Final Report of the key comparison CCRI(II)-K2.Fe-55.2019 of activity concentration measurements of a ^{55}Fe solution

R. Broda[1], I. Bonková[7], M. Capogni[2], P. Carconi[2], P. Cassette[4], R. Coulon[3], S. Courte[3], P. De Felice[2], T. Dziel[1], A. Fazio[2], C. Frechou[4], R. Galea[5], E. García-Toraño[10], E. Kołakowska[1], K. Kossert[6], M. Krivošík[7], E. Lech[1], K.B. Lee[8], J. Liang[9], A. Listkowska[1], H. Liu[9], N. Navarro[10], O.J. Nähle[6], M. Nowicka[1], M. van Rooy[11], B. Sabot[4], P. Saganowski[1], Y. Sato[12], Z. Tymiński[1], A. Yunoki[12], M. Zhang[9], T. Ziemek[1].

[1] *National Centre for Nuclear Research Radioisotope Centre POLATOM (NCBJ RC POLATOM), 7 Andrzeja Sołtana, 05-400 Otwock, Poland*
[2] *ENEA - Italian National Institute of Ionizing Radiation Metrology (ENEA-INMRI), Casaccia Research Centre, Via Anguillarese 301, I-00123, Italy*
[3] *Bureau International des Poids et Mesures (BIPM), Pavillon de Breteuil, F-92312, Sèvres, Cedex, France*
[4] *Université Paris-Saclay, CEA, LIST, Laboratoire National Henri Becquerel (LNE-LNHB), CEA-Saclay, F-91120 Palaiseau, France.*
[5] *National Research Council of Canada (NRC), 1200 Montreal Road, Building M-35, Ottawa, ON, K1A0R6, Canada*
[6] *Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany*
[7] *Slovenský metrologický ústav (SMU), Karlovenská 63, 842 55 Bratislava, Slovakia*
[8] *Korea Research Institute of Standards and Science (KRISS), Gaejong-ro 267, Yuseong-gu, Daejon, Republic of Korea*
[9] *National Institute of Metrology (NIM), 18 Bei San Huan Dong Lu, Chaoyang District, Beijing 100029, China*
[10] *Laboratorio de Metrologia de Radiaciones, Centro de Investigaciones Energéeticas, Medioambientales y Tecnologicas (CIEMAT), Avenida Complutense 40, 28040 Madrid, Spain*
[11] *National Metrology Institute of South Africa (NMISA), Radioactivity Standards Section, 15 Lower Hope Road, CSIR campus, Rosebank, Cape Town, 7700, South Africa*
[12] *National Metrology Institute of Japan (NMIJ), 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan*

E-mail : ryszard.broda@polatom.pl

Abstract

A comparison of the activity concentration of an ^{55}Fe solution was organized by the POLATOM under the auspices of the CCRI(II). The solution was sent to thirteen laboratories; ready-to-measure sources were also prepared using the same ^{55}Fe solution in diffusive (polyethylene) vials and sent to twelve laboratories. Twelve measurement reports were received. The results, as well as relevant information on the measurement techniques, are presented and analysed in this paper. The differences in the results obtained from measurements in non-diffusive (glass) and diffusive vials are indicated.

1 Introduction

Iron-55 is a radionuclide decaying by pure electron capture (EC) towards ^{55}Mn [1]. Iron-55 is a low-energy, difficult-to-measure [2], radionuclide. Institutes that demonstrate their capability

to measure the activity of an ^{55}Fe solution may be able to use the result as supporting evidence of their capability to measure other EC-radionuclides in their laboratory. Therefore, during the CCRI(II) meeting in 2019, POLATOM and ENEA-INMRI proposed to organize a new key comparison for an ^{55}Fe solution. The proposal was accepted by the CCRI(II) and the key comparison CCRI(II)-K2.Fe-55.2019 was registered.

An important aim of the comparison piloted by POLATOM was to develop a new activity standard of ^{55}Fe, as it had been fourteen years since the previous CCRI(II)-K2.Fe-55 comparison in 2006 [3]. It is worth mentioning here that 17 laboratories participated in the previous comparison. Various measurement methods were then used: the CIEMAT/NIST (C/N) method; the Triple to Double Coincidence Ratio (TDCR) method; a pressurized proportional counter working in coincidence or anticoincidence with a NaI(Tl) detector and using various tracers, i.e. ^{51}Cr, ^{54}Mn and ^{65}Zn; defined solid angle counting using a pressurized proportional counter or a calibrated Si(Li) detector; or the isothermal calorimetric method. The present international comparison has involved twelve laboratories listed in Table 1, using only the TDCR or C/N Liquid Scintillation Counting (LSC) technique. Some of the laboratories did not participate in the previous comparison.

The second aim of the comparison was to support the Bureau International des Poids et Mesures (BIPM) in validating the Extended International Reference System (ESIR). The ESIR reference system is based on a liquid-scintillation instrument that uses the TDCR method as a comparator for national standards of pure beta-emitting radionuclides. The goal of the ESIR is to provide an impartial, robust, long-term service for comparing national standards of pure beta-emitting radionuclides. This comparison should support and enable the BIPM to verify the operation of the ESIR system [4], the TDCR instrument and source preparation. The BIPM measurement results are not intended to be used to support claims for CMCs and are normally interpreted to be a parameter that is proportional to the activity rather than as a primary standard.

A further aim of the comparison was to compare the results from measurements of the activity concentration (activity per unit mass) of ^{55}Fe in ready-to-measure sources prepared by the pilot laboratory to the final result of the key comparison. The results from the ready-to-measure sources are not included in the CCRI(II) comparison official results but are an additional study to gain a greater understanding of the parameters that can affect liquid scintillation counting. Identical diffusive (polyethylene) vials, identical batch of Ultima Gold^{TM} scintillator and identical method of sources preparation should make it possible to reduce significantly the variation in results associated with the sources themselves. Furthermore, raw measurement data for the ready-to-measure sources were sent by participants to the pilot laboratory who recalculated the results using one selected code, enabling the comparison of the software packages used by participants.

The measurand was the activity concentration of the ^{55}Fe solution at the reference date of 19th October 2019, 12:00 UTC. The recommended half-life is $T_{1/2}(^{55}\text{Fe}) = 2.747\,(8)$ a [1].

2 Participants

Twelve laboratories participated in this comparison. The participating laboratories and the contact persons of each laboratory are presented in Table 1.

Two of the fourteen laboratories withdrew. One of the laboratories intended to carry out the measurements by using a PPC detector, but it obtained results of poor quality for an international comparison. The other laboratory encountered significant administrative and customs problems and was not allowed to receive the ^{55}Fe solution.

3 Solution shipment

The solution was prepared at the POLATOM by dilution of a stock solution to reach an activity concentration of about 100 kBq/g at the reference date. The chemical composition was $^{55}FeCl_2$ in a carrier solution containing 25 µg Fe in 1 mL of 1 M HCl. The radionuclidic purity of the ^{55}Fe solution was checked by gamma-ray spectrometry and by beta-ray spectrometry using a Tri-Carb 2910 TR liquid scintillation counter: a ^{60}Co impurity of $5.5(4) \times 10^{-3}$ % of the main radionuclide was detected.

Thirteen flame-sealed ampoules, each containing about 1.5 mL of solution, were prepared and 12 ampoules were sent to the laboratories who expressed their interest in participating in the exercise. The total activity of each ampoule was less than 150 kBq on the date of shipment.

Table 1. List of participants

Laboratory	Country	Responsible person
BIPM	France	Romain COULON
CIEMAT	Spain	Nuria NAVARRO
ENEA-INMRI	Italy	Marco CAPOGNI
KRISS	South Korea	Kyung Beom LEE
LNE-LNHB	France	Carole FRECHOU
NIM	China	Juncheng LIANG
NMIJ	Japan	Yasushi SATO
NMISA	South Africa	Milton VAN ROOY
NRC	Canada	Raphael GALEA
POLATOM*	Poland	Ryszard BRODA
PTB	Germany	Karsten KOSSERT
SMU	Slovakia	Matej KRIVOŠÍK

* Pilot Laboratory

In addition to the flame-sealed ampoules, a set of 78 ready-to-measure sources was prepared in 20 mL diffusive vials containing 10 mL of Ultima Gold^{TM}, as liquid scintillator, plus a mass of radioactive material ranging from 30 mg to 50 mg. The activity of sources was checked at POLATOM during the period of five weeks using a TDCR and a Tri-Carb 2910 TR counter. The homogeneity of sources (activity per gram) was within ± 0.22 %. Each source was measured for a time interval of 1000 s. A relative standard deviation of 0.22 % was obtained. The relative standard deviation of the tSIE parameter was found to be 0.56 %. A set of six ready-to-measure sources and an additional blank vial were sent to each participant. The total activity of each ready-to-measure source was less than 5.5 kBq at the date of shipment. The exact masses of the ready-to-measure sources were unknown by the participants.

The shipment of the ampoules and the ready-to-measure sources was organized by POLATOM as an exempted package shipment. Packages were sent to the participants on 7 September 2019 and delivered from the 9 to the 30 October 2019. After the measurements, five participating laboratories sent three of the ready-to-measure sources back to POLATOM where the long-term stability of the sources was confirmed.

4 Reception of the results

The deadline for the submission of the results was originally fixed as 3 January 2020. Some laboratories experienced delays and the final deadline was postponed to 30 June 2020. The last result was received in March 2020. The participants were asked to send their measurement reporting forms to the pilot laboratory, only after notification that the POLATOM had submitted its result to the executive secretary of the CCRI(II). This submission was done by e-mail, sent to the BIPM on 17 December 2019.

5 Measurement results

The LSC TDCR method was used by the majority of the laboratories. Most laboratories used their own bespoke TDCR counters. The ENEA-INMRI used the commercial TDCR counter, Hidex 300SL "Metro" version. The second ENEA-INMRI TDCR bespoke portable counter was used to evaluate the uncertainty budget based using the difference in the results between the two counters. The PTB used their own TDCR counters and presented the final result as the unweighted mean of the measurements in these counters. The NRC laboratory used a Wallac 1410 counter and submitted the result from a C/N efficiency tracing method with ^{3}H as the tracer. All measurement results of the activity concentrations of the ^{55}Fe solution obtained by participants are presented in Table 2 and are plotted in Figure 1.

Three laboratories, CIEMAT, NRC and SMU, additionally provided the results of other measurements made by the TDCR or C/N methods, which were not used by them to determine the final result. Nevertheless, these supplementary results are also shown in Table 2 and Figure 1 with the index "suppl".

Four laboratories, ENEA-INMRI, NMISA, NRC, and PTB, found a ^{60}Co impurity of $10(3)\times10^{-6}$ %, $5.7(8)\times10^{-3}$ %, $20(2)\times10^{-5}$ % and $50(7)\times10^{-6}$ % of the main radionuclide respectively.

Based on the activity of ready-to-measure sources reported by individual laboratories, POLATOM calculated the average activity per gram of the solution after taking into account the masses of the sources. The results are presented in Table 4 and plotted in Figure 2. The raw measurement results of ready-to-measure sources provided by the participants were analyzed at POLATOM using a simplified calculation code Fe55_6, developed by LNE-LNHB, considering only mono-energetic emission of ^{55}Fe (Table 4). The only decay data used by this code is K shell capture probability, P_K, and no emission energy is needed [5]. The parameter $kB = 0.010$ cm MeV^{-1} was adopted.

Information on the sources used, the measurement instruments and detection efficiency calculation methods are presented in Table A1 and A2 of the appendix.

During a meeting on 7 November 2019, the Key Comparison Working Group (KCWG(II)) of the CCRI(II) decided to provide information and recommendations concerning fractional electron-capture (EC) probabilities to the participants of this comparison. All participants received this information and were asked to analyze their data using new values for the EC probabilities of the ground-state to ground-state transition which were $P_K = 0.88044$ (34), $P_{L1} = 0.10188$ (23), $P_{L2} = 5.012$E-4 (26), $P_{M1} = 0.01599$ (10), $P_{M2} = 7.43$E-5 (7) and $P_{N1} = 0.001120$ (8). These probabilities were calculated with the new program BetaShape [6] which is considered to apply state-of-the-art calculations. These new recommended data had a significant effect on the measurement results: the activity calculated with these data is 0.6%

higher than the value obtained with the previous fractional EC-probabilities [7]. The recommended data were used by all participants except the KRISS.

6 Uncertainty budget

The uncertainty budgets for the measurements, using the TDCR method and reported by each participant, are presented in Table A3 of the appendix. The dominant uncertainty contribution generally comes from the counting model, the decay data and counting statistics. There is a significant dispersion of the reported combined uncertainties, the maximum reported uncertainty is 5.8 times the minimum. After omitting the largest and the smallest uncertainties, the two most extreme values differ by a factor of 2.1.

The uncertainty budget of the ^{55}Fe measurement using the C/N method reported by the NRC is presented in Table A4 of the appendix.

The uncertainty budgets for the ^{55}Fe ready-to-measure sources, using the TDCR method and reported by the participants, are presented in Table A5 of the appendix. The uncertainty budget of the ^{55}Fe ready-to-measure sources using the C/N method reported by the NRC is presented in Table A6 of the appendix. Uncertainties related to counting statistics were estimated at POLATOM using the masses of the ready-to-measure sources and the activity values reported by the participants. The dominant uncertainty contribution is similar to that identified for budgets in Table A3. The maximum reported uncertainty is twice the minimum.

7 Final results and evaluation of the *KCRV*

The final results of measurements of the ^{55}Fe activity concentration at the reference date of 19 October 2019 are presented in Table 2 and plotted in Figure 1. The different types of vial used for source preparation are indicated. It can be observed that all the results, except for the lowest result with a low estimated uncertainty, are in agreement within the reported uncertainties. The calculated power-moderated mean (*PMM$_{sol}$*) [8] and associated uncertainty was 100.88(30) kBq/g at the reference date of 19 October 2019.

The Peirce [9] and the Chauvenet [10] criteria were used for identifying outliers. According to both criteria, the reported KRISS results should be rejected as an outlier. The calculated *PMM$_{sol}$* and associated uncertainty was 101.07(24) kBq/g at the reference date of 19 October 2019.

While discussing the results, the participants pointed to the problem of internal light reflection in non-diffusive vials that results in a deviation from the Poisson distribution for light emitted. The expected number of photons emitted from the clear vial depends on the point of emission in the liquid scintillator. The optimum statistical distribution is in this case a compound Poisson distribution, which is not included in the TDCR calculation software used. In order to try to determine the best key comparison reference value (*KCRV*), taking into account the state-of-the-art, the participants decided to use only the results for diffusive vials for the determination of the *KCRV* [11, 12]. Results from non-diffusive vials and ready-to-measure sources were excluded from this calculation.

A new characterisation of the distribution of the results is reported in Table 3. The new calculated *PMM$_{sol}$* and associated uncertainty, is **101.49(23) kBq/g** at the reference date of **19 October 2019**. This *PMM$_{sol}$* value has been adopted to be the *KCRV*.

Table 2. Final results of the CCRI(II)-K2.Fe-55.2019 comparison of ^{55}Fe activity concentration at the reference date of 19 October 2019

Laboratory	Activity concentration (kBq/g)	Standard uncertainty (kBq/g)	Relative standard uncertainty (%)	Measure-ment method	Vials used for sources preparation	Mean date of measure-ments
BIPM	99.79	0.62	0.62	TDCR	non-diffusive	23 Oct 2019
CIEMAT	101.19	0.93	0.92	TDCR	diffusive & non-diffusive	19 Dec 2019
ENEA-INMRI	100.105	0.704	0.7	TDCR	non-diffusive	22 Oct 2019
KRISS	98.47	0.80	0.81	TDCR	non-diffusive	28 Feb 2020
LNE-LNHB	101.23	0.42	0.41	TDCR	diffusive	Oct-Nov 2019
NIM	100.34	0.46	0.45	TDCR	non-diffusive	10 Jan 2020
NMIJ	100.4	0.6	0.59	TDCR	non-diffusive	25 Dec 2019
NMISA	102.18	0.64	0.63	TDCR	diffusive	14 Nov 2019
NRC	102.1	1.6	1.57	C/N	diffusive	10 Nov 2019
POLATOM	101.27	0.39	0.39	TDCR	diffusive	17 Jul 2019
PTB	101.69	0.51	0.50	TDCR	diffusive	15 Sep 2019
SMU	101.91	0.47	0.46	TDCR	non-diffusive	01 Nov 2019
Supplementary results provided, not used to determine the final result						
CIEMAT_suppl	100.4	1.05	1.05	C/N [1]	non-diffusive	19 Dec 2019
NRC_suppl	101.3	0.45	0.44	TDCR [2]	diffusive	26 Nov 2019
SMU_suppl	101.24	0.5	0.49	TDCR [2]	non-diffusive	01 Nov 2019

[1] Wallac 1220 Quantulus counter was used   [2] Hidex 300 SL - Metro counter was used

Table 3. Statistical characteristics of the ^{55}Fe activity concentration from diffusive vials.

Quantity	Activity concentration (kBq/g)	Standard uncertainty (kBq/g)	Relative standard uncertainty (%)
Arithmetic mean	101.61	0.45	0.44
Weighted mean	101.46	0.15	0.15
Median	101.48	0.13	0.13
Power-moderated mean (PMM_{sol})	101.49	0.23	0.23

The dispersion of the reported relative standard uncertainties of the solution measurements is presented in Fig. 3.

The results of activity of ready-to-measure sources reported by individual laboratories and the results obtained at POLATOM using the Fe55_6 code, for analyzing all the data, are presented in Table 4. The characteristics of the distribution of the results are reported in Table 5. One can observe (Figure 2) that all the results are compatible within the reported uncertainties and that the values of the arithmetic mean, the weighted mean, the power-moderated mean (PMM_{r-t-m}) and the median are consistent (Table 5).

Table 4. Final results for the ^{55}Fe activity concentration on the reference date of 19 October 2019 obtained using the ready-to-measure sources

Laboratory	Reported by laboratory			Raw results calculated using Fe55_6 code		
	Activity concentration (kBq/g)	Standard uncert. (kBq/g)	Relative standard uncert. (%)	Activity concentration (kBq/g)	Standard uncert. (kBq/g)	Relative standard uncert. (%)
BIPM [1]	101.86	0.70	0.68	101.86	0.78	0.68
CIEMAT	101.64	0.91	0.90	-	-	-
ENEA-INMRI	101.91	0.70	0.69	102.08	0.70	0.69
KRISS	101.27	0.78	0.77	101.31	0.78	0.77
LNE-LNHB	102.13	0.43	0.42	101.96	0.43	0.42
NIM	101.86	0.46	0.45	101.7	0.46	0.45
NMIJ	101.64	0.54	0.53	101.7	0.54	0.53
NMISA	-	-	-	102.3	0.63	0.62
NRC [2]	102.93	1.79	1.74	-	-	-
POLATOM	101.16	0.46	0.46	101.76	0.46	0.46
PTB	102.04	0.51	0.50	102.44	0.51	0.50
SMU	103.23	0.45	0.44	102.76	0.45	0.44
Supplementary results provided, not used by laboratory to determine the final result						
NRC_suppl [3]	101.97	0.44	0.44	101.65	0.44	0.44
SMU_suppl [3]	101.86	0.49	0.49	101.49	0.49	0.49

[1] The BIPM result is not normally intended to be a standardization but a parameter which is proportional to the activity. The result is included as a test of the new BIPM instrument.
[2] Wallac 1410 counter was used   [3] Hidex 300 SL - Metro counter was used

Table 5. Statistical characteristics of the ^{55}Fe activity concentration from the ready-to-measure sources

Quantity	Reported by laboratory			Raw sources results calculated using Fe55_6 code		
	Activity concentration (kBq/g)	Standard uncert. (kBq/g)	Relative standard uncert. (%)	Activity concentration (kBq/g)	Standard uncert. (kBq/g)	Relative standard uncert. (%)
Arithmetic mean	101.97	0.63	0.61	102.04	0. 49	0.48
Weighted mean	101.98	0.19	0.19	102.00	0.17	0.17
Median	101.86	0.13	0.13	101.91	0.10	0.10
Power-moderated mean ($PMM_{r\text{-}t\text{-}m}$)	101.96	0.19	0.19	102.00	0.17	0.17

8 Degrees of equivalence

The degrees of equivalence (D_{sol} and D_{r-t-m}), determined from the reference value (PMM_{sol} or PMM_{r-t-m}), are presented in Figure 4 and Table 6.

Table 6. Degrees of equivalence of the solution and ready-to-measure sources measurement results.

Laboratory	Solution			Ready-to-measure sources		
	D_{sol} (kBq/g)	U (kBq/g)	$|D_{sol}|/U$	D_{r-t-m} (kBq/g)	U (kBq/g)	$|D_{r-t-m}|/U$
BIPM	-1.70	1.24	1.37	-0.10	1.39	0.07
CIEMAT	-0.30	1.86	0.16	-0.32	1.83	0.18
ENEA-INMRI	-1.39	1.41	0.98	-0.06	1.40	0.04
KRISS	-3.02	1.60	1.89	-0.69	1.55	0.44
LNE-LNHB	-0.26	0.84	0.31	0.17	0.87	0.19
NIM	-1.15	0.92	1.25	-0.10	0.92	0.11
NMIJ	-1.09	1.20	0.91	-0.32	1.08	0.30
NMISA	0.69	1.28	0.54	-	-	-
NRC	0.61	3.20	0.19	0.97	3.58	0.27
POLATOM	-0.22	0.78	0.28	-0.80	0.92	0.86
PTB	0.20	1.02	0.20	0.08	1.02	0.08
SMU	0.42	0.94	0.45	1.26	0.90	1.41

The value D_{sol} is defined as $D_{sol} = A_{sol} - PMM_{sol}$ with A_{sol} being the activity concentration reported by the laboratory (Table 2). The value D_{r-t-m} is defined as $D_{r-t-m} = A_{r-t-m} - PMM_{r-t-m}$ with A_{r-t-m} being the activity concentration of ready-to-measure sources reported by the laboratory (Table 4). U denotes two standard uncertainties of the laboratory result.

9 Conclusions

The CCRI(II)-K2.Fe-55.2019 comparison of an ^{55}Fe solution was completed successfully with the participation of twelve laboratories. The key comparison reference value taken as the power-moderated mean from the diffusive vials results, PMM_{sol}, was of **101.49(23) kBq/g** at the reference date of **19 October 2019**. The results were in good agreement except for the KRISS result which was identified as outlier.

One can see that the type of vials used by participants has influenced the results. The values of the activity concentration of the ^{55}Fe solution obtained by participants in non-diffusive vials was lower than in diffusive vials. The dispersion of the results in non-diffusive vials was greater than in diffusive vials.

The results of activity of the ready-to-measure sources reported by the participants were compared with results obtained at POLATOM using only the Fe55_6 code. The mean values were consistent within the measurement uncertainties and the spread between the results was about the same. The values of the activity concentration of the solution measured by the participants in diffusive vials were consistent with values obtained from the measurement of ready-to-measure sources. The use of diffusive vials for ^{55}Fe measurements in the ESIR system

at the BIPM is recommended.

After the improved calculation of fractional electron-capture probabilities by X. Mougeot [6], it seems that the ^{55}Fe half-life and the decay scheme should be reevaluated. However, the half life is of minor importance in this comparison, no greater than 0.03 %, since the participants carried out their measurements within a few months of the reference date.

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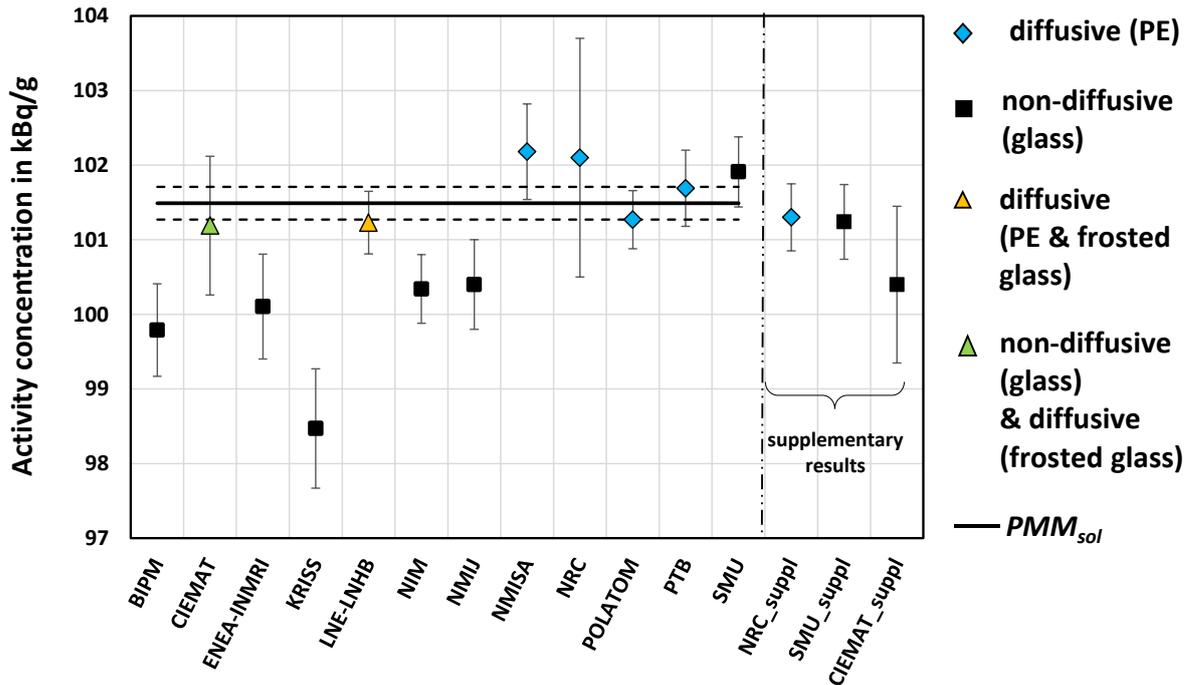
Figure 1. Final ^{55}Fe solution activity results for each laboratory. Three supplementary results were provided (as shown) but were not included in the calculation of the mean. Sources prepared in various vials are indicated using different symbols in the chart, only the results from diffusive vials are included in determining the mean (PMM_{sol}). The standard uncertainty of the PMM_{sol} value is marked (dashed lines).
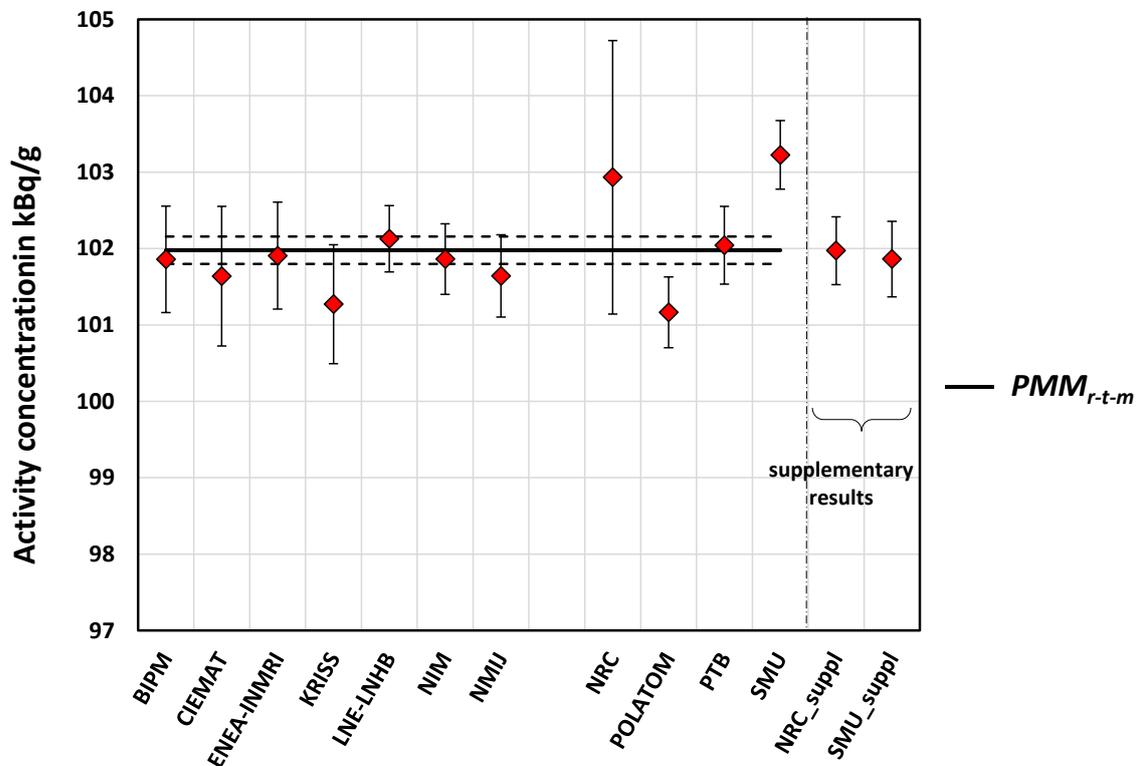
Figure 2. Final results of ready-to-measure sources. Two supplementary results provided, not used to the mean values determination, are marked. The standard uncertainty of the PMM_{sol} value is marked (dashed lines).
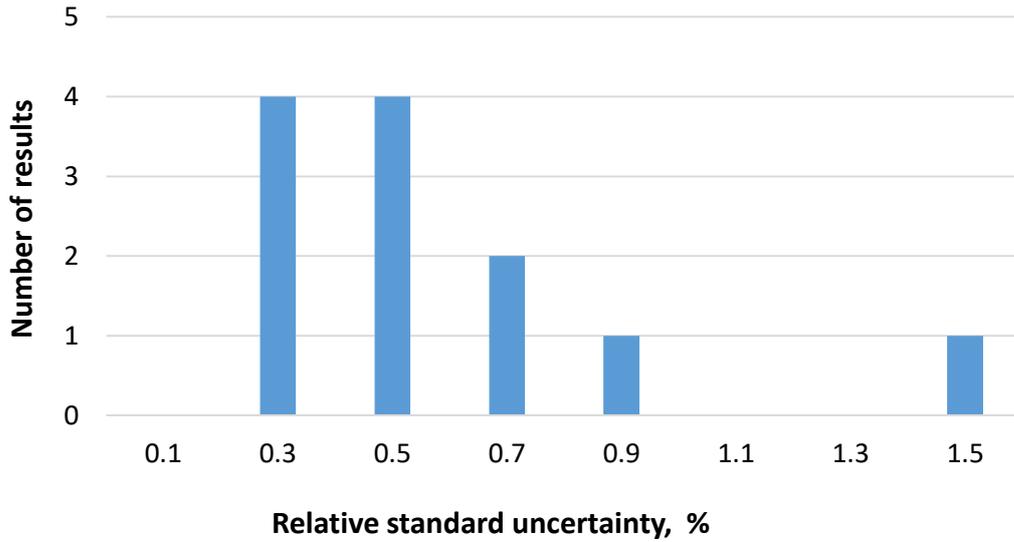
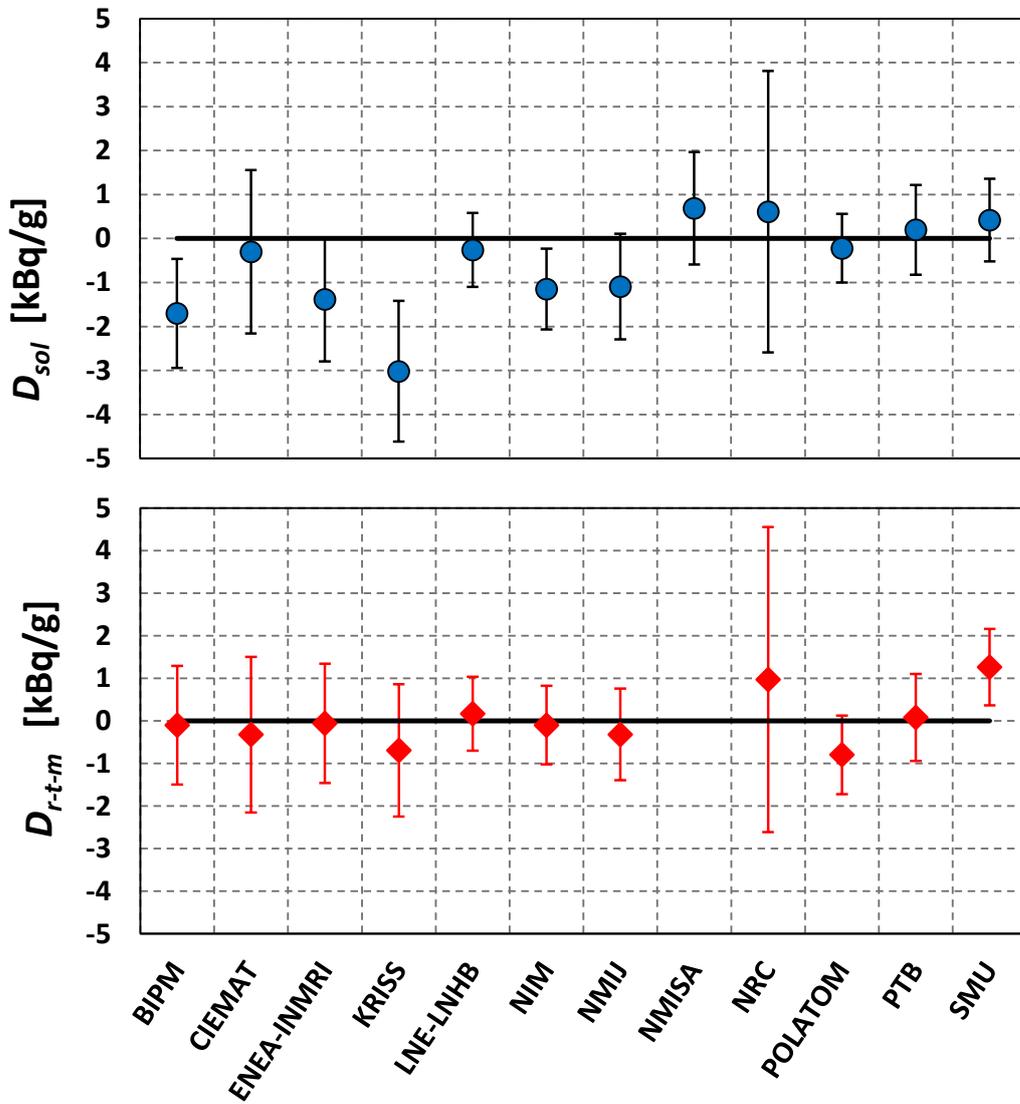**Figure 3.** Histogram of the reported relative standard uncertainties of the solution measurement.

Figure 4. D_{sol} and $D_{r\text{-}t\text{-}m}$ degrees of equivalence for each laboratory.

Appendix

Table A1. LS counters and measurement parameters

Laboratory	LS counter / measurement method	Age (y)	PMT	Coincidence resolving time (ns)	Dead-time type / base duration (μs)	Efficiency variation	Acquisition system	Scintillator	Typical TDCR value	Maximum efficiency achieved (%)	Type of LS vial	Number of sources	Typical count rate (s⁻¹)
BIPM	Locally developed / TDCR	~ 10	BURLE 8850	50	Extending / 50	Grey filters	Nano TDCR from LabZy	Ultima Gold	0.255	44.5	glass	6	2000
CIEMAT	CIEMAT Prototype / TDCR	4	BURLE 8850	40	Extending / 9 - 20	Optical filters and chemical quenching	MAC3	Ultima Gold & Hisafe 3	0.39 (UG) and 0.34 (Hisafe 3)	67 (UG) 61 (Hisafe 3)	High Performance Glass and frosted glass	16	2400 (UG) and 1900 (Hisafe 3)
ENEA-INMRI	Hidex 300SL "Metro" version / TDCR	12	ET 9102 KA	40	Non – extending / 50	none	Hidex	Ultima Gold	0.317	54.1	High Performance Glass	2	1495
KRISS	Locally developed / TDCR	16	BURLE 8850	40	Extending / 20	PMTs defocusing	MAC3	Ultima Gold	0.198	39.8	High Performance Glass	5	
LNE-LNHB	Locally developed / TDCR	>20	BURLE 8850	40 - 200	Extending / 50	none	MAC3 and Nano TDCR	UGAB	0.34	54	Polyethylene and frosted glass	7	5000 - 6000
NIM	Locally developed / TDCR	7	ET 9813B	50 - 200	Extending / 50	Grey filters	CAEN digitizer DT 5730	Ultima Gold AB	0.35	59.2	Low-potassium glass	5	1647
NMIJ	Locally developed / TDCR	20	Hamamatsu R331	20	Extending / 20	Optical filters	Analog NIM modules	Ultima Gold	0.29	50	glass	5	1000
NMISA	Locally designed and build / TDCR	~ 30	RCA 8850 & two Burle 8850	470	Non – extending / 1	none	Locally designed and build	QuickSafe A + HCl	0.5	21	Polyethylene	4	1500

Table A1. Continued

NRC	Wallac 1410 / CIEMAT/NIST	29	Hama-matsu R331-05	15	Non – extending / 30	Chemical quenching	Wallac 1410	Ultima Gold	-	42	Polyethylene	11	2000
POLATOM	Locally developed / TDCR	30	Burle 8850	40	Extending / 40	none	MAC3	Ultima Gold	0.2727	48.4	Polyethylene	10	2000
PTB	PTB developed TDCR-M27 & TDCR-M29 / TDCR	10	Hama-matsu R331	40 and other values	Extending / 30	Chemical quenching	MAC3 and 4KAM	Ultima Gold + 0.45 mL H_2O	0.51	74	Polyethylene	3	3900
SMU	POLATOM developed LSC-TDCRG / TDCR	1.5	ET Enter-prises 9214B	60	Extending / 30	Chemical quenching	MAC3	Ultima Gold	0.293	50.8	High Performance Glass	10	800

Table A2. Calculation methods, codes and parameters reported by the participants

Laboratory	Calculation code	Optimum *kB* value (cm/MeV)	d*E*/d*x* calculation model	Quenching formula	Lower integration bound	Asymmetry considered	Spectrum model	KCWG(II) recommended data
BIPM	Fe55_fom	0.010	ICRU37 and linear extrapolation under 100 eV	Birks	65 eV	yes	Betashape	yes
CIEMAT	PENNUC, NUR	0.012	NIST database ESTAR & Tan and Xia at low energies	Birks	0 eV	no	PENNUC & NUR	yes
ENEA-INMRI	Fe55_6	0.010	ICRU37+lin extrapolation	Birks	0 eV	yes	Fe55_6	yes
KRISS	Fe55_fom	0.010	Bethe-Bloch	Birks	60 eV	yes	Fe55_fom	no
LNE-LNHB	Fe55_fom	0.010	ICRU37 and linear extrapolation under 100 eV	Birks	0 eV	yes	Betashape & PENELOPE	yes
NIM	Fe55_fom	0.012	ICRU37 and linear extrapolation under 100 eV	Birks	60 eV	yes	KLM shell model & M-C simulation for X K-rays	yes
NMIJ	Locally developed	0.012	ICRU report 37	Birks	0 eV	yes	EADL & EPDL	yes
NMISA	Locally developed	0.009	Bethe formula	Birks	0 eV	yes	Locally implemented of the KL2L3M model	yes
NRC	MICELLE2	0.0075	Bethe and linear extrapolation	Birks	1 eV	No	MICELLE2	yes
POLATOM	MetroActivity v.1.0 & MICELLE2	0.010	Bethe-Bloch and d*E*/d*X* ~ *E* for *E* < 0.1 keV	Birks	62.457 eV	yes	MetroActivity v.1.0 & MICELLE2	yes
PTB	PTB code	0.0075	ICRU report 37 and d*E*/d*X* ~ $E^{-1.1}$ for *E* < 1 keV	Birks	0 eV	yes	Betashape	yes
SMU	Fe55_6	0.007	ICRU report 37	Birks	65 eV	yes	Fe55_6	yes

Table A3. Uncertainty budgets reported by the participants for solution measured using the TDCR method

Input quantity	BIPM Relative standard uncertainty %	ENEA-INMRI Relative standard uncertainty %	ENEA-INMRI Comments	KRISS Relative standard uncertainty %	KRISS Comments
counting statistics	0.152	0.15	repeated counting measurements	0.39	estimated from counting statistics of 5 samples
weighing	0.012	0.10	repeated weighing readings	0.10	estimated based on the information from the balance
background	0.014	0.05	repeated counting measurements		
dead/live time		0.20			
resolving time		0.20	by varying the resolving time in the range 20-100 ns		
decay data	0.291	0.35	by the decay-scheme uncertainties	0.65	estimated from decay data used for TDCR calculation
extra-/interpolation of efficiency curve		0.30	by varying TDCR parameter		
calibration factor	0.52				
decay correction	0.035	0.02	by uncertainty on half-life	0.03	estimated from the half-life uncertainty
impurities					
counting time					
kB parameter		0.15	by varying kB in the range 0.0070 - 0.0150 cm/MeV	0.19	estimated from series of activity calculation with different kB parameter values
counting model				0.15	estimated due to statistical model
counter effect		0.38	variability between HIDEX 300 SL 'Metro' version and ENEA portable TDCR counters		
PMTs asymmetry				0.14	estimated due to the unequal efficiency of the three PMTs
Combined standard uncertainty	0.62	0.70		0.81	

Table A3. Continued

Input quantity	CIEMAT			LNE-LNHB		NMIJ
	Relative standard uncertainty %	Comments		Relative standard uncertainty %	Comments	Relative standard uncertainty %
counting statistics	0.015			*	propagated by Monte Carlo method	0,17
weighing	0.1	From calibration certificate		0.1		0.05
background	0.01	Background variation at 1 sigma level		0.005		0.03
dead/live time	0.01	live time clock		0.01		0.05
resolving time	0.05			0.08		
decay data	0.3	From fractional EC probabilities		*	propagated by Monte Carlo method	0.42
decay correction	0.01			*	propagated by Monte Carlo method	0.02
impurities						
counting time				0.001		0.01
kB parameter	0.1	Efficiency variation depending on the kB selected		*	propagated by Monte Carlo method	0.13
counting model	0.7	Atomic rearrangement Monte Carlo model				0.35
PMTs asymmetry						
TDCR determination	0.34	Effect of TDCR determination in counting efficiency				
Eff. calculation	0.3	From NUR data processing following the PENNUC Monte Carlo model				
Scintillation composition	0.2	From data with 2 scintillation cocktails				
Sources dispersion				0.18		
Sum of uncertainties marked with *				0.35		
Combined standard uncertainty	0.92			0.41		0.59

Table A3. Continued

Input quantity	NIM			NMISA	
	Relative standard uncertainty %	Comments		Relative standard uncertainty %	Comments
counting statistics	0.11	Relative standard deviation of the mean of 5 samples, including the source dispersion.		0.06	Standard deviation of the mean using 20 values
weighing	0.1	From the calibration certificate		0.03	Weighing procedure, balance calibration
background	<0.01			0.02	BG increased by SQRT(BG) for singles, doubles and triple coincidence BG counts, 0.2 for single BG counts. 2.6 for double coincidence BG counts, 3.3 for triple coincidence BG counts
dead/live time	<0.01	The live time is calculated based on list mode data by codes from NIM		0.008	Increased dead time by 5 %
resolving time	0.19	comparing the results when the resolution time was 50 ns and 200 ns		0.02	Increased resolving time by 2 %
decay data	0.08	evaluated using Monte Carlo method		0.1	recommendations from POLATOM and parameters uncertainties
decay correction	<0.01	Uncertainty on decay correction to reference date		0.006	Using half-life uncertainty from Table of Radionuclides, Monographie BIPM-5, vol 3 , decay correction to reference date
impurities				0.0057	24 h measurement on HPGe, impurity ratio = 0.0057 (0.0008)
adsorption				0.11	Adsorption to counting vials. Count rate after 9 rinsing's with LS relative to count rate if no rinsing's were done
counting time	<0.01	Using 500 MS/s ADC sampling, the time stamp precision is 2 ns		0.001	Relative difference between expected number of counts in 300 s from a calibrated oscillator and actual number of counts measured, combined with uncertainty of the oscillator certificate
kB parameter	0.14	Comparing the difference in the results when kB value changes from 0.007 to 0.015 cm·MeV^{-1}		0.09	varied kB by 0.001
counting model	0.35	Statistical model			
after-pulsing				0.6	set afterpulsing in the triple coincidence channel to zero, half-range/sqrt(3)
Combined standard uncertainty	0.45			0.63	

Table A3. Continued

Input quantity	POLATOM Relative standard uncertainty %	POLATOM Comments	PTB Relative standard uncertainty %	PTB Comments	SMU (TDCRG) Relative standard uncertainty %
counting statistics	0.22		0.03		0.17
weighing	0.1		0.02		0.05
background	0.01		0.03		0.092
dead/live time			0.03		0.063
resolving time					0.065
decay data			0.35	e.g. fractional EC probabilities	0.2
extra-/interpolation of efficiency curve					0.1
calibration factor					0.2
decay correction	0.02		0.01		0.05
impurities			0.05	Co-60 detected	0.001
adsorption			0.05		0.05
counting time	0.001		0.01		0.01
kB parameter	<0.001	negligible impact for monoenergetical nuclide	0.16	and ionization quenching function	0.18
counting model	0.3		0.3		0.2
PMTs asymmetry			0.05	asymmetry is taken into account	
Combined standard uncertainty	0.39		0.50		0.46

Table A4. Uncertainty budgets reported by the participants for solution measured using the CIEMAT/NIST method

Input quantity	Relative standard uncertainty %	Comments
	NRC (Wallac)	
counting statistics	0.3	standard deviation of 11 LSC vials counted 10 times each
weighing	0.03	calibration of balances
background	0.01	standard deviation of blanks counted 6 times each for 1000s along with samples
dead/live time		
resolving time		
decay data	0.12	standard deviation of results having nuclear data varied according to uncertainties and propagated through MICELLE2 calculations of efficiencies
tracer	1.5	1% uncertainty on NIST traceable standard propagated through MC calculation of efficiency.
extra-/interpolation of efficiency curve	0.23	Interpolation method for determination of efficiency curves. (Lin.Interp.Fit:linear,quad,cubic)
decay correction	0.01	uncertainty on decay correction due to half-life uncertainty
impurities	0.002	application of impurity correction estimated by γ-spectrometer
adsorption	0.01	uncertainty on adsorption correction. Crushed glass was measured in an LSC cocktail repeatedly (10 times) to estimate the activity of the count rate above background.
counting time		
kB parameter		
MICELLE model corrections	0.2	spread of results of efficiencies in MICELLE2 corrections applied/not applied.
buoyancy correction	0.1	uncertainty of correction factor due to environmental conditions
Combined standard uncertainty	1.57	

Table A5. Uncertainty budgets reported by the participants for ready-to-measure sources measured using the TDCR method

Input quantity	BIPM — Relative standard uncertainty %	ENEA-INMRI — Relative standard uncertainty %	ENEA-INMRI — Comments	KRISS — Relative standard uncertainty %	KRISS — Comments
counting statistics *)	0.15	0.11		0.30	
background	0.014				
dead/live time		0.20			
resolving time		0.20	by varying the resolving time in the range 20-100 ns		
decay data	0.29	0.35		0.65	from decay data used for TDCR calculation
extra-/interpolation of efficiency curve		0.30	by varying TDCR parameter		
calibration factor	0.6 **)				
decay correction	0.034	0.02	by uncertainty on half-life	0.03	from the half-life uncertainty
impurities					
counting time					
kB parameter		0.15	by varying kB in the range 0.0070 - 0.0150 cm/MeV	0.19	from series of activity calculation with different kB parameter values
counting model				0.15	estimated uncertainty due to statistical model
Counter effect		0.38	variability between HIDEX 300 SL 'Metro' version and ENEA portable TDCR counters		
PMTs asymmetry				0.14	
Combined standard uncertainty	0.69	0.69		0.77	

*) The value estimated at POLATOM using the masses of ready-to-measure sources and the activity values reported.

**) Mean value estimated at POLATOM using values reported for various ready-to-measure sources.

Table A5. Continued

Input quantity	CIEMAT			LNE-LNHB	
	Relative standard uncertainty %	**Comments**		**Relative standard uncertainty %**	**Comments**
counting statistics *)	0.10			0.20	
background	0.02	Background variation at 1 sigma level		0.02	Gaussian distribution of the input parameters
dead/live time	0.01	live time clock		0.01	
resolving time	0.05			0.01	
pile-up		negligible for the counting rates measured		0.01	
decay data	0.3	From fractional EC probabilities		0.1	
extra-/inter-polation of efficiency curve					
calibration factor					
decay correction	0.01			0.08	
impurities					
counting time				0.01	
kB parameter	0.1	Efficiency variation depending on the *kB* selected		0.35	Gaussian distribution; propagated by Monte Carlo method
counting model	0.7	Atomic rearrangement Monte Carlo model			
PMTs asymmetry					
TDCR determination	0.34	Effect of TDCR determination in counting efficiency			
Eff. Calculation	0.3	From NUR data processing following the PENNUC Monte Carlo model			
Combined standard uncertainty	0.90			0.42	

*) The value estimated at POLATOM using the masses of ready-to-measure sources and the activity values reported.

Table A5. Continued

Input quantity	NIM Relative standard uncertainty %	NMIJ Relative standard uncertainty %	POLATOM Relative standard uncertainty %	PTB Relative standard uncertainty %	SMU (TDCRG) Relative standard uncertainty %
counting statistics *)	0.23	0.13	0.35	0.10	0.24
background	<0.01	0.01	0.01		0.092
dead/live time	<0.01	0.09			0.063
resolving time	0.07				0.065
decay data	0.08	0.39			0.2
extra-/inter-polation of efficiency curve					
calibration factor					
decay correction	<0.01	0.01			0.05
impurities					
counting time	<0.01	0.01	0.001		0.01
kB parameter	0.14	0.14			0.18
counting model	0.35	0.29	0.3		0.2
PMTs asymmetry					
Combined standard uncertainty	0.46	0.53	0.46	0.5	0.44

*) The value estimated at POLATOM using the masses of ready-to-measure sources and the activity values reported..

Table A6. Uncertainty budgets reported by the participants for ready-to-measure sources measured using the CIEMAT/NIST method

| Input quantity | NRC (HIDEX) | |
	Relative standard uncertainty %	Comments
counting statistics *)	0.23	
background	0.002	variation of 2.5 % of background
dead/live time		
resolving time		
decay data	0.12	standard deviation of results having nuclear data varied according to uncertainties and propagated through MICELLE2 calculations of efficiencies
extra-/inter-polation of efficiency curve	0.81	interpolation method for determination of efficiency curves. (Lin.Interp.Fit:linear,quad,cubic)
calibration factor		
decay correction	0.01	uncertainty on decay correction due to half-life uncertainty
impurities	0.002	application of correction factor
counting time		
kB parameter		
MICELLE model corrections	0.2	spread of results of efficiencies in MICELLE2 corrections applied / not applied
tracer	1.5	1% uncertainty on NIST traceable standard propagated through MC calculation of efficiency
Combined standard uncertainty	1.74	

*) The value estimated at POLATOM using the masses of ready-to-measure sources and the activity values reported..