European Commission

JRC TECHNICAL REPORT

Status of JRC reference measurements of radioactivity to realise the becquerel

Primary standardisation of activity and determination of decay properties

Pommé S.

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Joint Research Centre

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Acknowledgements

This work is performed in the frame of the Article 8 of the Euratom Treaty. Primary standardisation of activity at the highest level of accuracy involves the commitment of skilled physicists, chemists and engineers to the art of radionuclide metrology. JRC's participation to the key comparisons referred to in this report involved the efforts of Dr. S. Pommé, Dr. T. Altzitzoglou, R. Van Ammel, and G. Sibbens. We thank our colleagues of the radionuclide metrology sector of the JRC at Geel, for creating the stimulating environment in which this work could thrive.

Abstract

The Radionuclide Metrology team of JRC performs reference measurements of activity and nuclear decay characteristics in support of the common measurement system for radioactivity, as stipulated in Article 8 of the Euratom Treaty. Primary standardisations of activity performed at the JRC and National Metrology Institutes (NMI) are used at a global scale to establish radioactivity measurements in a traceable manner to the SI unit becquerel. To ensure equivalence of national standards, the NMIs and the JRC participate in **key comparisons**[1] of their primary standardisation measurements of a common mononuclidic solution. The Bureau International des Poids et Mesures (BIPM) in Sèvres (F) issues a report in which the Key Comparison Reference Value (KCRV) is calculated from a mean of the laboratory results, as well as a Degree of Equivalence (DoE) of each participating laboratory to the KCRV. Thus, the SI unit becquerel is established and international equivalence is demonstrated. Recently, the BIPM published final reports on various key comparisons, which validates standardisation work performed earlier at the JRC and other NMIs. This report gives an overview the corresponding key comparison results.

[1] https://www.bipm.org/en/cipm-mra/key_comparisons/

1 Introduction

The *Radionuclide Metrology* team of the JRC promotes a common terminology and measurement system for radioactivity, as laid down in Article 8 of the Euratom Treaty. Its laboratory in Geel[2] is equipped with extremely performant instruments for the most precise and sensitive measurements of activity and its highly experienced scientific and technical staff have a long-standing reputation of delivering world class quality research.

The JRC is represented in the Consultative Committee for Ionising Radiation Section II, CCRI(II), and its key comparison working group which organize the realisation of the becquerel in function of the demands in society. It takes part in key comparisons of primary standardisation measurements of activity to establish the SI unit becquerel and demonstrate equivalence of national standards on a global scale[3].

This report gives a summary of the final reports issued by the Bureau International des Poids et Mesures (BIPM) on the outcome of recently published key comparisons. With its wide suite of state-of-the-art primary standardisation techniques, the JRC has produced accurate and consistent reference values which contributed to a solid definition of the SI unit becquerel for several radionuclides.

What is a primary standardisation method for activity?

The measurand of an activity measurement is the expectation value at a reference time of the number of radioactive decays per second of a particular radionuclide in a material. It is expressed in the SI-derived unit becquerel (Bq), which corresponds to 1 aperiodic event per second.

Primary standardisation of radioactivity pertains to the indirect measurement of nuclear transitions occurring per unit time. What is considered a primary method for the realization of the unit becquerel differs from one radionuclide to another, depending on how their excess energy is emitted in the form of radiation.

Ideally, a primary standardisation method for a particular radionuclide is designed in such a way that (i) its calibration is based on basic physical principles, not on other radioactivity measurements, (ii) its result is independent of the various nuclear decay data and their associated uncertainties, (ii) it is under statistical control, i.e. all main sources of uncertainties are identified and quantified, and (iv) the total uncertainty of the result is reduced to a minimum.

In practice, a method is called primary when it has a combination of above characteristics which is competitive with the best methods available for the specific radionuclide. The transparency and completeness of the uncertainty budget as well as the accuracy of the measurement result are important criteria.

[2] https://ec.europa.eu/jrc/en/research-facility/radionuclide-metrology-laboratories

[3] https://www.bipm.org/metrology/ionizing-radiation/

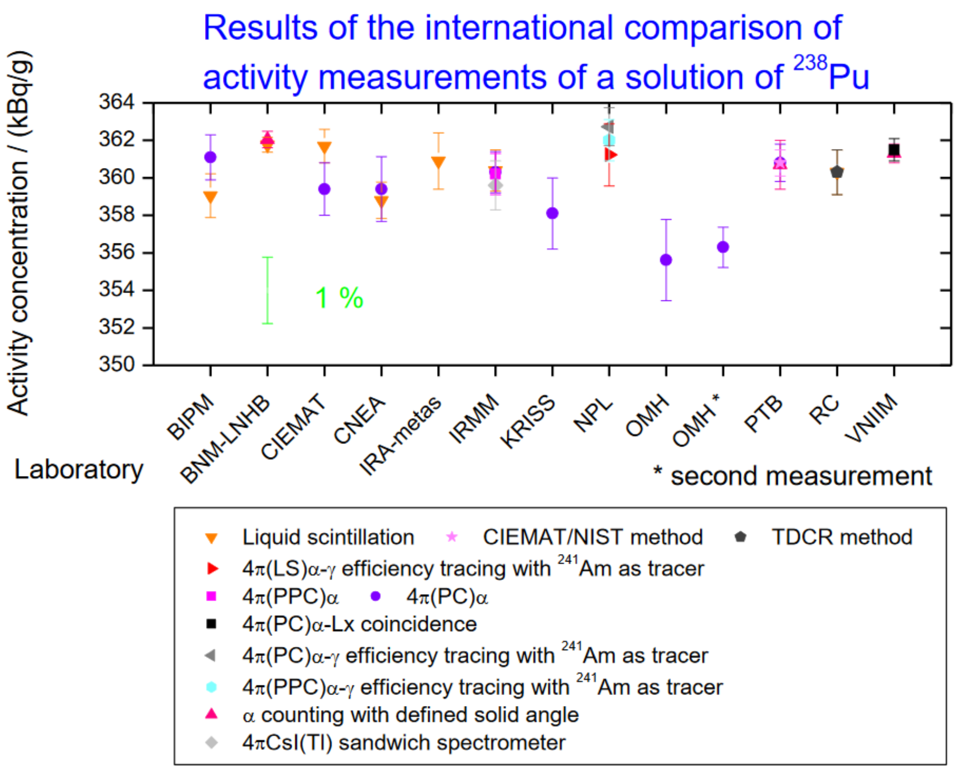
2 Primary standardisation of activity

The SI unit for activity, the becquerel, is realised through primary standardisation measurements at the NMIs (including the JRC). The KCRV (Key Comparison Reference Value) is calculated from a mean of the reference values of the participating laboratories. The degrees of equivalence (DoE) are calculated from the difference between the laboratory result and the KCRV, which preferably should not exceed the corresponding expanded uncertainty. Since 2013, the power-moderated mean is used for the KCRV (Pommé and Keightley, 2015), whereas the former KCRVs were obtained from an arithmetic mean. The official reports about key comparisons can be consulted in the Key Comparison Data Base (KCDB) (https://www.bipm.org/kcdb).

2.1 CCRI(II)-K2.Pu-238

Results published on 1 March 2018 by G. Ratel and C. Michotte in International comparison of activity measurements of a solution of ^{238}Pu, Metrologia 55 TS 06004, 2018. The JRC participated with four methods, leading to a consistent set of results. The method of counting at a defined low solid angle was upgraded by introducing autoradiography to locate the activity within the drop-deposited source, thus reducing systematic errors in the geometric correction factor.

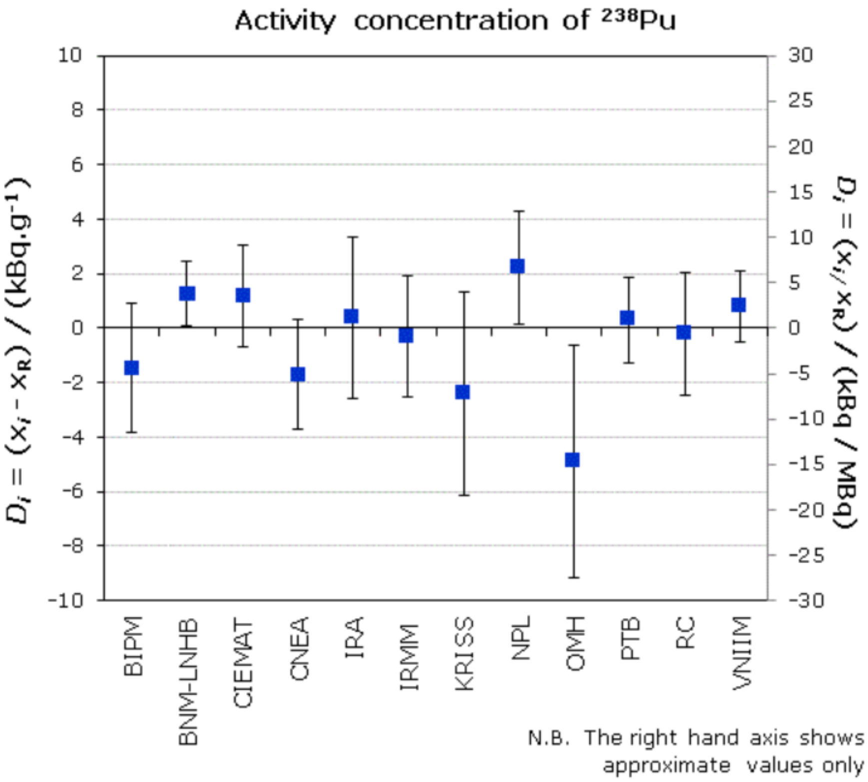
IRMM		
4π(PPC)α	360.2[1]	1.1
4π(LS)α	360.4	1.1
α(DLSA)	360.3	1.1
4πCsI(Tl)α	359.6	1.3



A weighted mean value of the four results was presented as JRC's (IRMM) reference value.

Lab i	x_i / kBq g^{-1}	u_i / kBq g^{-1}	Date of measurement
BIPM	359.1	1.2	2001
BNM-LNHB	361.77	0.40	2001
CIEMAT	361.7	0.9	2001
CNEA	358.8	1.0	2001
IRA	360.9	1.5	2001
IRMM	360.2	1.1	2001
KRISS	358.1	1.9	2001
NPL	362.7	1.0	2001
OMH	355.6	2.2	2001
PTB	360.8	0.7	2001
RC	360.3	1.1	2001
VNIIM	361.3	0.5	2001

JRC's result was in excellent agreement with the KCRV.



As a result, the degree of equivalence of JRC's reference value with the KCRV is very small, DoE=-0.3, and well within the expanded uncertainty, 2.2.

Lab i	D_i / kBq.g^{-1}	U_i
BIPM	-1.5	2.4
BNM-LNHB	1.3	1.2
CIEMAT	1.2	1.9
CNEA	-1.7	2.0
IRA	0.4	3.0
IRMM	-0.3	2.2
KRISS	-2.4	3.7
NPL	2.2	2.1
OMH	-4.9	4.3
PTB	0.3	1.6
RC	-0.2	2.2
VNIIM	0.8	1.3

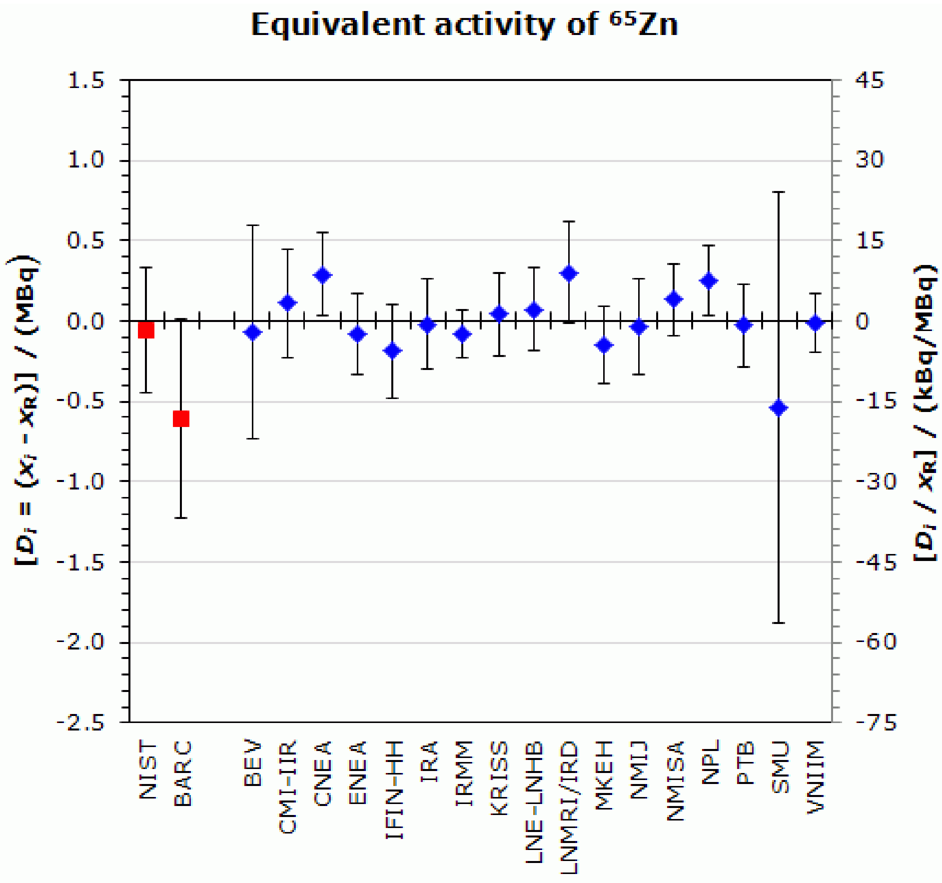
2.2 CCRI(II)-K2.Zn-65

Results were published on 09 February 2015 by C Michotte, G Ratel, S Courte, and L Joseph in BIPM comparison BIPM.RI(II)-K1.Zn-65 of activity measurements of the radionuclide 65Zn for the BARC (India) with linked results for the CCRI(II)-K2.Zn-65 comparison, Metrologia, Volume 52, Technical Supplement 06007.

IRMM	4π(PPC) β^+,e_A,x_e-γ coincidence [i] 4P-PP-MX-NA-GR-CO	55.06(10)	55.06(10) [##]
	4π(LS) CIEMAT/NIST 4P-LS-MX-00-00-CN	55.1(5)	

Lab i	x_i / kBq	u_i / kBq	Year of measurement
BEV	29670	330	2003
CMI	29850	170	2003
CNEA	30030	130	2003
ENEA-INMRI	29660	120	2003
IFIN-HH	29550	150	2003
IRA	29720	140	2003
IRMM	29661	68	2003
KRISS	29780	130	2003
LNE-LNHB	29810	130	2003
LNMRI/IRD	30040	160	2003
MKEH	29590	120	2003
NMIJ	29700	150	2003
NMISA	29870	110	2003
NPL	29990	110	2003
PTB	29710	130	2003
SMU	29200	670	2003
VNIIM	29727	87	2003

JRC realised a small uncertainty through coincidence counting and found its result close to the KCRV. Its DoE was -0.08, which is well within the expanded uncertainty of 0.15.



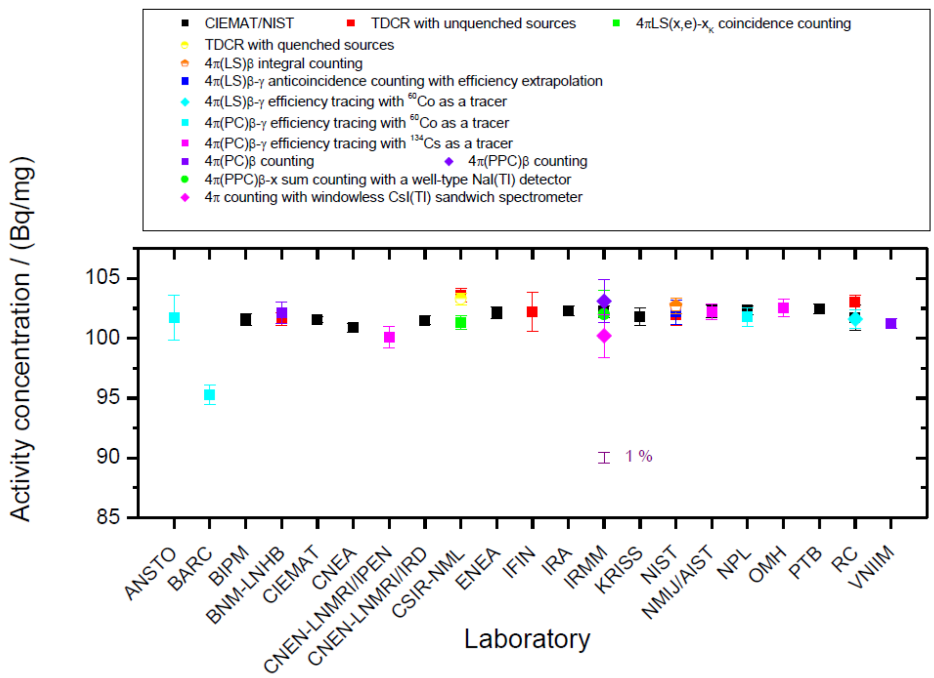
Lab i ⇩	D_i / MBq	U_i
NIST	-0.06	0.39
BARC	-0.61	0.61
BEV	-0.07	0.67
CMI	0.11	0.34
CNEA	0.29	0.26
ENEA-INMRI	-0.08	0.25
IFIN-HH	-0.19	0.29
IRA	-0.02	0.28
IRMM	-0.08	0.15
KRISS	0.04	0.26
LNE-LNHB	0.07	0.26
LNMRI/IRD	0.30	0.32
MKEH	-0.15	0.24
NMIJ	-0.04	0.30
NMISA	0.13	0.22
NPL	0.25	0.22
PTB	-0.03	0.26
SMU	-0.54	1.34
VNIIM	-0.01	0.18

2.3 CCRI(II)-K2.Tl-204

Results published on 26 June 2018 by G. Ratel in 'International comparison of activity measurements of a solution of ^{204}Tl'. The JRC participated with 4 primary standardisation methods. The results were consistent, although one method, liquid scintillation counting by CIEMAT/NIST was more precise than the other three, as it suffered less from self-absorption of the emitted low-energy beta particle. Nevertheless, the other methods relying on solid sources were not biased owing to sufficient diluting of the solution and fast drier using the JRC source drier.

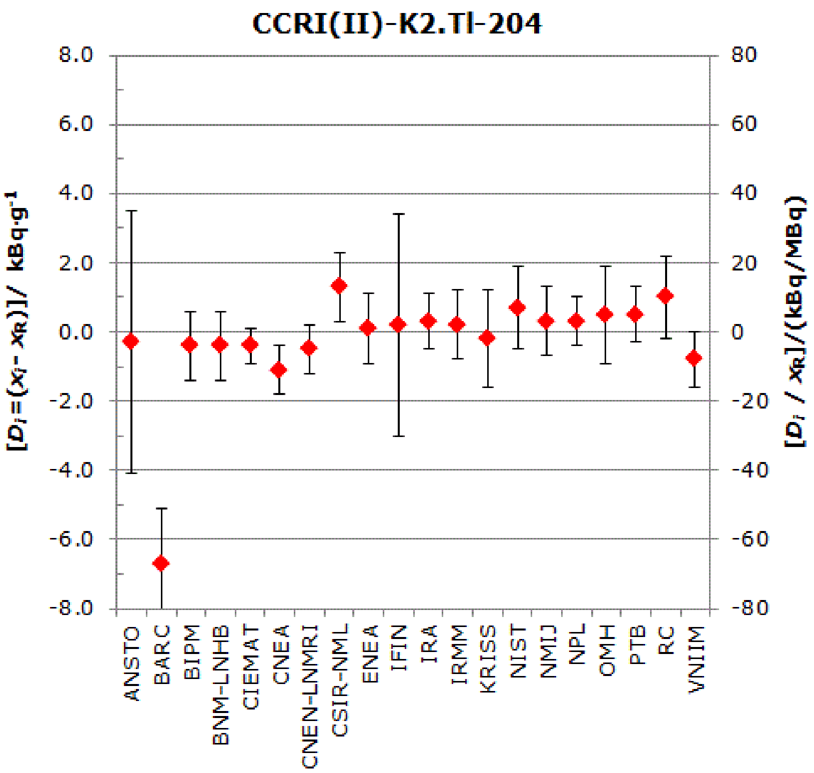
IRMM		
4π(PPC)β	103.1	1.8
4π(PPC)β-x sum counting	102.0	2.0
CIEMAT/NIST	102.2*	0.5
4πCsI(Tl)β	100.2	1.8

International comparison of activity measurements of a solution of ^{204}Tl



Each laboratory is allowed to present one reference value, which in the case of the JRC is identical to the result from LSC CIEMAT/NIST method.

Lab *i*	x_i / (kBq g^{-1})	u_i / (kBq g^{-1})	Date of measurement
ANSTO	101.7	1.9	2002
BARC	95.3	0.8	2002
BIPM	101.6	0.5	2002
BNM-LNHB	101.6	0.5	2002
CIEMAT	101.6	0.2	2002
CNEA	100.9	0.4	2002
CNEN-LNMRI	101.5	0.4	2002
CSIR-NML	103.3	0.5	2002
ENEA	102.1	0.5	2002
IFIN	102.2	1.6	2002
IRA	102.3	0.4	2002
IRMM	102.2	0.5	2002
KRISS	101.8	0.7	2002
NIST	102.7	0.6	2002
NMIJ	102.3	0.5	2002
NPL	102.3	0.4	2002
OMH	102.5	0.7	2002
PTB	102.5	0.4	2002
RC	103.0	0.6	2002
VNIIM	101.2	0.4	2002



The result of the JRC is very close to the KCRV, and consequently the DoE=0.2 is well within the expanded uncertainty U=1.0.

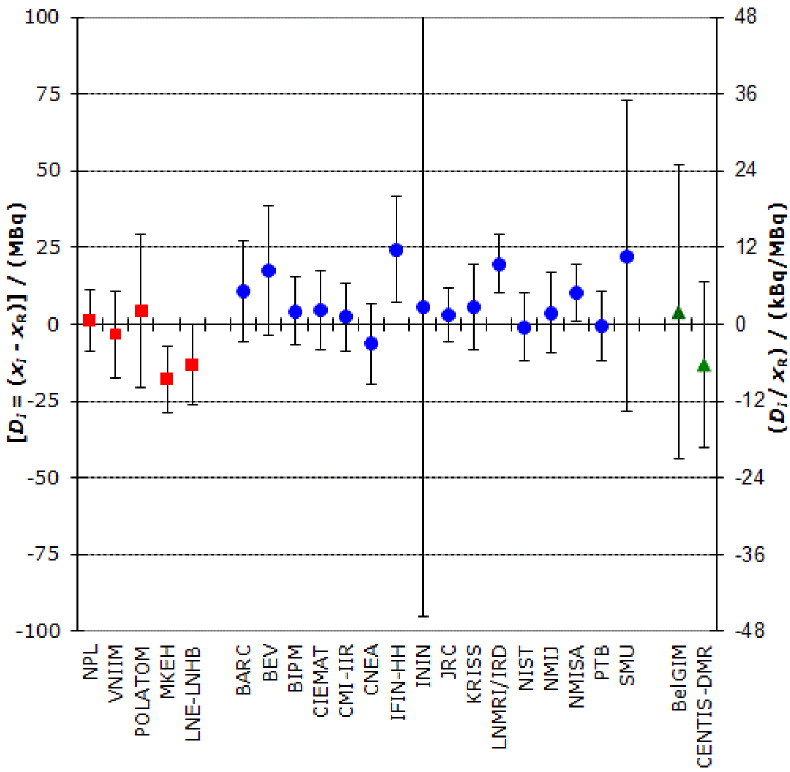
Lab *i*	D_i	U_i
	/ (kBq g^{-1})	
ANSTO	-0.3	3.8
BARC	-6.7	1.6
BIPM	-0.4	1.0
BNM-LNHB	-0.4	1.0
CIEMAT	-0.4	0.5
CNEA	-1.1	0.7
CNEN-LNMRI	-0.5	0.7
CSIR-NML	1.3	1.0
ENEA	0.1	1.0
IFIN	0.2	3.2
IRA	0.3	0.8
IRMM	0.2	1.0
KRISS	-0.2	1.4
NIST	0.7	1.2
NMIJ	0.3	1.0
NPL	0.3	0.7
OMH	0.5	1.4
PTB	0.5	0.8
RC	1.0	1.2
VNIIM	-0.8	0.8

2.4 CCRI(II)-K2.Am-241

Results were published on 1 Nov 2007 by G Ratel, C Michotte, L Johansson, S Judge and I A Kharitonov in Update of the BIPM comparison BIPM.RI(II)-K1.Am-241 of activity measurements of the radionuclide 241Am to include the 2006 VNIIM result, links for the 2003 international comparison CCRI(II)-K2.Am-241 and links for the 2006 regional comparison COOMET.RI(II)-K2.Am-241, in Metrologia, Volume 44, Technical Supplement 06007.

The JRC measured the activity within an accuracy of 0.06%, which is arguably the highest accuracy ever obtained. An additional uncertainty was added when making the link with the SIR, using the ampoule of the organising NMI of the key comparison.

Lab *i*	x_i / MBq	u_i / MBq	Year of measurement
BARC	2066.6	7.7	2003
BEV	2073	10	2003
BIPM	2060.0	4.8	2003
CIEMAT	2060.3	5.8	2003
CMI-IIR	2058.1	4.7	2003
CNEA	2049.5	6.0	2003
IFIN-HH	2080.1	8.1	2003
ININ	2061	50	2003
JRC	2058.8	3.3	2003
KRISS	2061.2	6.4	2003
LNMRI/IRD	2075.6	3.9	2003
NIST	2055.0	4.8	2003
NMIJ	2059.5	6.0	2003
NMISA	2066.1	3.6	2003
PTB	2055.2	4.8	2003
SMU	2078	25	2003



The red entries refer to the international reference system (SIR) based on standardised ampoules being measured in ionisation chambers at the BIPM, whereas the blue entries refer to a linked key comparison.

Lab *i*	D_i	U_i
	/ MBq	
NPL	1.0	10.0
VNIIM	-3.0	14.0
POLATOM	4.0	25.0
MKEH	-18.0	11.0
LNE-LNHB	-13.0	13.0
BARC	11.0	16.0
BEV	17.0	21.0
BIPM	4.0	11.0
CIEMAT	5.0	13.0
CMI-IIR	2.0	11.0
CNEA	-6.0	13.0
IFIN-HH	24.0	17.0
ININ	5.0	101.0
JRC	3.0	9.0
KRISS	5.0	14.0
LNMRI/IRD	20.0	10.0
NIST	-1.0	11.0
NMIJ	4.0	13.0
NMISA	10.0	9.0
PTB	-1.0	11.0
SMU	22.0	51.0
BelGIM	4.1	47.9
CENTIS-DMR	-13.1	27.2

2.5 CCRI(II)-K2.Mn-54

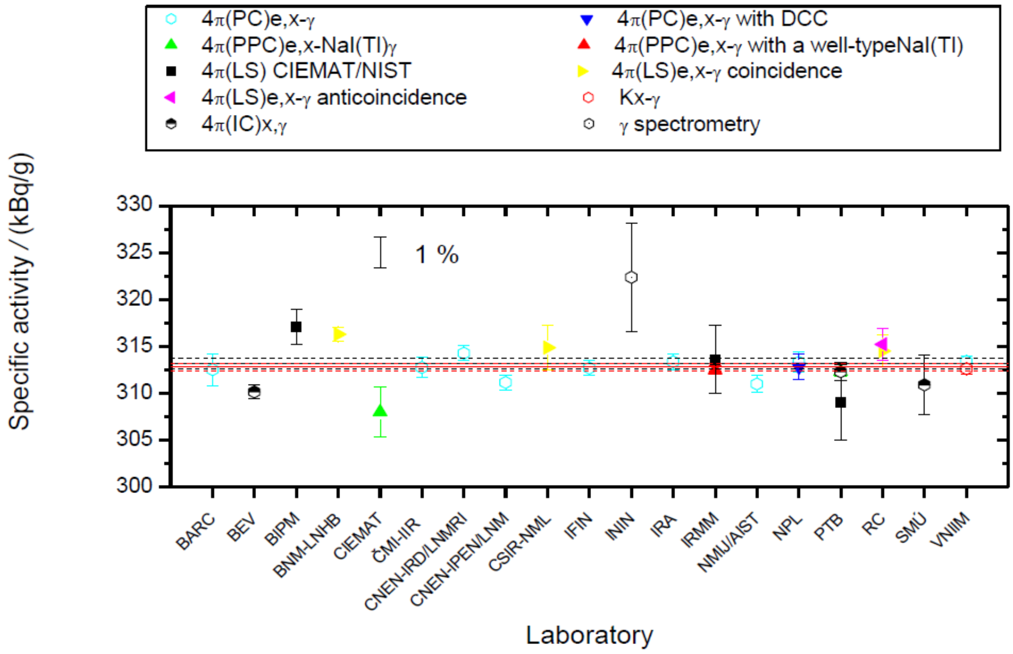
Results were published recently by G Ratel, and C Michotte in a BIPM report International comparison of activity measurements of a solution of ^{54}Mn. The JRC participated with two methods. The results were internally consistent and in good agreement with the KCRV.

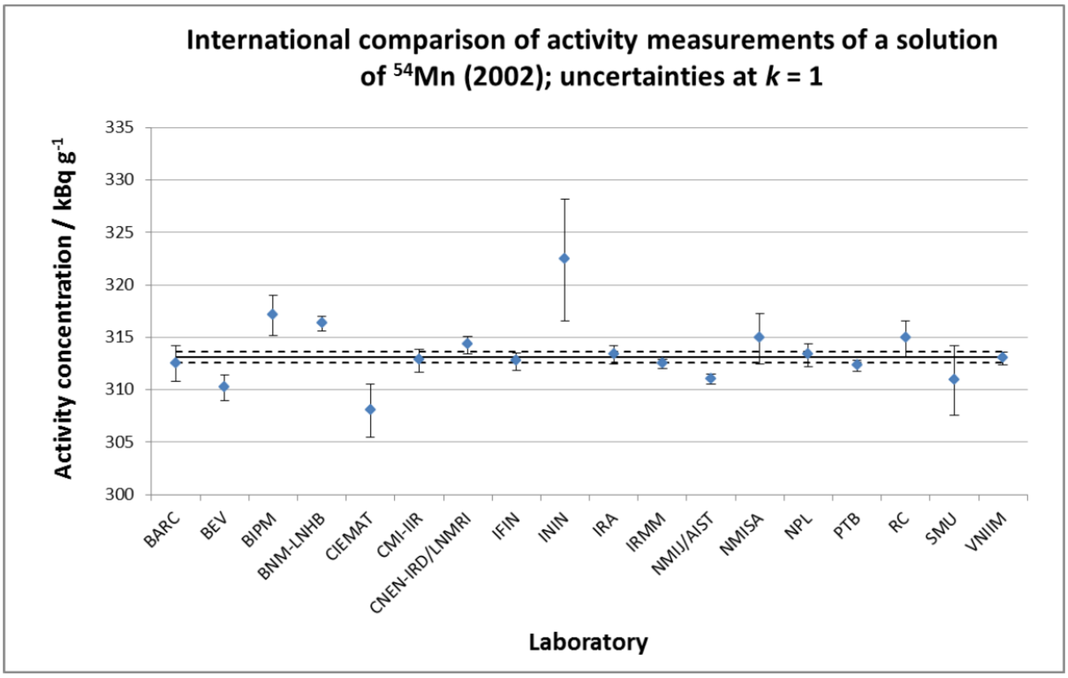
4π(PPC)e,x-γ 4P-PP-MX-NA-GR-CO	4πe,x-γ coincidence counting using a pressurized proportional counter and a well-type NaI(Tl) detector	IRMM (*P* = 0.5 MPa)
CIEMAT/NIST method 4π-LS-MX-00-00-CN	4π liquid-scintillation counting using the CIEMAT/NIST method	BIPM, IRMM, PTB

IRMM 4π(PPC)e,x-γ 4π(LS) CIEMAT/NIST	 312.45 313.6	 0.45 3.6	

Results of the international comparison of activity measurements of ^{54}Mn

July 2002

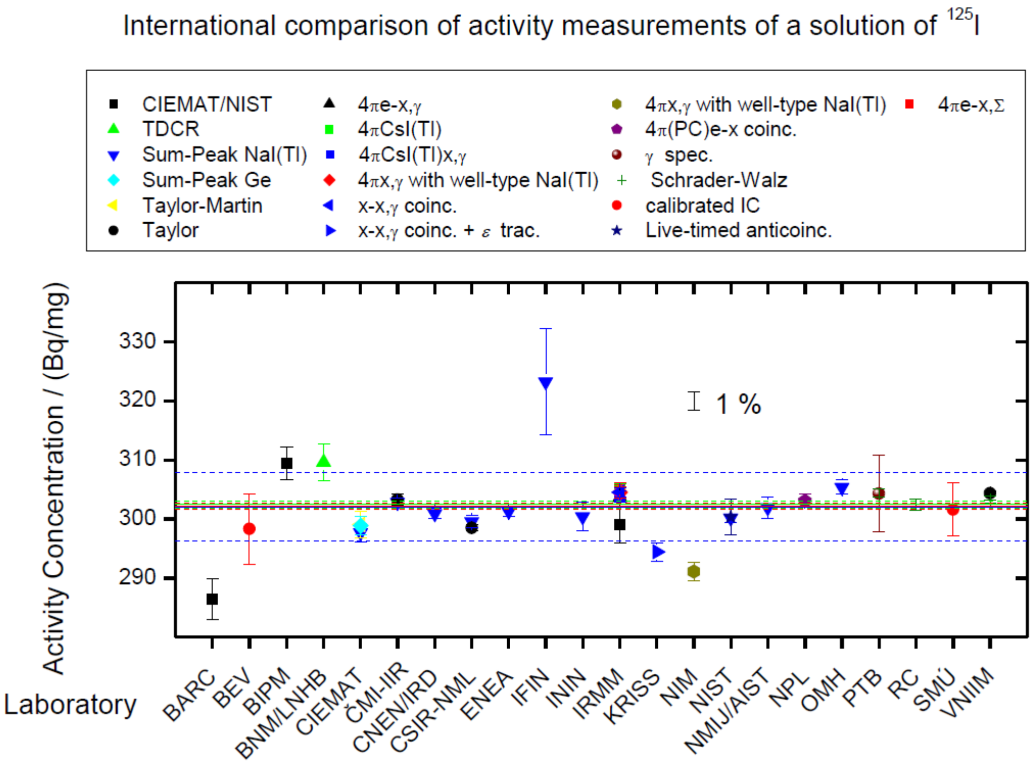


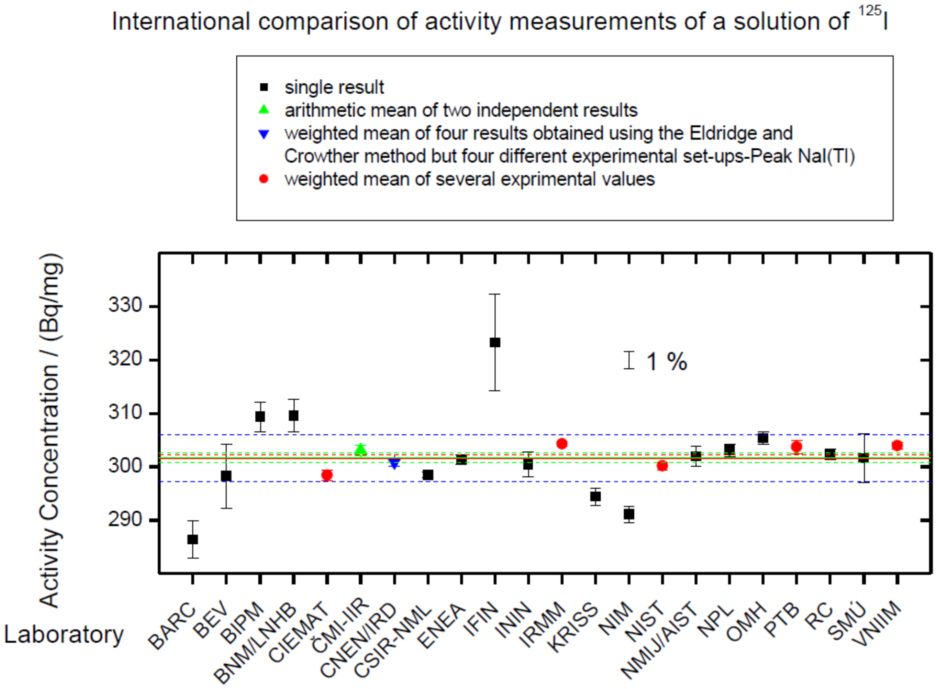


International comparison of activity measurements of a solution of ^{54}Mn (2002); uncertainties at k = 1
Activity concentration / kBq g^{-1}
335
330
325
320
315
310
305
300
BARC
BEV
BIPM
BNM-LNHB
CIEMAT
CMI-IIR
CNEN-IRD/LNMRI
IFIN
ININ
IRA
IRMM
NMIJ/AIST
NMISA
NPL
PTB
RC
SMU
VNIIM
Laboratory

2.6 CCRI(II)-K2.I-125(2)

Results were published recently by G Ratel in a BIPM report International comparison of activity measurements of a solution of ^{125}I. The JRC participated with a record number of seven methods: photon–photon coincidence counting with two NaI detectors, photon sum-peak counting in a NaI well detector and in a CsI(Tl) sandwich spectrometer, total emission counting in a windowless CsI(Tl) sandwich spectrometer, electron-X,γ coincidence counting and electron-X, γ sum counting in a pressurised proportional counter inside a NaI well detector and liquid scintillation counting with the CIEMAT/NIST method. The results were internally consistent, except for one method (LSC), and in good agreement with the KCRV. The JRC could demonstrate that the most commonly used method, LSC, gives a biased result due to incomplete modelling of the deexcitation process. Some results from other NMIs were underestimates of the true activity, due to volatility of iodine in the source, incorrect pileup correction, and modelling issues.

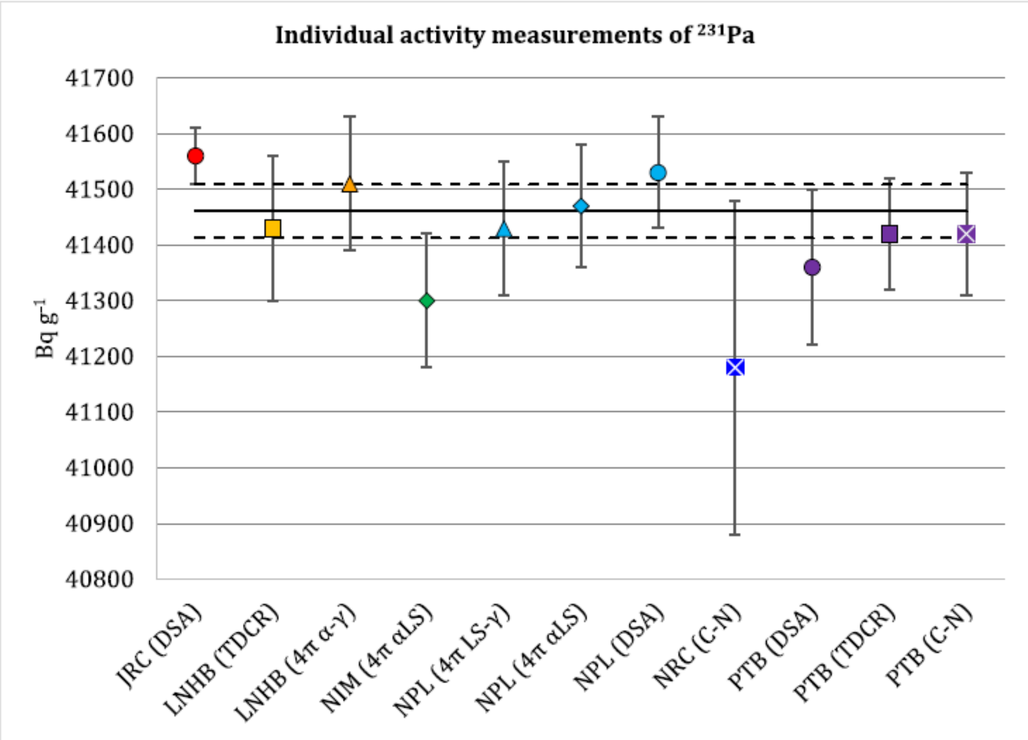


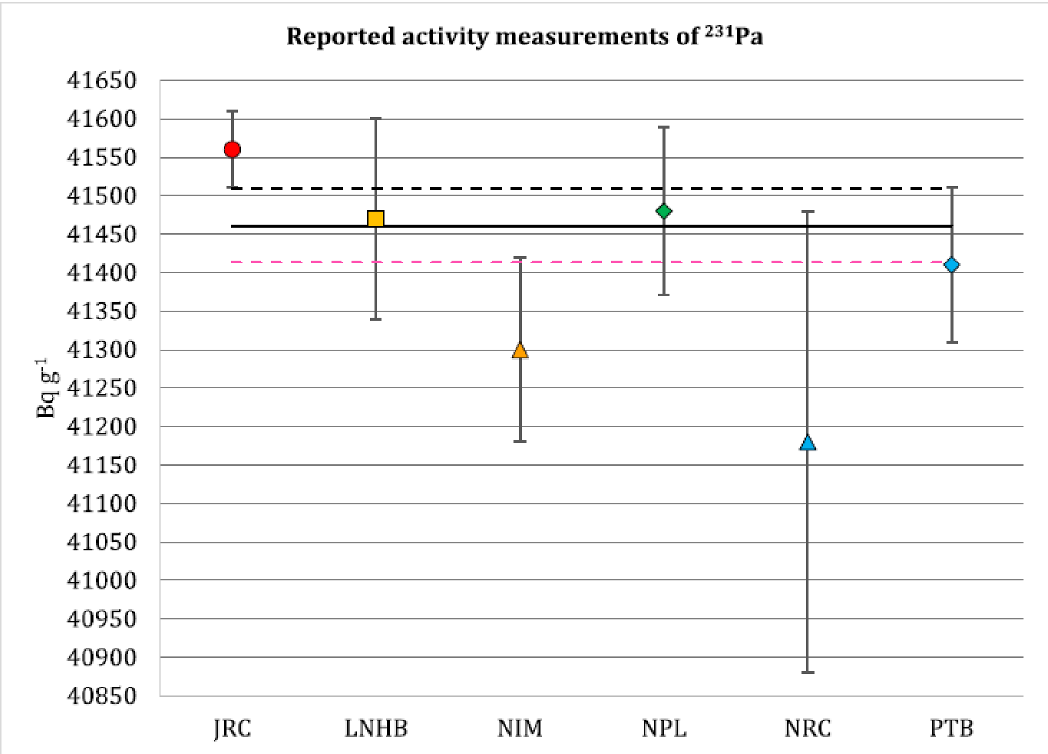


International comparison of activity measurements of a solution of 125I
single result
arithmetic mean of two independent results
weighted mean of four results obtained using the Eldridge and Crowther method but four different experimental set-ups-Peak NaI(Tl)
weighted mean of several exprimental values
Activity Concentration / (Bq/mg)
330
320
310
300
290
1 %
Laboratory
BARC
BEV
BIPM
BNM/LNHB
CIEMAT
ČMI-IIR
CNEN/IRD
CSIR-NML
ENEA
IFIN
ININ
IRMM
KRISS
NIM
NIST
NMIJ/AIST
NPL
OMH
PTB
RC
SMÚ
VNIIM

2.7 CCRI(II)-K2.Pa-231

Results were published by S. Jerome et al. in Appl. Radiat. Isot. 155 (2020) 108837 Half-life determination and comparison of activity standards of ^{231}Pa. The JRC contributed with alpha-counting at a defined solid angle. The result was the higher of the data set, but still compatible with results from other participants. A new value for the half-life could be derived from additional mass spectrometry measurements on the same ^{231}Pa in solution.



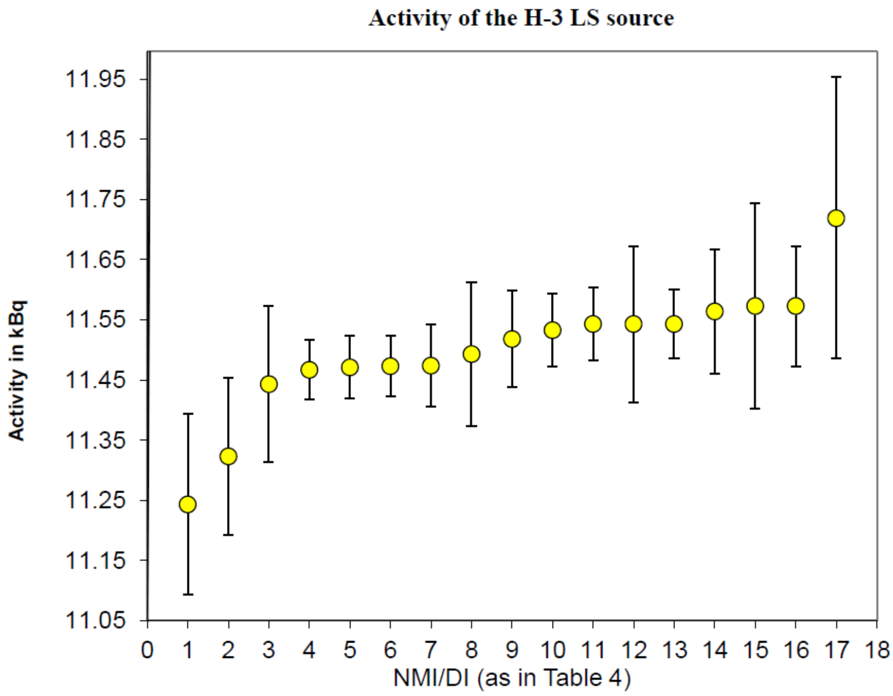


2.8 CCRI(II)-S12

A supplementary comparison was held on the data analysis of Triple-to-Double-Coincidence Ratio measuements (TDCR) supplied by the LNE-LNHB laboratory in France. The comparison results were published as "Results of the CCRI(II)-S12.H-3 supplementary comparison: Comparison of methods for the calculation of the activity and standard uncertainty of a tritiated-water source measured using the LSC-TDCR method" by Cassette, Altzitzoglou, et al. in Metrologia 56 Tech. Suppl. 06005 in 2019.

Table 4 Final results reported by the participants

Number	NMI/DI	Activity in kBq	Standard uncertainty in kBq	Relative standard uncertainty
1	KRISS	11.24	0.15	1.3%
2	NPL	11.32	0.13	1.1%
3	POLATOM	11.44	0.13	1.1%
4	ENEA	11.468	0.05	0.44%
5	VNIIM	11.468	0.05	0.44%
6	NIM	11.47	0.05	0.44%
7	IFIN-HH	11.471	0.07	0.61%
8	NIST	11.49	0.12	1.0%
9	EC-JRC	11.515	0.08	0.69%
10	LNE-LNHB	11.53	0.06	0.52%
11	FTMC	11.54	0.06	0.52%
12	CENTIS	11.54	0.13	1.1%
13	CNEA	11.54	0.06	0.52%
14	ANSTO	11.56	0.11	0.89%
15	IRA-METAS	11.57	0.17	1.5%
16	PTB	11.57	0.10	0.86%
17	NRC*	11.716	0.23	2.0%



The JRC result is realistic with respect to the calculated activity of tritium as well as the uncertainty evaluation associated with the data set and the method.

3 Conclusions

The BIPM has recently been catching up with its backlog of draft B reports on key comparisons of activity. Through the recent publication of final reports by the BIPM, the degree of equivalence of JRC's reference measurements of activity by primary standardisation techniques has been officialised. By the accuracy of its measurements in combination with a realistic uncertainty budget, the JRC has contributed to solid establishment of the becquerel for various radionuclides.

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