, the ^{94}Nb count rate was checked giving a value of 7630.4(15) s^{-1} in agreement with the measurements carried out at the NRC.

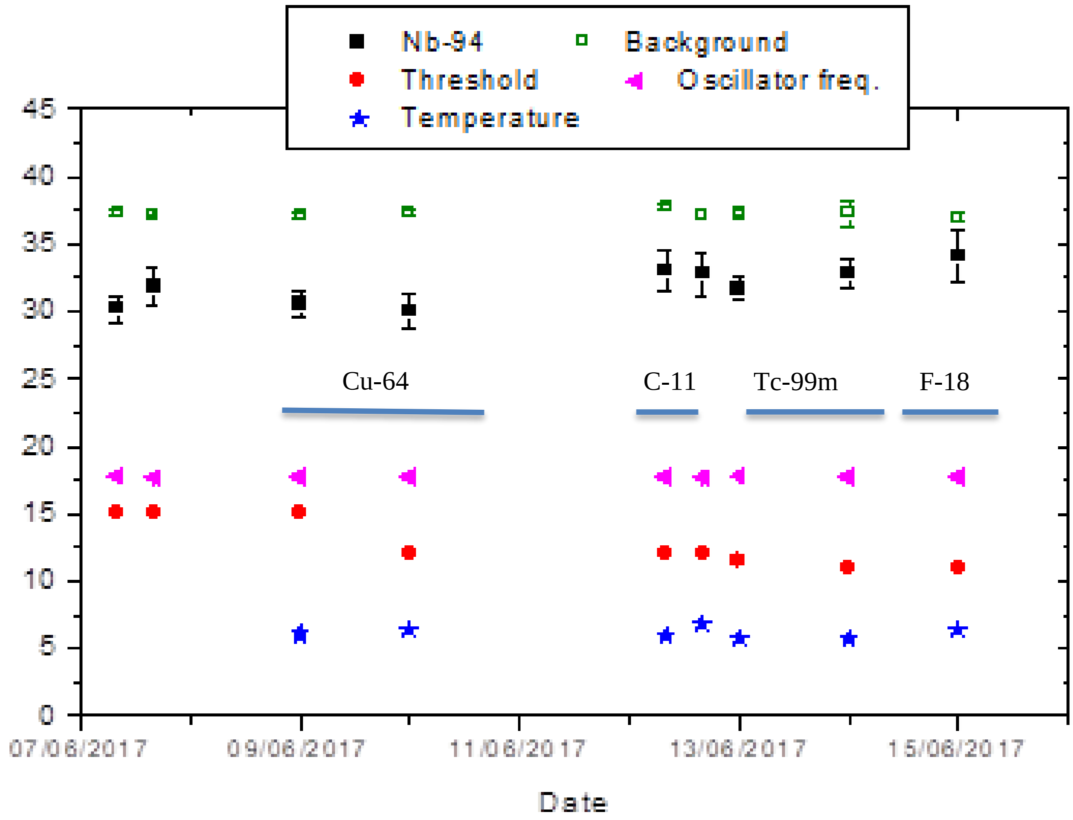


Figure 1: Fluctuation of the SIRTI at the NRC. Filled squares: ^{94}Nb No.3 count rate / s^{-1} above 7600 s^{-1}; triangles: frequency of the oscillator No.1 / Hz above 999 985 Hz; circles: threshold position / channel above 85 channels; asterisks: room temperature / °C above 15 °C; open squares: background count rate / s^{-1} above 110 s^{-1}. Statistical uncertainty (k = 1) for the Nb source, threshold position and background are shown (in some cases, the uncertainties are not visible in the plot as they are hidden by the character printed for the data point).

4. The ^{18}F, ^{64}Cu and ^{99m}Tc solutions standardized at the NRC

For the ^{11}C solution, see [30]. The ^{18}F, ^{64}Cu and ^{99m}Tc solutions measured in the SIRTI are described in Table 1, including any impurities, when present, as identified by the laboratory. Very small drops in the cylindrical part of the ampoules were noted only for ^{64}Cu ampoules and should have a negligible impact on the measurement results. The density and volume of the solutions in the ampoules conformed to the K4 protocol requirements.

The NRC activity measurement results are summarized in Table 2. The uncertainty budgets are given in Table 3.

Table 1: Characteristics of the solutions measured in the SIRTI

Radionuclide	Solvent / mol dm^{-3}	Carrier / µg g^{-1}	Density at 20 °C / g cm^{-3}	Ampoule number	Mass / g	Impurity*
^{18}F	water	NaCl / 1.2	0.9975(27)	17369	3.6088(3)	None detected
				17370	3.6001(1)	
^{64}Cu	water	$CuCl_2$ / 3.4	1.0092(14)	17277	3.6262(1)	^{60}Co: 2.66(3) × 10^{-8}
				17279	3.6090(1)	
^{99m}Tc	water	NaCl / 1.7	1.0044(47)	17340	3.6187(1)	None detected
				17352	3.5964(1)	

* Ratio of the impurity activity to the main radionuclide activity at the reference date

Table 2: The ^{18}F, ^{64}Cu and ^{99m}Tc standardizations by the NRC

Radionuclide	Measurement method ACRONYM*	Activity conc. / kBq g^{-1}	Standard uncert. / kBq g^{-1}	Reference date YYYY-MM-DD	Half-life used by the NMI / h
^{18}F	Ionization Chamber calibrated in January 2017 by 4π(PC)β^{+}-γ anti-coincidence counting 4P-IC-GR-00-00-00	8.85	0.03	2017-06-14 18:30 UTC	1.828 90(23)
^{64}Cu	Ionization Chamber calibrated in March 2017 by 4π(PC)β^{+}-γ anti-coincidence counting 4P-IC-GR-00-00-00	135	2	2017-06-08 16:00 UTC	12.7004(20)
^{99m}Tc	Ionization Chamber calibrated in October 2016 by 4π(PC)β^{+}-γ anti-coincidence counting 4P-IC-GR-00-00-00	13.43	0.16	2017-06-13 16:00 UTC	6.0067(10)

* See appendix 1

Table 3: The NRC uncertainty budgets for the activity standardizations of the ^{99m}Tc, ^{18}F and ^{64}Cu ampoules

Uncertainty contributions due to	Comments	Evaluation method	Relative standard uncertainty × 10^4									
			^{99m}Tc				^{18}F				^{64}Cu	
			4P-PC-MX-NA-GR-AC*	4P-PC-MX-NA-GR-CO	4P-LS-MX-00-00-CN	4P-LS-MX-00-00-TD	4P-PC-PO-NA-GR-AC*	4P-PC-PO-NA-GR-CO	4P-LS-PO-00-00-CN	4P-LS-PO-00-00-TD	4P-PC-PO-NA-GR-AC*	4P-PC-PO-NA-GR-CO
Counting statistics	Standard deviation of the mean of two independent measurements each of four ampoules. Each measurement is determined from five charge measurements	A	2				1				1	
Weighing	Balance Calibration	B	3				3				3	
Half-life	Uncertainty due to decay correction from IC measurement to reference time	B	0.3				1				1	
Dilution	Estimate of uncertainty in dilution factor of single serial dilution	B	1				1				1	
Measurement method	Standard deviation of four determinations of each IC measurement set	A	2				2				1	
Background	Standard deviation of ongoing background measurements	A	1				1				1	
Stability	Standard deviation of ongoing reference source measurements	A	3				3				3	
Charge collection	Electrometer calibration	B	5				5				5	
Impurities	^{60}Co impurity in the ^{64}Cu solution	B	–				–				< 1	
Calibration (see appendix 2)	Uncertainty in determination of the calibration factor from primary measurement.	B	100	86	120	62	33	31	45	44	130	120
Relative combined standard uncertainty**			100	86	120	62	34	32	46	45	130	120

* to be used for equivalence
** all quoted uncertainties are normally distributed.

4.1 ^{18}*F NRC measurement details*

The ^{18}F activity in ampoules 17369 and 17370 were determined from the measurement of a parent solution and a single dilution factor of 183.6(1). Two ampoules containing the parent solutions, 17359 and 17360, were measured in a Vinten ionization chambers (IC). A TPA IC was also used, as confirmatory measurement. These ICs were calibrated in January 2017 at NRC by 4P-PC-PO-NA-GR-AC and a detailed uncertainty budget for the ICs calibration is given in Appendix 2a. Several confirmatory primary methods 4P-PC-PO-NA-GR-CO, 4P-LS-PO-00-00-CN and 4P-LS-PO-00-00-TD were also carried out but are not intended for the calculation of degrees of equivalence. The calibration coefficients for these two ICs determined from four primary methods are presented in Table 4a and demonstrate the measurement compatibility.

Table 4a: ^{18}F calibration factors for two NRC ICs based on primary methods used to determine activity concentration

Primary method	Vinten calibration coefficient / pA MBq^{-1}	Vinten relative standard uncert. / 10^{-2}	TPA calibration coefficient / pA MBq^{-1}	TPA relative standard uncert. / 10^{-2}
4P-PC-PO-NA-GR-AC*	10.38	0.34	41.42	0.34
4P-PC-PO-NA-GR-CO	10.34	0.31	41.28	0.31
4P-LS-PO-00-00-CN	10.30	0.45	41.11	0.45
4P-LS-PO-00-00-TD	10.38	0.44	41.44	0.44

*used for degrees of equivalence

4.2 ^{64}*Cu NRC measurement details*

The ^{64}Cu activity in ampoules 17277 and 17279 were determined from the measurement of a parent solution and a single dilution factor of 22.01(1). Two ampoules containing the parent solutions, 17273 and 17275, were measured in a Vinten ionization chambers (IC). A TPA IC was also used, as confirmatory measurement. These ICs were calibrated in March 2017 at NRC by 4P-PC-PO-NA-GR-AC and a detailed uncertainty budget for the ICs calibration is given in Appendix 2b. A confirmatory primary method 4P-PC-PO-NA-GR-CO was also carried out but is not intended for the calculation of degrees of equivalence. The calibration coefficients for these two ICs determined from two primary methods are presented in Table 4b and demonstrate the measurement compatibility.

Table 4b: ^{64}Cu calibration factors for two NRC ICs based on primary methods used to determine activity concentration

Primary method	Vinten calibration coefficient / pA MBq^{-1}	Vinten relative standard uncert. / 10^{-2}	TPA calibration coefficient / pA MBq^{-1}	TPA relative standard uncert. / 10^{-2}
4P-PC-PO-NA-GR-AC*	1.940	1.29	7.751	1.29
4P-PC-PO-NA-GR-CO	1.893	1.23	7.751	1.23

*used for degrees of equivalence

4.3 *^{99m}Tc NRC measurement details*

The ^{99m}Tc activity in ampoules 17340 and 17353 were determined from the measurement of a parent solution and a single dilution factor of about 722.4(3). Two ampoules containing the parent solutions, 17335 and 17336, were measured in a Vinten ionization chambers (IC). A TPA IC was also used, as confirmatory measurement. These ICs were calibrated in October 2016 at NRC by 4P-PC-MX-NA-GR-AC and a detailed uncertainty budget for the ICs calibration is given in Appendix 2c. Several confirmatory primary methods 4P-PC-MX-NA-GR-CO, 4P-LS-MX-00-00-CN and 4P-LS-MX-00-00-TD were also carried out but are not intended for the calculation of degrees of equivalence. The calibration coefficients for these two ICs determined from four primary methods are presented in Table 4c and demonstrate the measurement compatibility.

Table 4c: ^{99m}Tc calibration factors for two NRC ICs based on primary methods used to determine activity concentration

Primary method	**Vinten calibration coefficient / pA MBq^{-1}**	**Vinten relative standard uncert. / 10^{-2}**	**TPA calibration coefficient / pA MBq^{-1}**	**TPA relative standard uncert. / 10^{-2}**
4P-PC-MX-NA-GR-AC*	1.259	1.0	8.266	1.0
4P-PC-MX-NA-GR-CO	1.250	0.9	8.208	0.9
4P-LS-MX-00-00-CN	1.259	1.2	8.267	1.2
4P-LS-MX-00-00-TD	1.239	0.6	8.132	0.6

*used for degrees of equivalence

5. The ^{18}F, ^{64}Cu and ^{99m}Tc measurements in the SIRTI at the NRC

For the ^{11}C SIRTI measurements, see [30]. The maximum live-time corrected count rate in the NaI(Tl) was 20 000 s^{-1} for ^{99m}Tc and ^{18}F, and 16 000 s^{-1} for ^{64}Cu, which conform to the limit of 20 000 s^{-1} set in the protocol [7]. In addition, a relative standard uncertainty of 3×10^{-4}, 2×10^{-4} and 4×10^{-4} for ^{18}F, ^{64}Cu and ^{99m}Tc respectively, was added to take account of a possible drift in the SIRTI at high count rate [9]. The time of each SIRTI measurement was obtained from the synchronization of the SIRTI laptop with the NRC NTP time server.

In principle, the live-time correction should be modified to take into account the decaying count rate [11]. In the present experiments, the duration of the measurements made at high rate has been limited to 400 s, 600 s and 600 s for ^{18}F, ^{64}Cu and ^{99m}Tc, respectively, so that the relative effect of decay on the live-time correction is less than one part in 10^4.

Two ampoules of each of the ^{18}F, ^{64}Cu and ^{99m}Tc solutions were measured alternatively for about 3 half-lives, respectively, and the results are shown in Figures 2a-2d.

For ^{18}F, the SIRTI preliminary results in Figure 2a show a discrepancy of 0.5 % between the two ampoules produced from the same dilution of the mother solution. Such a difference can hardly be explained except from an issue in the weighing of the solution. In addition, when switching the masses of solution in the two ampoules, the results are then in perfect agreement (see Figure 2b). This effect has also been observed in the IC measurement results of the NRC. So, it was deduced that an error happened in the labelling of the ampoules and the final result of the comparison has been obtained from the data in Figure 2b.

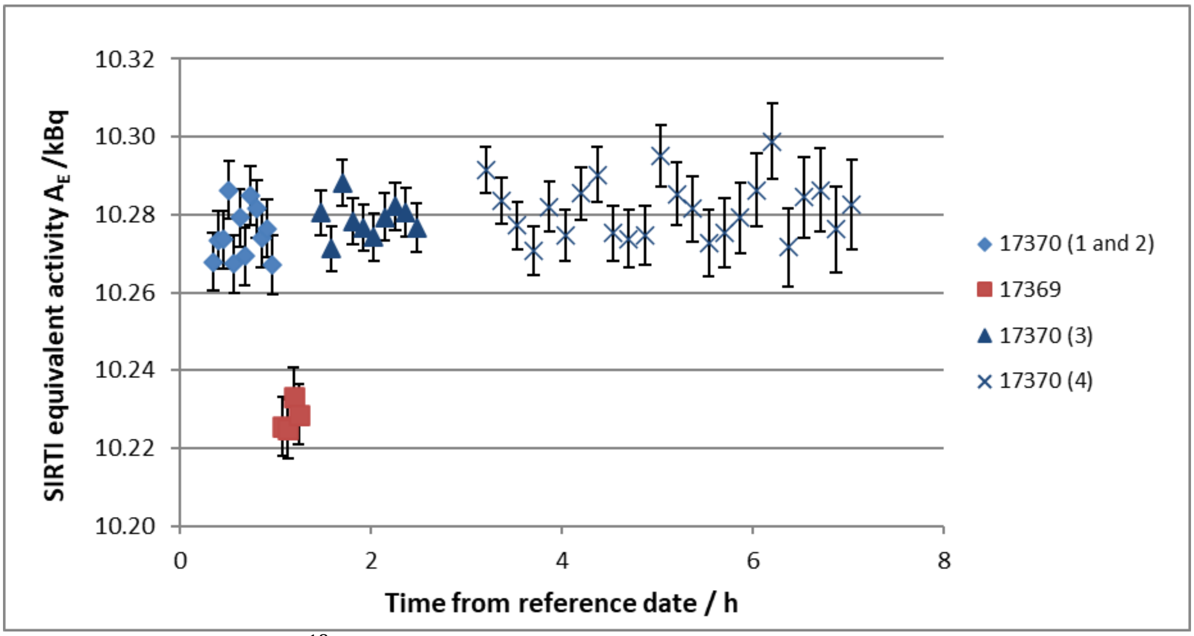


Figure 2a: Preliminary ^{18}F measurement results in the SIRTI at the NRC. The uncertainty of the ^{18}F activity concentration, which is constant over all the measurements, is not included in the uncertainty bars shown on the graph.

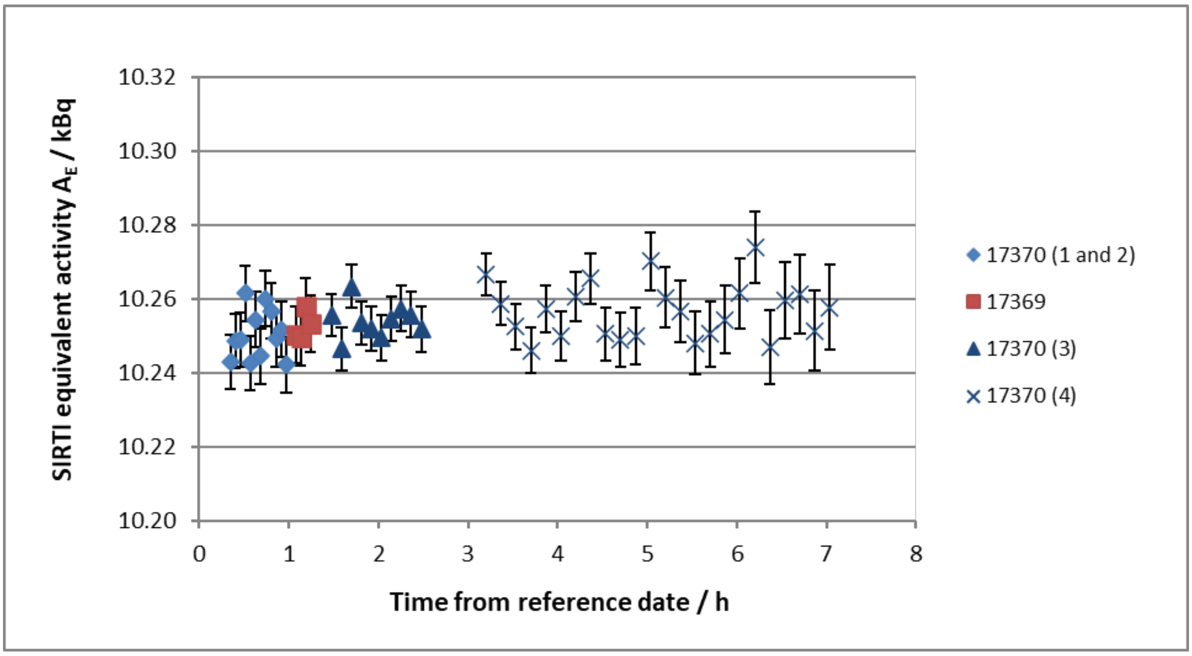


Figure 2b: Final ^{18}F measurement results in the SIRTI at the NRC. The uncertainty of the ^{18}F activity concentration, which is constant over all the measurements, is not included in the uncertainty bars shown on the graph.

The ^{64}Cu ampoules were sealed too high by about 6 mm and the PVC cap of the SIRTI ampoule holder could not be fully closed. Consequently, the ampoules were cut and resealed. It was assumed that the activity in the ampoule remained unchanged during the process. The relative difference in SIRTI result after/before re-sealing, measured with ampoule 17279, is 0.999 68(33) and agrees with the PENELOPE Monte-Carlo result of 0.999 73(40) for a change of position of the PVC cap.

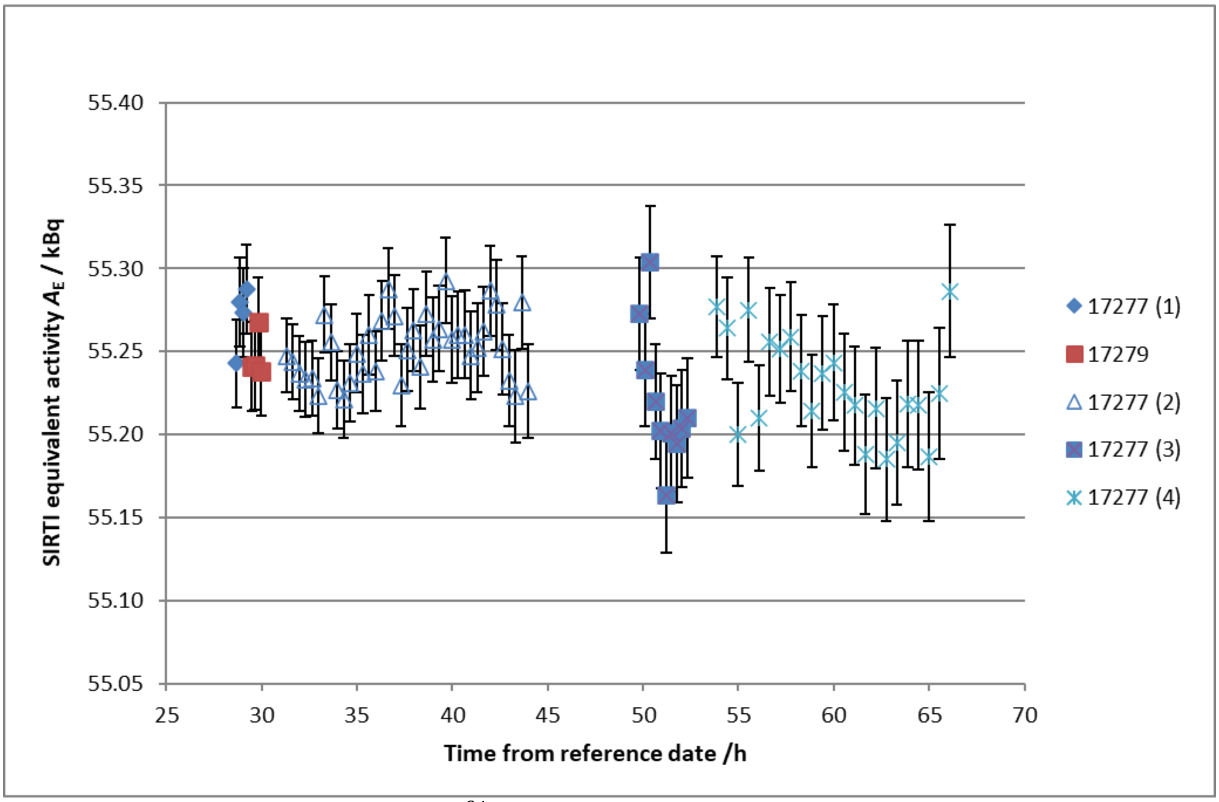


Figure 2c: As for Figure 2b, but for ^{64}Cu

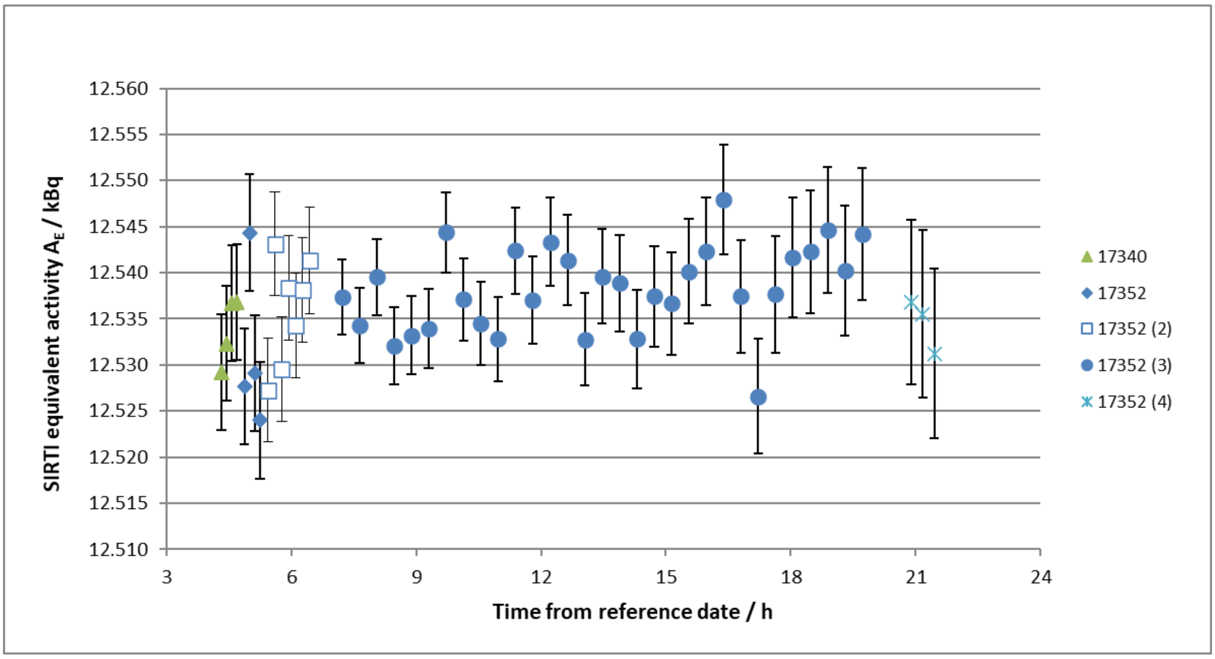


Figure 2d: As for Figure 2b, but for ^{99m}Tc.

The reduced chi-squared values evaluated for these series of measurements are 0.84, 0.87 and 0.89 for ^{18}F, ^{64}Cu and ^{99m}Tc, respectively. The absence of significant trend confirms the stability and adequate live-time corrections of the SIRTI. For ^{18}F and ^{99m}Tc, no impurity was detected by the NRC while for ^{64}Cu, the impurity correction for the SIRTI measurement is negligible ($< 4 \times 10^{-6}$).

The uncertainty budgets for the SIRTI measurements of the ^{18}F, ^{64}Cu and ^{99m}Tc ampoules are given in Table 5. In the SIRTI, the dominant part of the ^{64}Cu detection efficiency is defined by its beta plus decay. In this sense it is similar to ^{18}F, and the Monte-Carlo simulations carried out for ^{18}F were also used for the ^{64}Cu uncertainty evaluation. Further details are given in reference [9].

Table 5: Uncertainty budgets for the SIRTI measurement of the ^{18}F, ^{64}Cu and ^{99m}Tc ampoules

Uncertainty contributions due to	**Comments**	**Evaluation method**	**Relative standard uncertainty × 10^4**		
			^{18}F	**^{64}Cu**	**^{99m}Tc**
Radionuclide to ^{94}Nb measurement ratio	Including live-time, background, decay correction and threshold setting	A	2.2§§	3.6§§§	2.3§
^{94}Nb count rate	K4.Tc-99m: Standard uncertainty of the weighted mean of 9 series of 10 measurements K4.F-18 and Cu-64: Internal uncertainty of 1 series of 10 measurements, added quadratically with the uncertainty of threshold setting	A	1.4	1.6*	0.5
Long-term stability of the SIRTI	Weighted standard deviation of 90 series of 10 measurements	A	0.3	0.3	0.3
Effect of decay on the live-time correction	Maximum measurement duration evaluated from [12]	B	< 1	< 1	< 1
SIRTI drift at high count rate	Mean possible drift over the radionuclide series of measurements at the NRC	B	3	2	4
Ampoule dimensions	From the JRC report [13] and sensitivity coefficients from Monte-Carlo simulations	B	2.2*	2.2*	8.4
Ampoule filling height	Solution volume is 3.6(1) cm^3; sensitivity coefficients from Monte-Carlo simulations	B	2.3	2.3	2.8*
Solution density	Between 1 g/cm^3 and 1.01 g/cm^3 as requested in the protocol; sensitivity coefficients from Monte-Carlo simulations	B	0.7	0.7	0.8
Drops on the ampoule walls	From Monte-Carlo simulation	B	2	2	2
Relative combined standard uncertainty			5.0	5.3	9.7

§ Standard uncertainty of the weighted mean of 49 measurements, taking into account the correlation due to the ^{99m}Tc half-life
§§ Standard uncertainty of the weighted mean of 41 measurements, taking into account the correlation due to the ^{18}F half-life
§§§ Standard uncertainty of the weighted mean of 80 measurements, taking into account the correlation due to the ^{64}Cu half-life
* included in the uncertainty of the radionuclide to ^{94}Nb measurement ratio

6. Comparison results

The weighted mean and uncertainty of a selection of the measured A_E values is calculated taking into account correlations. The standard uncertainty $u(A_E)$ is obtained by adding in quadrature the SIRTI combined uncertainty from Table 4 and the uncertainty stated by the participant for the ^{18}F, ^{64}Cu and ^{99m}Tc measurements (see Table 2). The correlation between the NRC and the BIPM due to the use of the same ^{99m}Tc and ^{64}Cu half-life is negligible in view of the small contribution of these half-lives to the combined uncertainty of the comparison results.

The K4 comparison results are given in Table 6a as well as the linked results A_e in the corresponding BIPM.RI(II)-K1 comparisons which were obtained by multiplying A_E by the linking factors given in Table 6b:

Table 6a: BIPM.RI(II)-K4. comparison results and link to the BIPM.RI(II)-K1 comparisons

Radionuclide	**Measurement method** ACRONYM*	**Solution volume in the ampoules (calculated) /cm^3**	**A_E /kBq**	**$u(A_E)$ /kBq**	**Linked A_e /kBq**	**$u(A_e)$ /kBq**
^{11}C	IC calibrated in Feb. 2017 by CIEMAT/NIST method 4P-IC-GR-00-00-00	3.59(2) and 3.61(2)	9.90**	0.11**	–	–
^{18}F	IC calibrated in January 2017 by 4π(PC)β$^+$-γ anti-coincidence counting 4P-IC-GR-00-00-00	3.61(1) and 3.62(1)	10.255	0.035	15 332	56
^{64}Cu	IC calibrated in March 2017 by 4π(PC)β$^+$-γ anti-coinc. counting 4P-IC-GR-00-00-00	3.593(5) and 3.576(5)	55.25	0.71	81 900	1100
^{99m}Tc	IC calibrated in October 2016 by 4π(PC)-γ anti-coinc. counting 4P-IC-GR-00-00-00	3.603(17) and 3.581(17)	12.54	0.12	152 500	1500

* See appendix 1
** The result A_E = 9.906(99) kBq published in 2019 [30] was coming from the SIRTI measurement of ampoule 17 324 only.

Table 6b: Linking factors of BIPM.RI(II)-K4 comparison to BIPM.RI(II)-K1 comparison

Radionuclide	**Linking factor**	**Provider of solution for measurement of linking factor at the BIPM**
^{99m}Tc	12 165(23)	LNE-LNHB (2007 and 2014), NPL (2008)
^{18}F	1495.1(18)	LNE-LNHB (2014), AAA* (2014)
^{64}Cu	1482.2(25)	NPL (2016), CNRS/CEMHTI (2015)

* Advanced Accelerator Applications

7. **Key comparison reference values and degrees of equivalence**

7.1 KCRVs update

In May 2013 the CCRI(II) decided to calculate the key comparison reference value (KCRV) using the power-moderated weighted mean [24] 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. As proposed in [21], α is taken as $2 - 3/N$ where N is the number of results selected for the KCRV. Therefore, all SIR key comparison results can be selected for the key comparison reference value (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.

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "master-file". 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, or by consensus through electronic means (e.g., email) as discussed at the CCRI(II) meeting in 2013. In March 2015 and June 2017, the CCRI agreed for ^{99m}Tc and ^{18}F, respectively, that SIRTI linked results based on primary measurements or pressurized ionization chamber (IC) measurement are eligible for inclusion in the KCRV of the corresponding BIPM.RI(II)-K1 comparison when the IC is calibrated by a primary measurement of the same radionuclide within one year prior to the comparison date. In 2017, the CCRI also agreed that the limit of one year can be extended for practical reasons when the SIRTI comparison covers more than two nuclides at a time.

The ^{99m}Tc SIR and SIRTI results previously included in the KCRV are given in Table 7a together with the present eligible result. Using these results, the KCRV for ^{99m}Tc calculated using the power moderated weighted mean is 153 050(350) kBq, with the power α = 1.77. This can be compared successfully with the previous KCRVs of 153 070(460) kBq published in 2004 [28], 153 140(330) kBq in 2005 [29], 153 240(220) kBq in 2010 [27], 153 170(310) kBq in 2016 [22], 153 090(380) kBq in 2022 [31] and with the value of 153 400(410) kBq obtained using the SIRIC [26] efficiency curves of the SIR.

The ^{18}F SIR and SIRTI results previously included in the KCRV are given in Table 7b together with the present eligible result. Using these results, the KCRV for ^{18}F calculated using the power moderated weighted mean is 15 297(18) kBq, with the power α = 1.73. This can be compared with previous KCRVs of 15 241(71) kBq and 15 254(43) kBq published in 2003 [18, 19], 15 245(32) kBq in 2004 [16], 15 259(29) kBq in 2006 [25], 15 276(24) kBq published in 2016 [14] and 15 293(19) kBq in 2022 [31].

The KCRV for ^{64}Cu has been defined in the frame of the BIPM.RI(II)-K1.Cu-64 comparison using direct contributions to the SIR, and is equal to 80 990(340) kBq [23].

Table 7a: ^{99m}Tc SIR and SIRTI linked results included in the calculation of the KCRV

NMI	**Comparison and year of participation**	**Measurement method**	**Comparison result A_e / kBq**	**Reference**
IRA	SIR, 1984	IC* calibrated in1984 by 4π(PC)e_c-γ coincidence, HPGe detector** and 4π(NaI)γ counting	153 770(660)	[22]
PTB	SIR, 2005	IC calibrated in Nov. 2005 by 4π(PC)e_c-x and 4π(PPC)e_c-photon coincidences	152 710(640)	[22]
NPL	SIR, 2005	4π(PC)e_c-γ coincidence	153 310(660)	[22]
LNE-LNHB	SIR, 2007	4π(PC)β-γ anti-coincidence	153 180(790)	[22]
NIST	SIRTI, 2009	IC calibrated by anticoincidence measurements 2 months prior the comparison	152 840(730)	[22]
KRISS	SIRTI, 2010	IC calibrated by 4π(LS)β-γ coincidence measurements 3 months prior the comparison	154 100(1400)	[22]
NIM	SIRTI, 2012	IC calibrated by coincidence measurements 4 months prior the comparison	153 100(1200)	[22]
LNMRI/IRD	SIRTI, 2013	IC calibrated by 4π(LS)β-γ anticoincidence meas. 1 month prior the comparison	154 700(1700)	[22]
IFIN-HH	SIRTI, 2013	coincidence method	150 400(1800)	[22]
ENEA-INMRI	SIRTI, 2014	4π(LS)ec-γ(NaI) coincidence	150 750(770)	[22]
NMISA	SIRTI, 2015	IC calibrated by 4π(LS)β-γ coincidence measurements 2 months prior the comparison	156 100(2200)	[21]
POLATOM	SIRTI, 2016	4π(LS)ce-γ coincidence	154 600(1200)	[31]
NRC	SIRTI, 2017	IC calibrated by 4π(PC)-γ anti-coincidence 8 months prior the comparison	152 500(1500)	Present publication

* pressurized ionization chamber (IC) with correction factor for self-absorption in the solution of 1.0037(10)

** calibrated using ^{57}Co and ^{139}Ce calibrated sources

Table 7b: ^{18}F SIR and SIRTI linked results included in the calculation of the KCRV

NMI	Comparison and year of participation	Measurement method	Comparison result A_e / kBq	Reference
IRA	SIR, 2001	IC calibrated by 4πγ(NaI) counting and liquid scintillation	15 312(57)	[14]
NPL	SIR, 2003	4π(PC)β$^+$–γ coincidence method	15 281(39)	[14]
CIEMAT	SIR, 2004	IC calibrated by 4πβ$^+$(PPC)-γ coincidence	15 216(97)	[14]
PTB	SIR, 2005	IC calibrated by 4πβ$^+$(PC)-γ coincidence and CIEMAT/NIST	15 316(50)	[14]
LNE-LNHB	SIR, 2010	Liquid scintillation counting using TDCR	15 203(62)	[14]
VNIIM	SIRTI, 2014	4πγ(NaI) counting	15 197(97)	[20]
ENEA-INMRI	SIRTI, 2014	4π(LS)β$^+$–γ coincidence method, 4πγ(NaI) counting, and TDCR	15 368(49)	[20]
NMISA	SIRTI, 2015	IC calibrated by 4π(LS)β$^+$–γ coincidence method 1 month prior the comparison	15 328(96)	[21]
NIST	SIRTI, 2016	IC calibrated by 4π(LS)β$^+$–γ anticoincidence method 9 months prior the comparison	15 291(64)	[15]
POLATOM	SIRTI, 2016	4π(LS)ce-γ coincidence	15 323(88)	[31]
NRC	SIRTI, 2017	IC calibrated in January 2017 by 4π(PC)β$^+$-γ anti-coincidence counting 5 months prior the comparison	15 332(56)	Present publication

7.2 Degrees of equivalence

Every participant in a key comparison is entitled to have one result included in the key comparison database (KCDB) as long as the laboratory is a signatory or designated institute listed in the CIPM MRA. Normally, the most recent result is the one included. Any participant may withdraw its result only if all the participants agree.

The degree of equivalence of a particular NMI, *i*, with the KCRV is expressed as the difference D_i with respect to the KCRV

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

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 [24].

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

$$D_{ij} = D_i - D_j = A_{\mathrm{e}i} - A_{\mathrm{e}j} \qquad (3)$$

and the expanded uncertainty of this difference U_{ij} where

$$U_{ij}^2 = 4u^2(D_{ij}) = 4\left\lfloor u_i^{\,2} + u_j^{\,2} - 2u(A_{\mathrm{e}i}, A_{\mathrm{e}j})\right\rfloor \qquad (4)$$

where any obvious correlations between the NMIs (such as a traceable calibration) are subtracted using the covariance $u(A_{\mathrm{e}i}, A_{\mathrm{e}j})$, as is the correlation coming from the link of the SIRTI to the SIR. The covariance between two participants in the K4 comparison is given by

$$u(A_{\mathrm{e}i}, A_{\mathrm{e}j}) = A_{\mathrm{e}i}\, A_{\mathrm{e}j}\, (u_{\mathrm{L}} / L)^2 \qquad (5)$$

where u_{L} is the standard uncertainty of the linking factor L given above. 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.

Tables 8(a,b,c) show the matrices of the degrees of equivalence with the KCRV 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 $A_{\mathrm{e}i}$ replaced by x_i. The introductory text is that agreed for the comparison. The graph of the degrees of equivalence with respect to the KCRV (identified as x_{R} in the KCDB) is shown in Figure 3(a,b,c) in relative terms. 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.

The degrees of equivalence of the BIPM.RI(II)-K1 key comparisons for ^{99m}Tc and ^{18}F have been updated following the update of the KCRV for ^{99m}Tc and ^{18}F.

The CCRI(II)-K3.F-18 and APMP.RI(II)-K3.F-18 key comparisons linked to the BIPM.RI(II)-K1.F-18 key comparison earlier [17] are now older than 20 years and have been withdrawn from the KCDB.

Conclusion

In 2017, the NRC (Canada) hosted the SIRTI to participate in the BIPM ongoing key comparison for activity measurement of ^{11}C, ^{18}F, ^{64}Cu and ^{99m}Tc (the BIPM.RI(II)-K4 series). The results of the ^{11}C comparison published earlier have been slightly updated. The other three K4 comparisons are linked to the corresponding BIPM.RI(II)-K1 comparisons (the SIR comparisons).

The key comparison reference values for ^{99m}Tc and ^{18}F, usually defined in the frame of the K1 comparisons, have been updated to include the NRC result from the K4 comparisons. The degrees of equivalence with the respective key comparison reference values have been evaluated for NRC's participation in the BIPM.RI(II)-K4 comparisons, and degrees of equivalence for other K1 or K4 participants have been updated. The degrees of equivalence have been approved by the CCRI(II) and are published in the BIPM key comparison database.

Other results may be added when other NMIs contribute with ^{11}C, ^{18}F, ^{64}Cu and ^{99m}Tc activity measurements to the K4 or K1 comparisons or take part in other linked Regional Metrology Organization comparisons. 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 [7] are those available in the KCDB.

Table 8a. Table of degrees of equivalence and introductory text for ^{99m}Tc

Key comparison BIPM.RI(II)-K1.Tc-99m

MEASURAND : Equivalent activity of ^{99m}Tc

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 153.05 MBq
with a standard uncertainty, u_R = 0.35 MBq (see 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
$U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ where w_i is the weight of laboratory *i* contributing to the calculation of x_R.

Linking BIPM.RI(II)-K4.Tc-99m to BIPM.RI(II)-K1.Tc-99m

The value x_i is the SIRTI equivalent activity for laboratory *i* participant in BIPM.RI(II)-K4.Tc-99m
having been normalized using the NPL and the LNE-LNHB as linking laboratories (see Final report).
The degree of equivalence of laboratory *i* participant in BIPM.RI(II)-K4.Tc-99m 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,
$U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ where w_i is the weight of laboratory *i* contributing to the calculation of x_R.

These statements make it possible to extend the BIPM.RI(II)-K1.Tc-99m matrices of equivalence to the other participants in BIPM.RI(II)-K4.Tc-99m.

Table 8a continued

Lab *i*	D_i / MBq	U_i / MBq
BEV	2.6	2.7
MKEH	1.3	3.3
PTB	-0.4	1.3
LNE-LNHB	0.1	1.6
NPL	0.2	1.6
NIST	-0.3	1.5
KRISS	1.0	2.7
NMIJ	-0.7	2.3
NIM	0.0	2.4
CNEA	7.1	4.3
LNMRI/IRD	1.6	3.3
IFIN-HH	-2.2	2.9
VNIIM	3.5	4.9
ENEA-INMRI	-2.3	1.5
NMISA	3.0	4.3
POLATOM	1.5	2.4
NRC	-0.6	2.9

Figure 3a. Graph of degrees of equivalence with the KCRV for ^{99m}Tc

(as it appears in Appendix B of the MRA)

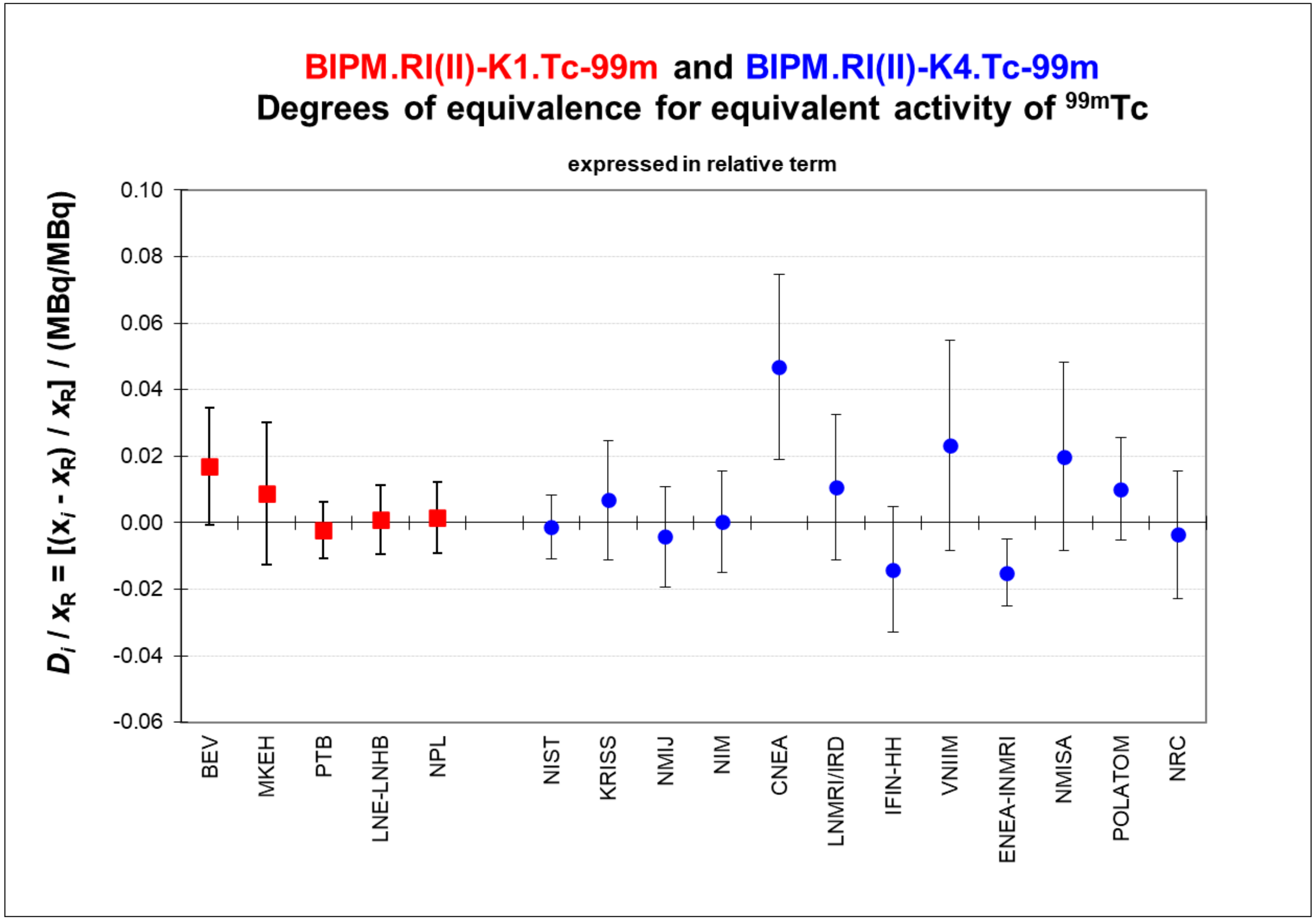


Table 8b. Introductory text and table of degrees of equivalence for ^{18}F
Key comparison BIPM.RI(II)-K1.F-18

MEASURAND : Equivalent activity of ^{18}F

Key comparison reference value: the SIR reference value for this radionuclide is x_R = 15.297 MBq
with a standard uncertainty, u_R = 0.018 MBq (see 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.

Linking BIPM.RI(II)-K4.F-18 to BIPM.RI(II)-K1.F-18

The value x_i is the SIRTI equivalent activity for laboratory *i* participant in BIPM.RI(II)-K4.F-18
multiplied by the linking factor to BIPM.RI(II)-K1.F-18 (see Final Report).

The degree of equivalence of laboratory *i* participant in BIPM.RI(II)-K4.F-18 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.

These statements make it possible to extend the BIPM.RI(II)-K1.F-18 matrices of equivalence
to all participants in the BIPM.RI(II)-K4.F-18 comparisons.

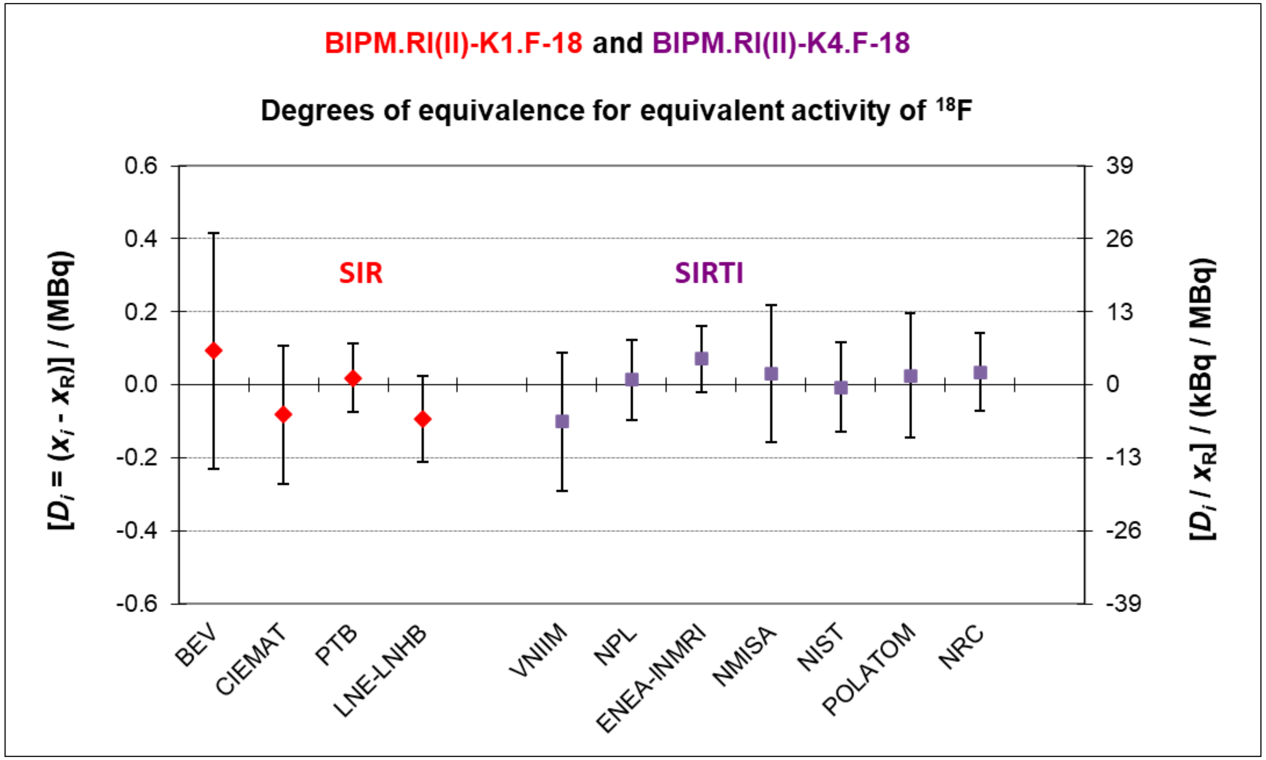
Table 8b continued

Lab *i*

	D_i / MBq	U_i / MBq
BEV	**0.09**	0.32
CIEMAT	**-0.08**	0.19
PTB	**0.019**	0.094
LNE-LNHB	**-0.09**	0.12
VNIIM	**-0.10**	**0.19**
NPL	**0.01**	**0.11**
ENEA-INMRI	**0.071**	**0.092**
NMISA	**0.03**	**0.19**
NIST	**-0.01**	**0.12**
POLATOM	**0.03**	**0.17**
NRC	**0.03**	**0.11**

Figure 3b. 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

Table 7c. Table of degrees of equivalence and introductory text for ^{64}Cu

Key comparison BIPM.RI(II)-K1.Cu-64

MEASURAND : **Equivalent activity of ^{64}Cu**

Key comparison reference value: **the SIR reference value x_R for this radionuclide is 80.99 MBq, with a standard uncertainty u_R of 0.34 MBq.**
The value x_i is taken as the equivalent activity for laboratory *i*.

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms:
$D_i = (x_i - x_R)$ and U_i, its expanded uncertainty (k = 2), both expressed in MBq, and
$U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$ when each laboratory has contributed to the calculation of x_R.

Linking BIPM.RI(II)-K4.Cu-64 to BIPM.RI(II)-K1.Cu-64

The value x_i is the SIRTI equivalent activity for laboratory *i* participant in BIPM.RI(II)-K4.Cu-64 multiplied by the linking factor to BIPM.RI(II)-K1.Cu-64 (see Final Report).

The degree of equivalence of laboratory *i* participant in BIPM.RI(II)-K4.Cu-64 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.

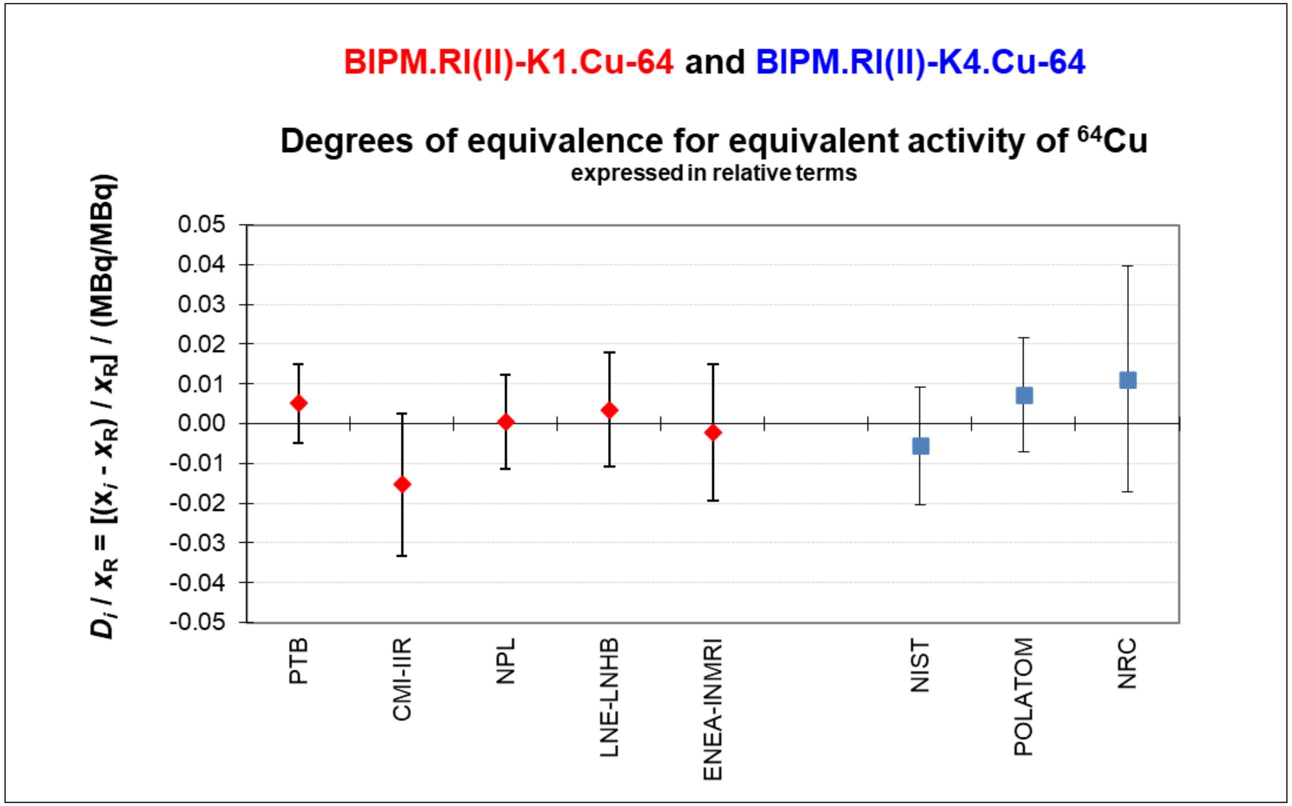
These statements make it possible to extend the BIPM.RI(II)-K1.Cu-64 matrices of equivalence to all participants in the BIPM.RI(II)-K4.Cu-64 comparisons.

Lab *i* ⇩	D_i / MBq	U_i / MBq
PTB	**0.41**	0.80
CMI-IIR	**-1.2**	1.4
NPL	**0.0**	1.0
LNE-LNHB	**0.3**	1.2
ENEA-INMRI	**-0.2**	1.4

Table 7c continued

	D_i	U_i
	/ MBq	
NIST	**-0.4**	**1.2**
POLATOM	**0.6**	**1.2**
NRC	**0.9**	**2.3**

Figure 3c. **Graph of degrees of equivalence with the KCRV for ^{64}Cu**
(as it appears in Appendix B of the MRA)



References

[1] Ratel G., 2007, The Système International de Référence and its application in key comparisons, *Metrologia* **44**(4), S7-S16.
[2] Remit of the CCRI(II) Transfer Instrument Working Group, 2009, CCRI(II) working document CCRI(II)/09-15.
[3] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Helmer R., Nichols A., Schönfeld E., Dersch R., 2004, Table of radionuclides, *Monographie BIPM-5*, volume 1.
[4] Bé M.-M., Helmer R., 2011, Decay Data Evaluation Project working group, www.nucleide.org/ DDEP_WG/DDEPdata.htm.
[5] NUDAT2.5, National Nuclear Data Center, Brookhaven National Laboratory, based on ENSDF and the Nuclear Wallet Cards.
[6] Bouchard J., 2000, *Appl. Radiat. Isot.* **52**, 441-446.
[7] SIR Transfer Instrument. Protocol for the ongoing comparisons on site at the NMIs, BIPM.RI(II)-K4. Published on the CIPM MRA KCDB website.
[8] CIPM MRA: *Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes*, International Committee for Weights and Measures, 1999, 45 pp. http://www.bipm.org/pdf/mra.pdf.
[9] Michotte C. *et al.*, The SIRTI, a new tool developed at the BIPM for comparing activity measurements of short-lived radionuclides world-wide, *Rapport BIPM*-2013/02.
[10] Michotte C. *et al.*, Calibration of the SIRTI against the SIR and trial comparison of ^{18}F and ^{99m}Tc at the NPL. *In preparation*.
[11] Baerg A.P. *et al.*, 1976, Live-timed anti-coincidence counting with extending dead-time circuitry, *Metrologia* **12**, 77-80.
[12] Fitzgerald R., 2016, Corrections for the combined effects of decay and dead time in live-timed counting of short-lived radionuclides, *Appl. Radiat. Isot.* **109**, 335-340.
[13] Sibbens G., 1991, A comparison of NIST/SIR-, NPL-, and CBNM 5 ml ampoules, GE/R/RN/14/91, CEC-JRC Central Bureau for Nuclear Measurements, Belgium.
[14] Michotte C. et al., 2016, Update of the BIPM comparison BIPM.RI(II)-K1.F-18 of activity measurements of the radionuclide ^{18}F to include the 2010 result of the LNE-LNHB (France). *Metrologia*, 2016, **53,** *Tech. Suppl.*, 06004.
[15] Michotte C., Nonis M., Bergeron D., Cessna J., Fitzgerald R., Pibida L., Zimmerman B., Fenwick A., Ferreira K., Keightley J., Da Silva I., Activity measurements of the radionuclides ^{18}F and ^{64}Cu for the NIST, USA, in the ongoing comparisons BIPM.RI(II)-K4.F-18 and BIPM.RI(II)-K4.Cu-64, *Metrologia*, 2017, **54**, Tech. Suppl. 06011.
[16] Ratel G., Michotte C., García-Toraño E., Los Arcos J.-M., Update of the BIPM comparison BIPM.RI(II)-K1.F-18 of activity measurements of the radionuclide ^{18}F to include the CIEMAT, *Metrologia*, 2004, **41**, *Tech. Suppl.*, 06016.
[17] Ratel G., Michotte C., Woods M.J., Comparisons CCRI(II)-K3.F-18 and APMP.RI(II)-K3.F-18 of activity measurements of the radionuclide ^{18}F and links to the key comparison reference value of the BIPM.RI(II)-K1.F-18 comparison, *Metrologia*, 2005, **42**, *Tech. Suppl.*, 06007.
[18] Ratel G., Michotte C., BIPM comparison BIPM.RI(II)-K1.F-18 of activity measurements of the radionuclide ^{18}F, *Metrologia*, 2003, **40**, *Tech. Suppl.*, 06005.
[19] Ratel G., Michotte C., Woods M.J., Update of the BIPM comparison BIPM.RI(II)-K1.F-18 of activity measurements of the radionuclide ^{18}F to include the NPL, *Metrologia*, 2003, **40**, *Tech. Suppl.*, 06027.

[20] Michotte C., Nonis M., Alekseev I.V., Kharitonov I.A., Tereshchenko E.E., Zanevskiy A.V., Keightley J.D., Fenwick A., Ferreira K., Johansson L., Capogni M., Carconi P., Fazio A., De Felice P., Comparison of ^{18}F activity measurements at the VNIIM, NPL and the ENEA-INMRI using the SIRTI of the BIPM, *Applied Radiation and Isotopes*, 2016, **109**, 17–23.

[21] Michotte C., Nonis M., Van Rooy M.W., Van Staden M.J. and Lubbe J., Activity measurements of the radionuclides ^{18}F and ^{99m}Tc for the NMISA, South Africa in the ongoing comparisons BIPM.RI(II)-K4.F-18 and BIPM.RI(II)-K4.Tc-99m, *Metrologia*, 2017, **54**, *Tech. Suppl.*, 06001.

[22] Michotte C., Nonis M., Alekseev I.V., Kharitonov I.A., Tereshchenko E.E., Zanevskiy A.V., Capogni M., De Felice P., Fazio A., Carconi P., Activity measurements of the radionuclide ^{99m}Tc for the VNIIM, Russian Federation and ENEA-INMRI, Italy, in the ongoing comparison BIPM.RI(II)-K4.Tc-99m and KCRV update in the BIPM.RI(II)-K1.Tc-99m comparison, *Metrologia*, 2016, **53**, *Tech.Suppl.*, 06014.

[23] Michotte C., *et al.*, Update of the BIPM comparison BIPM.RI(II)-K1.Cu-64 of activity measurements of the radionuclide ^{64}Cu to include the 2009 results of the CMI–IIR (Czech Rep.) and the NPL (UK), the 2010 result of the LNE–LNHB (France) and the 2011 result of the ENEA–INMRI (Italy). *Metrologia*, 2013, **50**, *Tech. Suppl.*, 06021.

[24] Pommé S., Keightley, J., Determination of a reference value and its uncertainty through a power-moderated mean, *Metrologia*, 2015, **52(3)**, S200-S212.

[25] Ratel G., Michotte C., Kossert K., Janßen H., Update of the BIPM comparison BIPM.RI(II)-K1.F-18 of activity measurements of the radionuclide ^{18}F to include the PTB, *Metrologia*, 2006, **43**, *Tech. Suppl.*, 06001.

[26] Cox M.G., Michotte C., Pearce A.K., Measurement modelling of the International Reference System (SIR) for gamma-emitting radionuclides, Monographie BIPM-7, 2007, 48 pp.

[27] Michotte C., Courte S., Ratel G., Moune M., Johansson L., Keightley J., Update of the BIPM.RI(II)-K1.Tc-99m comparison of activity measurements for the radionuclide ^{99m}Tc to include new results for the LNE-LNHB and the NPL, *Metrologia,*, 2010, **47**, *Tech. Suppl.*, 06026.

[28] Ratel G., Michotte C., BIPM comparison BIPM.RI(II)-K1.Tc-99m of activity measurements of the radionuclide ^{99m}Tc, *Metrologia*, 2004, **41**, *Tech. Suppl.*, 06005.

[29] Ratel G., Michotte C., Johansson L., Update of the BIPM.RI(II)-K1.Tc-99m comparison of activity measurements for the radionuclide ^{99m}Tc to include the NPL, *Metrologia*, 2005, **42**, *Tech. Suppl.*, 06015.

[30] Galea R., Michotte C., Nonis M., Moore K., El Gamal I., Keightley J., Fenwick A., *Applied Radiation and Isotopes*, 2019, **154,** 108834.

[31] Michotte C., Dziel T., Listkowska A., Ziemek T., Tymiński Z. and Da Silva I., Activity measurements of the radionuclides ^{99m}Tc, ^{18}F and ^{64}Cu for the POLATOM, Poland, in the ongoing comparisons BIPM.RI(II)-K4 series and KCRV update in the corresponding BIPM.RI(II)-K1 comparison, *Metrologia*, 2022, **59**, *Tech. Suppl.*, 06004.

Appendix 1. **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
		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
		CeBr3	CB

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 radiations	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

Appendix 2a. Uncertainty budget for the IC calibration done by ^{18}F primary measurements (January 2017). The results of the 4P-PC-PO-NA-GR-AC in **bold red** are submitted for equivalence and the other is presented for comparison.

Uncertainty Contributions due to	**Comments**	**Evaluation method**	**Relative standard uncertainties x10^4** **4P-PC-PO-NA-GR-AC**	**Relative standard uncertainties x10^4** 4P-PC-PO-NA-GR-CO	**Relative standard uncertainties x10^4** 4P-LS-PO-00-00-CN	**Relative standard uncertainties x10^4** 4P-LS-PO-00-00-TD
Counting statistics (for primary determination)	Weighted uncertainty on the measurement of 6 dried point sources on VYNS* films.	A	**2**	1		
Decay data	Uncertainty in positron decay branching ratio 0.9686(19) and half life 1.828 90(23) h.	B	**21**	21		
Live time	Previous sensitivity tests.	B	**5**			
Dead time	Previous sensitivity tests.	B		1	1	1
Resolving time	Previous sensitivity tests.	B		1		
Extrapolation	Maximum uncertainty of a statistical fit of 10 PC thresholds x 6 sources.	A	**20**	13		
Quenching parameter	Standard deviation of calculated activity for model kb=0.005-0.012 with zero slope fits	B			41	41
^3H Tracer	Propagated error on tritium traceable tracer.	B			10	
Stability of LSC cocktail	Standard uncertainty of 10 repeated measurements and one LSC cocktails.	A			10	10
Background (primary)	Repeated measurement of background under experimental conditions.	A	**11**	14		
Weighing	Estimate of uncertainty in dilution factor, masses of point sources and ampoule filling volumes due to balance calibration	B	**4**	4	4	4

*ploy-vinyl chloride, poly-vinyl acetate compound.

Appendix 2a Continued:

Uncertainty Contributions due to	Comments	Evaluation method	Relative standard uncertainties x10^4 4P-PC-PO-NA-GR-AC	Relative standard uncertainties x10^4 4P-PC-PO-NA-GR-CO	Relative standard uncertainties x10^4 4P-LS-PO-00-00-CN	Relative standard uncertainties x10^4 4P-LS-PO-00-00-TD
^{18}F half life	Uncertainty due to decay correction from IC measurement to reference time.	B	**6**	6	6	6
Counting statistics (IC response determination)	Standard deviation of the mean of two repeated measurements each of four ampoules. Each measurement is determined from five charge measurements.	A	**1**	1	1	1
Measurement method	Standard deviation of four determinations of each IC measurement set.	A	**1**	1	1	1
Background (IC)	Standard deviation of ongoing background measurements.	A	**1**	1	1	1
Stability	Standard deviation of ongoing reference source measurements.	A	**3**	3	3	3
Charge collection	Electrometer calibration.	B	5	5	5	5
Impurities	None detected.					
Relative combined standard uncertainty* **			**33**	31	45	44

Appendix 2b. Uncertainty budget for the IC calibration done by ^{64}Cu primary measurements (March 2017). The results of the 4P-PC-PO-NA-GR-AC in **bold red** are submitted for equivalence and the other is presented for comparison.

Uncertainty Contributions due to	**Comments**	**Evaluation method**	**Relative standard uncertainties x10^4** **4P-PC-PO-NA-GR-AC**	**Relative standard uncertainties x10^4** 4P-PC-PO-NA-GR-CO
Counting statistics (for primary determination)	Weighted uncertainty on the measurement of 10 dried point sources on VYNS* films.	A	**24**	7
Decay data	Uncertainty in decay scheme data.	B	**82**	82
Live time	Previous sensitivity tests.	B	**5**	
Dead time	Previous sensitivity tests.	B		1
Resolving time	Previous sensitivity tests.	B		1
Extrapolation	Weighted uncertainty of a statistical fit of 10 PC thresholds x 10 sources for each of two extrapolation schemes.	A	**96**	91
Background (primary)	Repeated measurement of background under experimental conditions.	A	**10**	10
Weighing	Estimate of uncertainty in dilution factor, masses of point sources and ampoule filling volumes due to balance calibration	B	**4**	4
^{64}Cu half life	Uncertainty due to decay correction from IC measurement to reference time.	B	**1**	1
Counting statistics (IC response determination)	Standard deviation of the mean of two repeated measurements each of four ampoules. Each measurement is determined from five charge measurements.	A	**1**	1

*poly-vinyl chloride, poly-vinyl acetate compound.

Appendix 2b Continued

Uncertainty Contributions due to	Comments	Evaluation method	Relative standard uncertainties x10^4 4P-PC-PO-NA-GR-AC	Relative standard uncertainties x10^4 4P-PC-PO-NA-GR-CO
Measurement method	Standard deviation of four determinations of each IC measurement set.	A	1	1
Background (IC)	Standard deviation of ongoing background measurements.	A	1	1
Stability	Standard deviation of ongoing reference source measurements.	A	3	3
Charge collection	Electrometer calibration.	B	5	5
Impurities	Gamma spectroscopy identified ^{192}Ir (2.3e^{-5}% of ^{64}Cu activity at ref. time) and ^{60}Co (1.0e^{-5}% of ^{64}Cu activity at ref. time).	B	1	1
Relative combined standard uncertainty* **			**129**	123

Appendix 2c. Uncertainty budget for the IC calibration done by ^{99m}Tc 4P-PC-MX-NA-GR-AC primary measurements (October 2016).

Uncertainty Contributions due to	**Comments**	**Evaluation method**	**Relative standard uncertainties x10^4**
Counting statistics (for primary determination)	Weighted uncertainty on the measurement of 15 dried point sources on VYNS* films.	A	18
Live time	Previous sensitivity tests.	B	5
Extrapolation	Statistical fit of 10 PC thresholds x 15 sources.	A	98
Weighing	Estimate of uncertainty in dilution factor, masses of point sources and ampoule filling volumes due to balance calibration	B	4
^{99m}Tc half life	Uncertainty due to decay correction from IC measurement to reference time.	B	5
Counting statistics (IC response determination)	Standard deviation of the mean of two repeated measurements each of three ampoules. Each measurement is determined from five charge measurements.	A	1
Measurement method	Standard deviation of four determinations of each IC measurement set.	A	2
Background	Standard deviation of ongoing background measurements.	A	1
Stability	Standard deviation of ongoing reference source measurements.	A	3
Charge collection	Electrometer calibration.	B	5
Filling amount correction	Monte Carlo evaluation for protocol filling height variance.	B	4
Impurities	None detected.		
Relative combined standard uncertainty* **			100

*ploy-vinyl chloride, poly-vinyl acetate compound.