BIPM comparison BIPM.RI(II)-K1.I-131 of activity measurements of the radionuclide ^{131}I

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Abstract

Since 1977, fifteen national metrology institutes (NMI) and one other laboratory have submitted thirty-five samples of known activity of ^{131}I to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures. The activities ranged from about 200 kBq to 67 MBq. The degrees of equivalence between each equivalent activity measured in the SIR and the key comparison reference value (KCRV) have been calculated and the results are given in the form of a matrix for thirteen NMIs. A graphical presentation is also given. The results of this comparison have been approved by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), comparison identifier BIPM.RI(II)-K1.I-131.

1. Introduction

The SIR for activity measurements of γ-ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid (or gaseous) form. The NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of ^{226}Ra using pressurized ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity are all given in [1].

Since its inception, until 31 December 2002, the SIR has measured 835 ampoules to give 606 independent results for 62 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.I-131 key comparison.

2. Participants

Fifteen NMIs and one other laboratory have submitted thirty-five ampoules for the comparison of ^{131}I activity measurements since 1977. Some early results have been withdrawn. The laboratory details are given in Table 1. In cases where the laboratory has changed its name since the original submission, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB.

Table 1. Details of the participants in the BIPM.RI(II)-K1.I-131

Original acronym	**NMI**	**Full name**	**Country**	**Regional metrology organization**	**Date of measurement at the BIPM**
NBS	NIST	National Institute of Standards and Technology	United States	SIM	1977-05-24 1978-06-05 1979-09-03 1980-03-06 1981-03-06 1998-02-04
ASMW *	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1978-03-10 1989-10-31 1993-03-15
–	BARC	Bhabha Atomic Research Centre	India	APMP	1979-07-17 1994-06-15
NAC*	CSIR-NML	National Metrology Laboratory	South Africa	SADCMET	1980-02-11
–	NPL	National Physical Laboratory	United Kingdom	EUROMET	1980-10-08 1999-03-18
–	OMH	Országos Mérésügyi Hivatal	Hungary	EUROMET	1983-03-04 1991-11-25 1998-10-28
IER	IRA	Institut de Radiophysique Appliquée	Switzerland	EUROMET	1984-05-24 1996-09-19

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Table 1 continued. Details of the participants in the BIPM.RI(II)-K1.I-131

Original acronym	**NMI**	**Full name**	**Country**	**Regional metrology organization**	**Date of measurement at the BIPM**
PDS	P3KRBiN	Pusat Penelitian & Pengembangan Keselamatan Radiasi & Biomedika Nuklir	Indonesia	APMP	1985-06-17
ETL	NMIJ	National Metrology Institute of Japan	Japan	APMP	1986-02-05
UVVVR	CMI-IIR	Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation	Czech Republic	EUROMET	1986-10-02 2001-09-25
LMRI	BNM-LNHB	Bureau national de métrologie-Laboratoire national Henri Becquerel	France	EUROMET	1987-06-22
NIRH	–	National Institute of Radiation Hygiene	Denmark	EUROMET	1991-03-27
–	ANSTO	Australian Nuclear Science and Technology Organisation	Australia	APMP	1994-06-02
–	LNMRI	Laboratorio Nacional de Metrologia das Radiaçoes Ionizantes	Brazil	SIM	1999-11-17
–	CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnològicas	Spain	EUROMET	2002-06-26

* another laboratory in the country

3. NMI standardization methods

Each NMI that submits ampoules to the SIR has measured the activity either by a primary standardization method or by using a secondary method, for example a calibrated ionization chamber. In the latter case, the traceability of the calibration needs to be clearly identified to ensure that any correlations are taken into account.

A brief description of the standardization methods for each laboratory, the activities submitted and the relative standard uncertainties ($k = 1$) are given in Table 2. The list of acronyms used to summarize the methods is given in Appendix 3. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. When submitted by the NMIs, the uncertainty budgets are given in Appendix 1 attached to this report. Consequently, the NPL, LNMRI, CMI-IIR and the CIEMAT have provided uncertainty budgets.

The half-life used by the BIPM is 8.021 (1) days [3].

Table 2. Standardization methods of the participants for ^{131}I

NMI	Method used and acronym (see Appendix 3)	Half-life / d	Activity A_i / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
NIST	Pressurized IC 4P-IC-GR-00-00-00 calibrated by 4πβ(PC) 4P-PC-BP-00-00-00	–	7 170 7 190†	77-05-11 17 h UT	0.02	0.57
		8.02 (1)	4 648	78-05-24 12 h UT	0.01	0.63
	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1979 by 4πβ-γ coincidence 4P-PP-BP-NA-GR-CO	8.02 (1)	4 475	79-07-17 19 h UT	0.01	0.64
			4 395	80-02-21 7 h UT	0.01	0.61
			5 363 ^{a}	81-02-24 20 h UT	0.04	0.47
			19 850	98-01-27 22 h UT	0.02	0.35
PTB	Pressurized IC * 4P-IC-GR-00-00-00	–	32 160	78-03-08 0 h UT	0.01	0.15
	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	–	4 005	89-11-01 12 h UT	0.01	0.15
	Pressurized IC * 4P-IC-GR-00-00-00	–	3 719 3 708†	93-03-12 0 h UT	0.03	0.20

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Table 2 continued. Standardization methods of the participants for ^{131}I

NMI	Method used and acronym (see Appendix 3)	Half-life / d	Activity A_i / kBq	Reference date	Relative standard uncertainty × 100 by method of evaluation	
				YY-MM-DD	A	B
BARC	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	–	1 901	79-07-05 6 h 30 UT	0.19	0.43
		–	259.2	94-06-01 6 h 30 UT	0.11	0.29
CSIR-NML	4π(LS)β-γ coinc. 4P-LS-BP-NA-GR-CO	–	22 510 22 580[†]	80-02-06 10 h UT	0.03	0.50
NPL	Pressurized IC** 4P-IC-GR-00-00-00	–	6 899	80-10-06 0 h UT	0.06	0.33
	4π(PPC)-γ coinc. 4P-PP-BP-NA-GR-CO	–	3 750 3 719[†]	99-03-02 12 h UT	0.09	0.27
OMH	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	8.020 (2) [4]	3 702	83-03-08 12 h	0.02	0.63
		[3]	10 140	91-12-01 12 h UT	0.02	0.39
	4π(PC)-γ coinc. 4P-PC-BP-NA-GR-CO	[3]	9 398	98-10-27 0 h UT	0.02	0.31
IRA	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	–	3 634	84-05-16 0 h UT	0.04	0.49
	Pressurized IC* 4P-IC-GR-00-00-00	–	3 453	96-09-10 10 h UT	0.03	0.50
P3KRBiN	Pressurized IC 4P-IC-GR-00-00-00	–	2 360 2 360[†]	85-06-06 2 h UT	1.1	
NMIJ	4π (PC)β-γ coinc. 4P-PC-BP-NA-GR-CO	8.040	2 822 2 809[†]	86-02-04 12 h UT	0.08	0.26
CMI-IIR	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	8.02	20 290	86-10-01 12 h UT	0.04	0.27
	Pressurized IC* 4P-IC-GR-00-00-00	8.021 (1)	9 701	01-09-18 12 h UT	0.28	0.28
BNM-LNHB	4πβ-γ coincidence 4P-PC-BP-NA-GR-CO	8.021 (1)	2 116 2 114[†]	87-06-24 12 h UT	0.10	0.13
NIRH	Pressurized IC 4P-IC-GR-00-00-00	–	67 200	91-03-20 23 h UT	0.5	2.5
ANSTO	Pressurized IC [††] 4P-IC-GR-00-00-00	–	6 491	94-05-17 23 h UT	0.02	0.88

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Table 2 continued. Standardization methods of the participants for ^{131}I

NMI	Method used and acronym (see Appendix 3)	Half-life / d	Activity A_i / kBq	Reference date YY-MM-DD	Relative standard uncertainty × 100 by method of evaluation	
					A	B
LNMRI	Pressurized IC ††† 4P-IC-GR-00-00-00	[3]	11 610	99-11-01 0 h UT	0.18	0.43
CIEMAT	4π(PC)β-γ (NaI) coincidence 4P-PC-BP-NA-GR-CO	8.021	35 940	02-06-05 10 h UT	0.45	0.10

† two ampoules submitted

[a] corrected for the growth of ^{131}Xe^{m} which was assumed to be zero at the time of sealing the ampoule

†† calibrated by 4πβ-γ coincidence measurements of ^{131}I in1978

* traceable to 4πβ-γ coincidence measurements 4P-PC-BP-NA-GR-CO of ^{131}I (in 2001 for the CMI-IIR)

** calibrated by primary measurements of ^{131}I

††† calibrated by 4πβ-γ coincidence measurements of ^{131}I in1993.

Details regarding the solution submitted are shown in Table 3, including any impurities, when present, as identified by the laboratories. When given, the standard uncertainties on the evaluations are shown. Recently the BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [5]. The CCRI(II) agreed in 1999 [6] that this method should be followed according to the protocol described in [7] when an NMI makes such a request or when there appear to be discrepancies.

Table 3. Details of the solution of ^{131}I submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity[†]
NIST	KI and Na_2SO_3 in diluted LiOH and NaOH	LiOH: 1.3×10^{-3} NaOH: 0.05×10^{-3}	KI: 60 Na_2SO_3: 30	0.999 (2)	^{125}I : 0.3 % *
		LiOH: 0.1×10^{-3} NaOH: 0.08×10^{-3}	KI: 6 Na_2SO_3: 3	0.999 (2)	–
				0.998 (2)	–
	KI and Na_2SO_3 in diluted $LiOH.H_2O$ and NaOH	$LiOH.H_2O$: 0.05×10^{-3} NaOH: 0.01×10^{-3}	KI: 5 Na_2SO_3: 2	0.998 (2)	–
		$LiOH.H_2O$: 0.07×10^{-3} NaOH: 0.02×10^{-3}	KI: 6 Na_2SO_3: 3	0.998 (2)	–
	KI in LiOH	0.01	KI: 68	0.999	–
PTB	NaI and $Na_2S_2O_3$ in water with 0.1 % Formalin	–	NaI and $Na_2S_2O_3$: 100	1.00	< 0.01 %
	KI and $Na_2S_2O_3$ in water	–	KI : 20 $Na_2S_2O_3$: 50	0.9982	< 0.03 %
	NaI and $Na_2S_2O_3$ and 0.1 % Formalin in NaOH	1	NaI : 60 $Na_2S_2O_3$: 45	1.0	–
BARC	KI and $Na_2S_2O_3$ in water	–	KI: 200 $Na_2S_2O_3$: 60	–	–
	NaI and $Na_2S_2O_3$ in water	–	KI: 100	1	–

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Table 3 continued. Details of the solution of ^{131}I submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(μg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity[†]
CSIR-NML	KI and Na_2SO_3 in diluted NaOH	0.2 × 10^{-3}	KI: 4800 Na_2SO_3: 1600	1.007	–
NPL	KI in NaOH	0.01	KI: 100	1.0004	–
	NaI and $Na_2S_2O_3$ in diluted NaOH	0.0025	NaI : 55 $Na_2S_2O_3$: 25	1	–
OMH	KI; KIO_3 and $Na_2S_2O_3$ in water	–	KI : 50 KIO_3 : 50 $Na_2S_2O_3$: 50	–	< 0.01 %
			idem	–	–
	I in NaOH	0.01	–	–	–
IRA	KI and Na_2SO_3 in diluted LiOH	0.9 × 10^{-3}	KI : 50 Na_2SO_3 : 20	1.00	–
			KI : 20 Na_2SO_3 : 20	–	–
P3KRBiN	KI and Na_2SO_3 in diluted LiOH	0.9 × 10^{-3}	KI : 50 Na_2SO_3 : 20	0.990 (1)	–
NMIJ	NaI and Na_2SO_3 in diluted LiOH	0.9 × 10^{-3}	NaI: 50 Na_2SO_3 : 30	0.9982	–
CMI-IIR	KI and $Na_2S_2O_3$ in $NaHCO_3$	0.7 × 10^{-3}	KI : 50 $Na_2S_2O_3$: 50	–	< 0.1 %
	KI and $Na_2S_2O_3$ in water	–	KI : 50 $Na_2S_2O_3$: 50	–	< 0.1 %
BNM-LNHB	KI and $Na_2S_2O_3$ in diluted NaOH	0.5 × 10^{-3}	KI : 50 $Na_2S_2O_3$: 50	0.9983	< 2 10^{-3} %
NIRH	–	–	–	1	–
ANSTO	NaI	–	–	–	–

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Table 3 continued. Details of the solution of ^{131}I submitted

NMI	Chemical composition	Solvent conc. / (mol dm^{-3})	Carrier: conc. /(µg g^{-1})	Density /(g cm^{-3})	Relative activity of impurity[†]
LNMRI	NaCl in water	–	–	1.01	–
CIEMAT	Na_2HPO_4 in water	–	Na_2HPO_4 : 6800	–	–

[†] the ratio of the activity of the impurity to the activity of ^{131}I at the reference date
* negligible effect on the SIR measurement.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The activity measurements for ^{131}I arise from thirty-five ampoules and the SIR equivalent activity for each ampoule, A_{ei}, is given in Table 4 for each NMI, i. The dates of measurement in the SIR are given in Table 1.

As the half-life of the daughter radionuclide ^{131}Xe^{m} ($T_{1/2}$ of 11.93 d [3]) is longer than that of the parent, no equilibrium can be reached. However, the contribution of ^{131}Xe^{m} to the ionization current is negligible at any time, because of the low branching ratio and gamma-emission probability.

The relative standard uncertainties arising from the measurements in the SIR are also shown. This uncertainty is additional to that declared by the NMI for the activity measurement shown in Table 2. Although activities submitted are compared with a given source of ^{226}Ra, all the SIR results are normalized to the radium source number 5 [1].

No impurity measurements were made at the BIPM.

Measurements repeated at the BIPM after periods of up to about one month later produced the same comparison results within the SIR combined uncertainties for the OMH (1983 and 1991), IRA (1984), NMIJ, CMI-IIR (1986) and for the NIST (1998). These measurements confirm the validity of the half-life value used and the absence of impurity in these solutions.

The most recent value from the NIST contained a larger activity of ^{131}I than earlier submissions and also was measured at the BIPM closer to the reference date than had been possible previously. This results in a reduced SIR uncertainty that together with the smaller NIST uncertainty produces a much smaller combined uncertainty than the 1979 submission. Consequently, the1998 result is used in the KCRV.

No recent submission has been identified as a pilot study so the most recent result of each NMI is normally eligible for Appendix B of the MRA. However, the NIRH no longer undertakes the metrology of activity and Indonesia (P3KRBiN) has not signed the MRA, therefore none of these results is included in the KCDB.

No international or regional comparison for this radionuclide has been held to date so no linking data are identified.

Table 4. Results of SIR measurements of ^{131}I

NMI	Mass of solution m_i / g	Activity submitted A_i / kBq	N° of Ra source used	SIR A_e / kBq	Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / kBq
NIST	3.665 65 3.676 07	7 170 7 190	3	40 770 40 780	8×10^{-4}	240 240
	3.671 13	4 648	2	40 690	9×10^{-4}	260
	3.620 48	4 475	1	40 110	23×10^{-4}	270
	3.611 43	4 395	2	40 370	9×10^{-4}	250
	3.580 17	5 363	3	40 330	7×10^{-4}	190
	3.596 14	19 850	4	40 500	7×10^{-4}	140
PTB	3.682 7 (1)	32 160	5	40 391	6×10^{-4}	66
	3.511 6	4 005	3	40 533	7×10^{-4}	67
	3.641 8 3.631 0	3 719 3 708	3	40 411 40 348†	7×10^{-4}	85 85
BARC	3.599 7	1 901	1	40 440	13×10^{-4}	200
	3.585 2	259.2	1	40 360	23×10^{-4}	160
CSIR-NML	3.649 3.585 *	22 510 22 580	4	40 430 40 430†	6×10^{-4}	200 200
NPL	3.701 4	6 899	3	40 320	7×10^{-4}	140
	3.721 70 3.690 49	3 750 3 719	2	40 190 40 130†	9×10^{-4}	120 120
OMH	3.604 4	3 702	3	40 010	8×10^{-4}	260
	3.610 8	10 140	4	40 430	6×10^{-4}	160
	3.617 4	9 398	4	40 330	7×10^{-4}	130
IRA	3.584 7 (1)	3 634	3	40 310	7×10^{-4}	200
	3.363 6 (1)	3 453	2	40 260	10×10^{-4}	210
P3KRBiN	3.575 3.574	2 360 2 360	2	40 250 40 190†	9×10^{-4} 10×10^{-4}	440 440
NMIJ	3.602 6 3.586 5	2 822 2 809	3	40 320 40 320†	8×10^{-4}	110 110

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Table 4 continued. Results of SIR measurements of ^{131}I

NMI	Mass of solution m_i / g	Activity submitted A_i / kBq	N° of Ra source used	SIR A_e / kBq	Relative uncertainty from SIR	Combined uncertainty $u_{c,i}$ / kBq
CMI-IIR	3.588 0	20 290	3	40 330	6×10^{-4}	110
	3.610 4	9 701	3	40 640	7×10^{-4}	170
BNM-LNHB	3.623 94 3.621 13	2 116 2 114	3	40 316 40 300[†]	8×10^{-4} 8×10^{-4}	72 72
NIRH	3.316	67 200	5	40 200	6×10^{-4}	1000
ANSTO	3.584	6 491	2	40 390	10×10^{-4}	360
LNMRI	3.510 04	11 610	3	40 670	8×10^{-4}	190
CIEMAT	3.546 3	35 940	3	40 130	7×10^{-4}	190

[†] the mean of the two A_e values is used with an averaged uncertainty, as attributed to an individual entry [8]

* mass of active solution before dilution: 1.188 28 g and 1.192 01 g respectively.

4.1 The key comparison reference value

The key comparison reference value is derived from the unweighted mean of all the results submitted to the SIR with the following provisions:

a) only primary standardized solutions are accepted, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;

b) each NMI or other laboratory has only one result (normally the most recent result or the mean if more than one ampoule is submitted);

c) any outliers are identified using a reduced chi-squared test and, if necessary, excluded from the KCRV using the normalized error test with a test value of four;

d) exclusions must be approved by the CCRI(II).

The reduced data set used for the evaluation of the KCRVs is known as the KCRV file and is the reduced data set from the SIR mother-file. 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 only made by the CCRI(II), normally during one of its biennial meetings.

Consequently, the KCRV for ^{131}I has been identified as 40 390 (40) kBq using the most recent results from the CSIR-NML, NMIJ, BNM-LNHB, PTB, ANSTO, BARC, NIST, OMH, NPL, LNMRI, CMI-IIR and the CIEMAT and the earlier results from the IRA (1984) and the ASMW (1989).

4.2 Degrees of equivalence

Every NMI that has submitted ampoules to the SIR is entitled to have one result included in Appendix B of the KCDB as long as the NMI is a signatory or designated institute listed in the MRA. Normally, the most recent result is the one included. Any NMI may withdraw its result only if all the participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the key comparison reference value [2]. The degree of equivalence is expressed quantitatively in terms of the deviation from the key comparison reference value and the expanded uncertainty of this deviation ($k = 2$). The degree of equivalence between any pair of national measurement standards is expressed in terms of their difference and the expanded uncertainty of this difference and is independent of the choice of key comparison reference value.

4.2.1 *Comparison of a given NMI with the KCRV*

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

$$D_i = A_{ei} - \mathrm{KCRV} \tag{1}$$

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

$$U_i = 2u_{D_i}, \tag{2}$$

taking correlations into account as appropriate (see Appendix 2).

4.2.2 *Comparison of any two NMIs with each other*

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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \tag{3}$$

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

$$u_{D_{ij}}^{\;2} = u_i^{\;2} + u_j^{\;2} - \sum_k \left(f_k u_{\mathrm{corr},k}\right)_i^2 - \sum_k \left(f_k u_{\mathrm{corr},k}\right)_j^2 \tag{4}$$

and any obvious correlations in the standard uncertainties for a given component, $u_{\mathrm{corr},k,}$, between the NMIs (such as a traceable calibration) are subtracted using an appropriate correlation coefficient, f_k, as are normally those correlations coming from the SIR.

The uncertainties of the differences between the values assigned by individual NMIs and the key comparison reference value (KCRV) are not necessarily the same uncertainties that enter into the calculation of the uncertainties in the degrees of equivalence between a pair of participants. Consequently, the uncertainties in the table of degrees of equivalence cannot be generated from the column in the table that gives the uncertainty of each participant with respect to the KCRV. However, the effects of correlations have been treated in a simplified way as the degree of confidence in the uncertainties themselves does not warrant a more rigorous approach.

Table 5 shows the matrix of all the degrees of equivalence as they will appear in Appendix B of the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{ei} replaced by x_i. The introductory text is that agreed for the comparison. The graph of the first column of results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV (identified as x_R in the KCDB), is shown in Figure 1. This representation indicates in part the degree of equivalence between the NMIs but does not take into account the correlations between the different NMIs. However, the matrix of degrees of equivalence shown in yellow in Table 5 does take the known correlations into account.

Conclusion

The BIPM ongoing key comparison for ^{131}I, BIPM.RI(II)-K1.I-131 currently comprises thirteen results. These have been analysed with respect to the KCRV determined for this radionuclide, and with respect to each other. The matrix of degrees of equivalence has been approved by the CCRI(II) and is published in the BIPM key comparison database. Other results may be added as and when other NMIs contribute ^{131}I activity measurements to this comparison.

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Table 5. Table of degrees of equivalence and introductory text for ^{131}I

Key comparison BIPM.RI(II)-K1.I-131

MEASURAND : **Equivalent activity of ^{131}I**

Key comparison reference value: the SIR reference value x_R for this radionuclide is 40 390 kBq, with a standard uncertainty, u_R = 40 kBq (see Section 4.1 of the Final Report). 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 $U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\Sigma u_i^2)^{1/2}$ when each laboratory has contributed to the calculation of x_R, with n the number of laboratories.

The degree of equivalence between two laboratories is given by a pair of numbers: $D_{ij} = D_i - D_j = (x_i - x_j)$ and U_{ij}, its expanded uncertainty (k = 2), both expressed in MBq. The approximation $U_{ij}^2 \sim 2^2(u_i^2 + u_j^2)$ is used in the following table.

Lab *j* ⇨

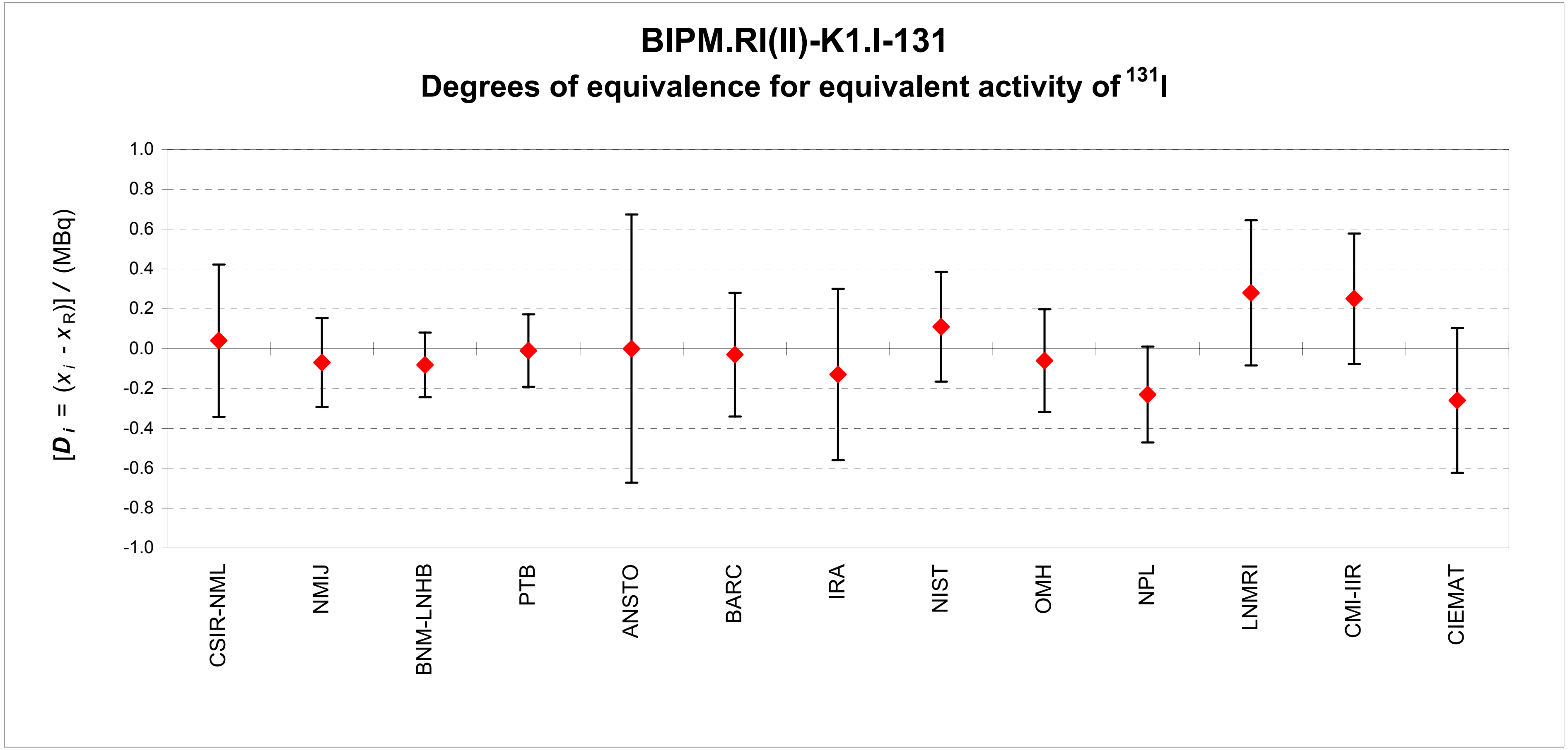
Lab *i* ⇩

			CSIR-NML		NMIJ		BNM-LNHB		PTB		ANSTO		BARC		IRA	
	D_i	U_i	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	/ MBq		/ MBq		/ MBq		/ MBq		/ MBq		/ MBq		/ MBq		/ MBq	
CSIR-NML	**0.04**	0.38			**0.11**	0.46	**0.12**	0.43	**0.05**	0.43	**0.04**	0.82	**0.07**	0.51	**0.17**	0.58
NMIJ	**-0.07**	0.22	**-0.11**	0.46			**0.01**	0.26	**-0.06**	0.28	**-0.07**	0.75	**-0.04**	0.39	**0.06**	0.47
BNM-LNHB	**-0.08**	0.16	**-0.12**	0.43	**-0.01**	0.26			**-0.07**	0.22	**-0.08**	0.73	**-0.05**	0.35	**0.05**	0.44
PTB	**-0.01**	0.18	**-0.05**	0.43	**0.06**	0.28	**0.07**	0.22			**-0.01**	0.74	**0.02**	0.36	**0.12**	0.45
ANSTO	**0.00**	0.67	**-0.04**	0.82	**0.07**	0.75	**0.08**	0.73	**0.01**	0.74			**0.03**	0.79	**0.13**	0.83
BARC	**-0.03**	0.31	**-0.07**	0.51	**0.04**	0.39	**0.05**	0.35	**-0.02**	0.36	**-0.03**	0.79			**0.10**	0.53
IRA	**-0.13**	0.43	**-0.17**	0.58	**-0.06**	0.47	**-0.05**	0.44	**-0.12**	0.45	**-0.13**	0.83	**-0.10**	0.53		
NIST	**0.11**	0.28	**0.07**	0.49	**0.18**	0.36	**0.19**	0.31	**0.12**	0.33	**0.11**	0.77	**0.14**	0.43	**0.24**	0.50
OMH	**-0.06**	0.26	**-0.10**	0.48	**0.01**	0.34	**0.02**	0.30	**-0.05**	0.31	**-0.06**	0.77	**-0.03**	0.41	**0.07**	0.49
NPL	**-0.23**	0.24	**-0.27**	0.47	**-0.16**	0.33	**-0.15**	0.28	**-0.22**	0.29	**-0.23**	0.76	**-0.20**	0.40	**-0.10**	0.48
LNMRI	**0.28**	0.36	**0.24**	0.55	**0.35**	0.44	**0.36**	0.41	**0.29**	0.42	**0.28**	0.81	**0.31**	0.50	**0.41**	0.57
CMI-IIR	**0.25**	0.33	**0.21**	0.52	**0.32**	0.40	**0.33**	0.37	**0.26**	0.38	**0.25**	0.80	**0.28**	0.47	**0.38**	0.54
CIEMAT	**-0.26**	0.36	**-0.30**	0.55	**-0.19**	0.44	**-0.18**	0.41	**-0.25**	0.42	**-0.26**	0.81	**-0.23**	0.50	**-0.13**	0.57

Lab *i* ⇩

			NIST		OMH		NPL		LNMRI		CMI-IIR		CIEMAT	
	D_i	U_i	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}	D_{ij}	U_{ij}
	/ MBq		/ MBq		/ MBq		/ MBq		/ MBq		/ MBq		/ MBq	
CSIR-NML	**0.04**	0.38	**-0.07**	0.49	**0.10**	0.48	**0.27**	0.47	**-0.24**	0.55	**-0.21**	0.52	**0.30**	0.55
NMIJ	**-0.07**	0.22	**-0.18**	0.36	**-0.01**	0.34	**0.16**	0.33	**-0.35**	0.44	**-0.32**	0.40	**0.19**	0.44
BNM-LNHB	**-0.08**	0.16	**-0.19**	0.31	**-0.02**	0.30	**0.15**	0.28	**-0.36**	0.41	**-0.33**	0.37	**0.18**	0.41
PTB	**-0.01**	0.18	**-0.12**	0.33	**0.05**	0.31	**0.22**	0.29	**-0.29**	0.42	**-0.26**	0.38	**0.25**	0.42
ANSTO	**0.00**	0.67	**-0.11**	0.77	**0.06**	0.77	**0.23**	0.76	**-0.28**	0.81	**-0.25**	0.80	**0.26**	0.81
BARC	**-0.03**	0.31	**-0.14**	0.43	**0.03**	0.41	**0.20**	0.40	**-0.31**	0.50	**-0.28**	0.47	**0.23**	0.50
IRA	**-0.13**	0.43	**-0.24**	0.50	**-0.07**	0.49	**0.10**	0.48	**-0.41**	0.57	**-0.38**	0.54	**0.13**	0.57
NIST	**0.11**	0.28			**0.17**	0.38	**0.34**	0.37	**-0.17**	0.47	**-0.14**	0.44	**0.37**	0.47
OMH	**-0.06**	0.26	**-0.17**	0.38			**0.17**	0.35	**-0.34**	0.46	**-0.31**	0.43	**0.20**	0.46
NPL	**-0.23**	0.24	**-0.34**	0.37	**-0.17**	0.35			**-0.51**	0.45	**-0.48**	0.42	**0.03**	0.45
LNMRI	**0.28**	0.36	**0.17**	0.47	**0.34**	0.46	**0.51**	0.45			**0.03**	0.51	**0.54**	0.54
CMI-IIR	**0.25**	0.33	**0.14**	0.44	**0.31**	0.43	**0.48**	0.42	**-0.03**	0.51			**0.51**	0.51
CIEMAT	**-0.26**	0.36	**-0.37**	0.47	**-0.20**	0.46	**-0.03**	0.45	**-0.54**	0.54	**-0.51**	0.51		

Figure 1. Graph of degrees of equivalence with the KCRV for ^{131}I (as it appears in Appendix B of the MRA)



Appendix 1. Uncertainty budgets for the activity of ^{131}I submitted to the SIR

Uncertainty budget for the NPL (1999)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	9	–
weighing, dilution	–	6
dead time	–	5
coincidence resolving time	–	7
background	–	2
half-life	–	1
extrapolation	–	25
radionuclide impurities	–	–
Quadratic summation	**9**	**27**
Relative combined standard uncertainty, u_c	**29**	

Uncertainty budget for ^{131}I at the CMI-IIR (2001)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contribution due to	**A**	**B**
counting statistics	10	–
weighing	–	5
dead time	–	5
resolving time	–	8
background	–	15
adsorption	–	5
radionuclide impurities (none measured with HPGe)	–	5
half-life (8.021 (1) d)	–	1
extrapolation of efficiency curve	–	20
ionization chamber	26	–
Quadratic summation	**28**	**28**
Relative combined standard uncertainty, u_c	**40**	

LNMRI Uncertainty budget for ^{131}I (1999)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contribution due to	**A**	**B**
counting statistics (15 × 10^{-4} included in fitting)	*	–
weighing	–	8
dead time	–	32
resolving time	–	21
background	–	7
delay mismatch	–	16
adsorption	–	–
radionuclide impurities (none measured with HPGe)	–	–
half-life (8.021 (1) d)	–	–
extrapolation of efficiency curve	18	–
ionization chamber	5	–
Quadratic summation	**19**	**43**
Relative combined standard uncertainty, u_c	**47**	

Uncertainty budget for the CIEMAT (2002)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
Contributions due to	**A**	**B**
counting statistics	20	-
weighing, dilution	-	1
dead time	-	10
timing	-	1
extrapolation	40	-
Quadratic summation	**45**	**10**
Relative combined standard uncertainty, u_c	**46**	

Appendix 2. Evaluation of the uncertainty of the degree of equivalence

Table 5 indicates for each laboratory the degree of equivalence D_i with its associated uncertainty U_i. This appendix presents the procedure used to evaluate these uncertainties.

The degree of equivalence of one laboratory is defined as the difference between the individual value of the equivalent activity A_{ei} for an NMI *i* and a suitable reference value which has been evaluated by the KCDB Working Group and the expanded uncertainty of this difference. Currently, the reference value, KCRV, for a given radionuclide is calculated as the arithmetic mean value of the SIR experimental entries for this radionuclide. Briefly at least four situations can occur depending on the consistency of the experimental SIR data sets :

1. All data are consistent and contribute to the reference value; this is the general case;

2. The value obtained by a laboratory that no longer exists, is used as long as it fits the usual quality criteria; it is taken into account when evaluating the reference value but does not appear in the matrices of results;

3. A value, that has been identified for example as an outlier, is not taken into account for the evaluation of the reference value but, nevertheless, the corresponding laboratory appears in the matrices of results.

The situation where a laboratory that no longer exists but contributes to the reference value and where an outlier has been identified in the data set can occur. This is a combination of both situation 2) and situation 3). The results, deduced from these two preceding cases, are also presented here, case 4.

In the following, the expression of the uncertainty for these four cases is considered on the assumption that the uncertainties of the different equivalent activities A_{ei} are not correlated. For the sake of coherence with the definition of the variables used in the text, the following notation is used :

$x_i = A_{ei}$ and $u_i = u_{Aei}$ its uncertainty.

Case 1. All *n* laboratories contribute to the reference value, and appear in Table 5. In this case obviously we have

$$x_{\mathrm{ref}} = \bar{x} = \frac{\sum_{j=1}^{n} x_j}{n} \tag{A-1}$$

$$D_i = x_i - x_{\mathrm{ref}} \tag{A-2}$$

$$D_i = x_i - \frac{\sum_{j=1}^{n} x_j}{n} = x_i\left(1 - \frac{1}{n}\right) - \frac{\sum_{j \neq i}^{n} x_j}{n} \tag{A-3}$$

At this stage the uncertainty of D_i has to be calculated. Applying the method of Gauß for the propagation of the uncertainties it is necessary to calculate the partial derivatives of D_{i} with respect to the x_i.

So $\dfrac{\partial D_i}{\partial x_i} = \left(1 - \dfrac{1}{n}\right)$, and (A – 4)

$\dfrac{\partial D_i}{\partial x_j} = -\dfrac{1}{n}, (j \neq i).$ (A – 5)

Then the total combined uncertainty becomes

$$u_{c_i}^2 = \left(\frac{\partial D_i}{\partial x_i}\right)^2 u_i^2 + \sum_{j \neq i}\left(\frac{\partial D_j}{\partial x_j}\right)^2 u_j^2 \tag{A-6}$$

$$= \left(1 - \frac{1}{n}\right)^2 u_i^2 + \frac{1}{n^2}\sum_{j \neq i} u_j^2 \tag{A-7}$$

or, after recombination

$$= \left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2}\sum_{j=1}^{n} u_j^2. \tag{A-8}$$

When a coverage factor of 2 is used (A-8) becomes

$$U_i^2 = 2^2\left[\left(1 - \frac{2}{n}\right) u_i^2 + \frac{1}{n^2}\sum_{j=1}^{n} u_j^2\right]. \tag{A-9}$$

Case 2. A laboratory was used to evaluate the reference value but does not appear in Table 5.

Let us assign the subscript *n* to the additional laboratory that contributes to the reference value. The uncertainty of this laboratory will appear only in the second part of equation (A-9). Accordingly, equation (A-9) becomes

$$U_i^2 = 2^2\left[\left(1-\frac{2}{n}\right)u_i^2+\frac{1}{n^2}\left(\sum_{j=1}^{n}u_j^2\right)\right], \text{for } i = 1, n-1. \qquad (\text{A}-10)$$

Case 3. The reference value was evaluated with all reported values except one.

For the sake of simplicity let us assign the subscript *n* + 1 to the ineligible laboratory so that the subscript for the other laboratories will run from 1 to *n*. Under this assumption the treatment of the ineligible laboratory will be slightly different and two formulae are deduced.

The ineligible laboratory does not contribute to the reference value, so the term (1 – 2/*n*) in (A-9) reduces to 1 and the uncertainty is simply given by

$$U_{n+1}^2 = 2^2\left[u_{n+1}^2+\frac{1}{n^2}\sum_{j=1}^{n}u_j^2\right]. \qquad (\text{A}-11)$$

In the evaluation of the uncertainty related to the *n* other laboratories the contribution from laboratory *n* + 1 disappears totally and the uncertainty remains given by the expression (A-10) without restriction over the subscript range i. e.

$$U_i^2 = 2^2\left[\left(1-\frac{2}{n}\right)u_i^2+\frac{1}{n^2}\sum_{j=1}^{n}u_j^2\right]. \qquad (\text{A}-12)$$

Case 4. A laboratory that no longer exists contributes to the reference value and an outlier has been identified for another laboratory.

Let us assign the subscript *n* to the defunct existing laboratory so that the expression for the mean (A-1) remains applicable. In addition the outlier will be labelled by *n* + 1. For the (*n* – 1) first laboratories which contribute to the mean value and appear in Table 5 the uncertainty of D_i is given by

$$U_i^2 = 2^2\left[\left(1-\frac{2}{n}\right)u_i^2+\frac{1}{n^2}\sum_{j=1}^{n}u_j^2\right], \text{for } i = 1, n-1. \qquad (\text{A}-13)$$

For the laboratory *n* + 1 that is ineligible for the KCRV, its coefficient (1 – 2/*n*) in (A-13) reduces to 1 and the expression of the uncertainty in Table 5 becomes

$$U_{n+1}^2 = 2^2\left[u_{n+1}^2+\frac{1}{n^2}\sum_{j=1}^{n}u_j^2\right], \qquad (\text{A}-14)$$

similar to (A-11).

Appendix 3. Acronyms used to identify different measurement methods

Each acronym has six components, geometry-detector (1)-radiation (1)-detector (2)-radiation (2)-mode. When a component is unknown, ?? is used and when it is not applicable 00 is used.

Geometry	**acronym**	**Detector**	**acronym**
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop counter	PP
2π	2P	liquid scintillation counting	LS
undefined solid angle	UA	NaI(Tl)	NA
		Ge(HP)	GH
		Ge-Li	GL
		Si-Li	SL
		CsI	CS
		ionization chamber	IC
		grid ionization chamber	GC
		bolometer	BO
		calorimeter	CA
		PIPS detector	PS

Radiation	**acronym**	**Mode**	**acronym**
positron	PO	efficiency tracing	ET
beta particle	BP	internal gas counting	IG
Auger electron	AE	CIEMAT/NIST	CN
conversion electron	CE	sum counting	SC
bremsstrahlung	BS	coincidence	CO
gamma ray	GR	anti-coincidence	AC
X - rays	XR	coincidence counting with efficiency tracing	CT
alpha - particle	AP	anti-coincidence counting with efficiency tracing	AT
mixture of various radiation e.g. X and gamma	MX	triple-to-double coincidence ratio counting	TD
		selective sampling	SS

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(GeHP)-anticoincidence counting	4P-PP-MX-GH-GR-AC
4π CsI-β,AX,γ counting	4P-CS-MX-00-00-00
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG