

**BIPM comparison BIPM.RI(II)-K1.Tl-201 of
activity measurements of the radionuclide ^{201}Tl**

G. Ratel and C. Michotte
BIPM

Abstract

Since 1979, six national metrology institutes (NMI) and another laboratory have submitted 16 samples of known activity of ^{201}Tl to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM). The activities ranged from about 3.5 MBq to 207 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 six NMIs. A graphical presentation is also given. The results of this comparison have been approved for publication by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), with comparison identifier BIPM.RI(II)-K1.Tl-201.

1. Introduction

The SIR for activity measurements of γ -ray-emitting radionuclides was established in 1976. Each NMI may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid form. For radioactive gases, a different standard ampoule is used. 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, A_e , are all given in [1].

Since its inception until 31 December 2003, the SIR has measured 849 ampoules to give 615 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.Tl-201 key comparison.

2. Participants

Six NMIs and another laboratory have submitted sixteen ampoules for the comparison of ^{201}Tl activity measurements since 1979. 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.TI-201

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	1979-07-23 1982-12-16 1983-06-29 1998-06-29
–	PTB	Physikalisch-Technische Bundesanstalt	Germany	EUROMET	1981-05-27
NIRH	–	National Institute of Radiation Hygiene	Denmark	EUROMET	1982-12-14
LMRI	BNM-LNHB	Bureau national de métrologie-Laboratoire national Henri Becquerel	France	EUROMET	1983-06-09 2003-09-19
NAC*	CSIR-NML	National Metrology Laboratory	South Africa	SADCMET	1991-10-29
–	ANSTO	Australian Nuclear Science and Technology Organisation	Australia	APMP	1994-06-02
–	OMH	Országos Mérésügyi Hivatal	Hungary	EUROMET	1997-11-07

* 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, only the BNM-LNHB has provided an uncertainty budget.

The original half-life used by the BIPM was 3.044 (9) d [3] and this has been changed to 3.0422 (17) d, the most recent evaluation available [4]. The equivalent activities given in Table 4 have been recalculated accordingly.

Table 2. Standardization methods of the participants for ^{201}Tl

NMI	Method used and acronym (see Appendix 3)	Half-life / d	Activity A_i / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
NIST	Pressurized IC 4P-IC-GR-00-00-00 calibrated in 1976 by $4\pi(e,x)-\gamma$ coinc. 4P-PC-MX-NA-GR-CO	3.047	48 360	79-06-21 20 h UT	0.02	0.49
		3.0465 (10)	13 118	82-12-15 12 h UT	0.01	0.48
		—	11 226	83-06-22 19 h UT	0.01	0.48
		3.045 (1)	68 500	98-06-29 12 h UT	0.03	0.38
PTB	$4\pi\text{PPC-}\gamma$ (GeLi) coincidence 4P-PP-MX-GL-GR-CO	—	16 441 [†] 16 418	81-05-28 0 h UT	0.11	0.56
NIRH	Pressurized IC* 4P-IC-GR-00-00-00	—	207 400 [†] 198 390	82-12-13 12 h UT	1.4	1.4

continued overleaf.

Table 2 continued. Standardization methods of the participants for ^{201}Tl

NMI	Method used and acronym (see Appendix 3)	Half-life / d	Activity A_i / kBq	Reference date YY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
BNM-LNHB	$4\pi\gamma$ 4P-NA-GR-00-00-00	—	3 512 [†] 3 449	83-06-10 12 h UT	0.04	0.54
	$4\pi\text{LSe}_c\text{-}\gamma$ anti-coincidence 4P-LS-CE-GH-GR-AC	3.0409 (29) [8]	7 605 [†] 8 134 7 714	03-09-16 12 h UT	0.37	0.20
CSIR-NML	$4\pi\text{LS}(e,x)\text{-}\gamma$ coincidence [9] 4P-LS-MX-NA-GR-CO	3.038 [10]	56 800	91-10-24 12 h UT	0.06	0.52
ANSTO	Pressurized IC** 4P-IC-GR-00-00-00	—	11 460	94-05-19 23 h 01 UT	0.04	3.02
OMH	$4\pi(\text{PPC})\text{-}\gamma$ coinc. and anti-coinc. 4P-PP-MX-NA-GR-CO 4P-PP-MX-NA-GR-AC	3.043 (3) [11]	32 060	97-11-07 0 h UT	0.06	1.20

[†] more than one ampoule submitted

* result determined from the efficiency curve for the NIRH IG11 IC

** calibrated by interpolation from the standardization of other radionuclides at the ANSTO.

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 ^{201}Tl submitted

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. / (μg g ⁻¹)	Density / (g cm ⁻³)	Relative activity of impurity [†]
NIST	TlNO ₃ in HNO ₃	1	TlNO ₃ : 48	1.029 (2)*	²⁰⁰ Tl : 0.008 (1) % ²⁰² Tl : 0.21 (2) %
	TlCl in HCl	0.1	TlCl: 117	1.002	²⁰² Tl : 2.1 (2) %
	TlNO ₃ in HNO ₃	1	TlNO ₃ : 10	1.031	²⁰⁰ Tl : 0.012 (2) % ²⁰² Tl : 0.291 (44) %
	TlNO ₃ in HNO ₃	1.15	TlNO ₃ : 150	1.035	²⁰⁰ Tl : 0.008 (1) % ²⁰² Tl : 0.60 (1) % ^{††}
PTB	Tl ₂ SO ₄ in HNO ₃	0.1	Tl ₂ SO ₄ : 40	1.0	²⁰² Tl : 0.41 (2) % ²⁰³ Pb : 0.056 (2) %
NIRH	TlCl in benzyl alcohol C ₇ H ₈ O and NaCl	0.08	NaCl: 9000	–	²⁰⁰ Tl : 0.050 (5) % ²⁰² Tl : 0.16 (1) %
BNM-LNHB	TlCl in HCl	0.1	TlCl: 10	0.999	²⁰⁰ Tl : 0.152 (15) % ²⁰² Tl : 0.29 (3) %
	Tl in HCl	0.1	Tl : 30	1	²⁰⁰ Tl : 0.19 (2) % ²⁰² Tl : 0.26 (3) % ²⁰⁰ Pb : 0.035 (4) % ²⁰³ Pb : 0.033 (4) %
CSIR-NML	TlNO ₃ in HNO ₃	0.1	Tl ⁺ : 500	1.0020	²⁰⁰ Tl : 0.0437 (13) % ²⁰² Tl : 0.0230 (12) %
ANSTO	TlCl	–	Tl	–	²⁰⁰ Tl : 0.22 (2) % ²⁰² Tl : 0.17 (2) %
OMH	TlCl in isotonic NaCl	–	NaCl: 9000	–	²⁰⁰ Tl : 0.009 (2) % ²⁰² Tl : 0.76 (6) % [#]

[†] the ratio of the activity of the impurity to the activity of ^{201}Tl at the reference date

* at 23.6 °C

^{††} confirmed by γ -spectrometry measurements made at the BIPM

[#] γ -spectrometry measurement at the BIPM gives 0.801 (12) % for ^{202}Tl (see section 4).

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 ^{201}Tl arise from sixteen 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 and are used in the KCDB and all references in this report.

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].

The correction factors for impurities are typically between 1.02 and 1.08 but reach 1.16 for the ANSTO, 1.23 for the NIST (1982) and 5.40 for the NIST (1979). They are dominated by the contribution of ^{202}Tl . The ^{202}Tl corrections were revised for the present report using a half-life of 12.65 (32) d [12] and photon emission probabilities from [13]. The equivalent activity of ^{201}Tl used in all the impurity corrections is the current KCRV (314 MBq). These three changes produce better coherence in the repeated measurements described hereafter and several decreasing trends previously observed are now resolved. However, more half-life measurements for both ^{201}Tl and ^{202}Tl are required in order to resolve the discrepancies between previously published values [4], [14], [15] and [16].

Measurements repeated at the BIPM after periods of up to about 5 days later produced comparison results in agreement within the SIR standard uncertainty for NIST (1983) and BNM-LNHB (1983 and 2003). The agreement is within one SIR combined uncertainty for the ANSTO and within two SIR combined uncertainties for the PTB and the NIST (1982).

Measurements repeated at the BIPM after periods of up to about 3 weeks later produced slightly increasing results for the NIRH and decreasing results at a rate of about 2×10^{-3} per day for the OMH (see below). The SIR measurement of the NIST (1998) ampoule was repeated after 3 weeks giving a result in agreement within the SIR uncertainty.

Measurements were made at the BIPM concerning the impurities noted by the OMH. A slightly higher ^{202}Tl activity value was measured (see Table 3), which resolves, within the SIR uncertainty, the decrease observed in the repeated SIR measurements. However, as the OMH and BIPM ^{202}Tl activities agree within the uncertainty, this does not produce a significant effect on the final SIR result (always taken as the first of the series): the difference in A_e is 1200 kBq, $u = 1800$ kBq. Therefore, the OMH decided to use their impurity values in Table 3 for the final result in Table 4, leaving the trend observed in the repeated SIR measurements unresolved.

In principle, the chemical composition of the solutions could have an influence on the SIR measurements due to the intense x-ray emission from ^{201}Tl . Indeed, using the efficiency curve of the SIR [17], the relative contribution of the x-rays to the ionization current is estimated to be 65 %. As a consequence, a supplementary

contribution¹ of 9×10^{-4} was added to the SIR relative uncertainty when the density of the solution is 1.03 g cm^{-3} or more. Further study of this phenomenon is desirable.

One earlier submission was withdrawn and does not appear here. The recent BNM-LNHB submission was identified as a pilot study and is not included in the KCDB. For all other NMIs, the most recent result is normally eligible for Appendix B of the MRA. However, the NIRH no longer undertakes the metrology of activity so this result is not included in the KCDB.

Table 4. Results of SIR measurements of ²⁰¹Tl

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 $u_{c,i} / \text{kBq}$
NIST	3.714 20	48 360	1	329 000	$7.8 \times 10^{-2} \#$	26 000
	3.740 46	13 118	3	311 800	$2.2 \times 10^{-2} \#\#$	7 000
	3.691 6	11 226	1	313 600	$1.3 \times 10^{-2} \#\#$	4 200
	3.745 77	68 500	4	318 000	$16 \times 10^{-4} *$	1 300
PTB	3.706 4(1)	16 441	3	308 150	$19 \times 10^{-4} \#\#$	1 900
	3.701 1(1)	16 418		308 340 [†]		1 900
NIRH	3.684 5	207 400	5	321 440	$16 \times 10^{-4} \#\#$	6 400
	3.524 4	198 390		314 520 [†]		6 200
BNM-LNHB	3.699 60	3 512	2	300 100	$62 \times 10^{-4} \#\#$	2 500
	3.632 80	3 449		299 820 [†]	59×10^{-4}	2 400
	3.5619 (1)	7 605	1	318 770	$47 \times 10^{-4} \#\#$	2 000
	3.8099 (1)	8 134		318 040	47×10^{-4}	2 000
	3.6133 (1)	7 714		317 120 ^{††}	48×10^{-4}	2 000 ^{††}
CSIR-NML	3.599 0**	56 800	3	312 500	12×10^{-4}	1 700
ANSTO	3.569	11 460	1	305 800	$1.6 \times 10^{-2} \#$	10 500
OMH	3.605 8	32 060	3	314 600	$56 \times 10^{-4} \#\#$	4 200

[†] the mean of the A_e values shown for the same measurement date is used with an averaged uncertainty, as attributed to an individual entry [18]

^{††} pilot study: result not included in the KCDB

* uncertainty increased from 13×10^{-4} to 16×10^{-4} due to solution density (see paragraph 4)

** mass before dilution : 1.67887 g

due to a long delay between the measurement in the SIR and the reference date, combined with the uncertainty on the impurity as given by the laboratory. Revised uncertainty evaluation according to the present SIR procedure

\#\# reflecting the uncertainty on the impurity as given by the laboratory.

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

¹ Based on an experimental study of the SIR carried out by Dr Rytz using ²⁴¹Am [19].

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.

On the advice of the BIPM, the KCWG has agreed that although the BNM-LNHB (2003) result is a pilot study it may be included in the KCRV rather than the previous 1983 result. This situation is unusual but has been accepted, as the new BNM-LNHB measurement method is believed to produce a more reliable value for the measurement of ^{201}Tl than that of 1983. Consequently, the KCRV for ^{201}Tl has been identified as 314.3 (1.8) MBq using the results from the PTB, CSIR-NML, OMH, NIST (1998) and the BNM-LNHB (2003). None of the earlier results has been used for the NIST even though the most recent result is furthest in time from the calibration of the ionization chamber. This is appropriate because of the large SIR uncertainty due to the delay between the SIR measurement and the reference date or the large NIST uncertainty for the impurity in the earlier results.

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

$$D_i = A_{ei} - \text{KCRV} \quad (1)$$

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

$$U_i = 2u_{D_i}, \quad (2)$$

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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \quad (3)$$

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

$$u_{D_{ij}}^2 = u_i^2 + u_j^2 - \sum_k (f_k u_{\text{corr},k})_i^2 - \sum_k (f_k u_{\text{corr},k})_j^2 \quad (4)$$

and any obvious correlations in the standard uncertainties for a given component, $u_{\text{corr},k}$, 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 where, following the advice of the CCRI, measurements made prior to 1983 are indicated as black squares. This graphical 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 ^{201}Tl , BIPM.RI(II)-K1.Tl-201 currently comprises six 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 ^{201}Tl activity measurements to this comparison.

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Table 5. Table of degrees of equivalence and introductory text for ²⁰¹Tl

Key comparison BIPM.RI(II)-K1.TI-201

MEASURAND : Equivalent activity of ²⁰¹Tl

Key comparison reference value: the SIR reference value for this radionuclide is $x_R = 314.3$ MBq, with a standard uncertainty $u_R = 1.8$ MBq. x_R is computed as the mean of the results obtained by primary methods.

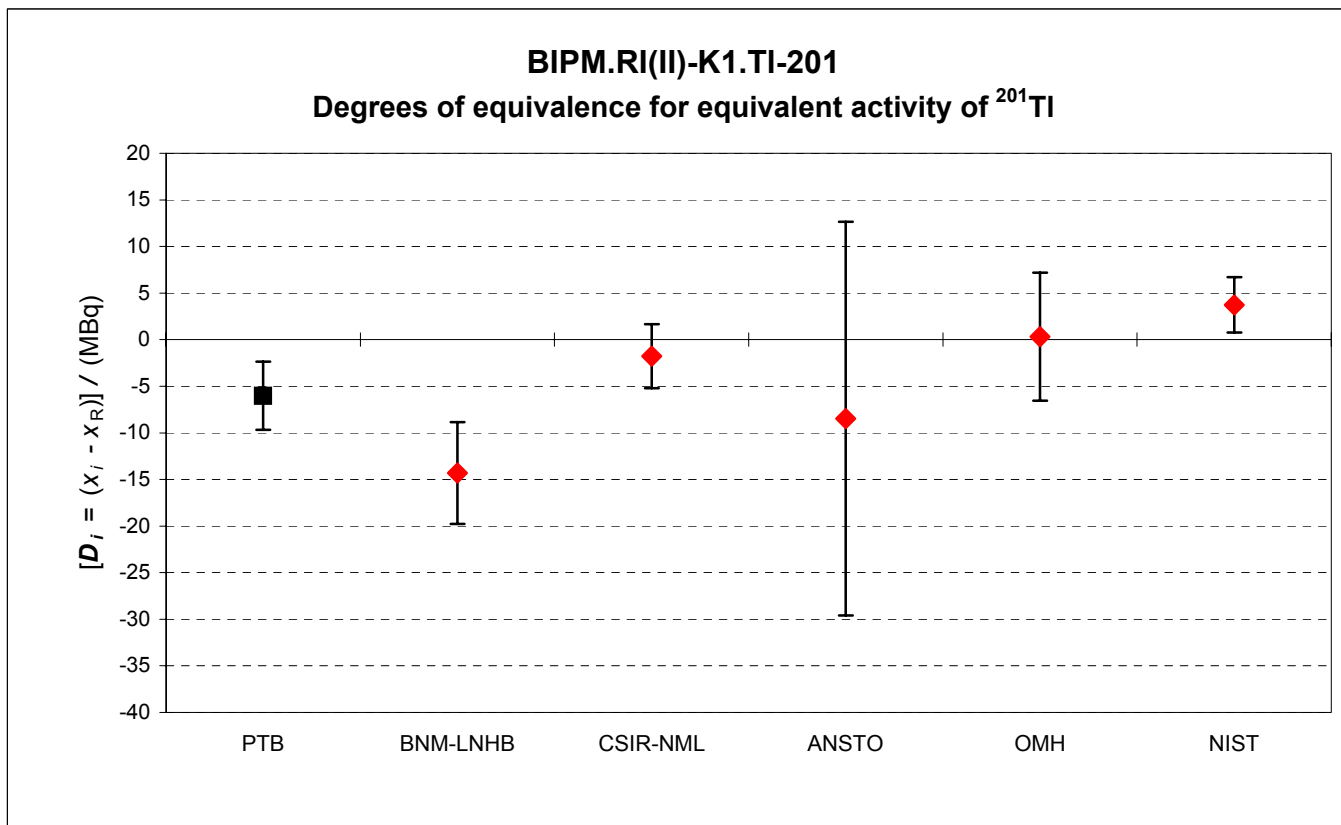
The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_R)$ and U_i , its expanded uncertainty ($k = 2$), both expressed in MBq, with n the number of laboratories, $U_i = 2((1-2/n)u_i^2 + (1/n^2)\sum u_i^2)^{1/2}$ when each laboratory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

The degree of equivalence between two laboratories is given by a pair of terms: $D_{ij} = D_i - D_j = (x_i - x_j)$ and U_{ij} , its expanded uncertainty ($k = 2$), both expressed in MBq.

The approximation $U_{ij} \sim 2(u_i^2 + u_j^2)^{1/2}$ is used in the following table.

Lab <i>i</i> ↓			Lab <i>j</i> →											
	D_i	U_i	PTB		BNM-LNHB		CSIR-NML		ANSTO		OMH		NIST	
	/ MBq		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	
PTB	-6.0	3.7			8.3	6.3	-4.3	5.1	2.5	21.3	-6.4	9.2	-9.8	4.6
BNM-LNHB	-14.3	5.5	-8.3	6.3			-12.5	6.0	-5.8	21.6	-14.6	9.8	-18.0	5.6
CSIR-NML	-1.8	3.4	4.3	5.1	12.5	6.0			6.7	21.3	-2.1	9.1	-5.5	4.3
ANSTO	-8.5	21.1	-2.5	21.3	5.8	21.6	-6.7	21.3			-8.8	22.6	-12.2	21.2
OMH	0.3	6.9	6.4	9.2	14.6	9.8	2.1	9.1	8.8	22.6			-3.4	8.8
NIST	3.7	3.0	9.8	4.6	18.0	5.6	5.5	4.3	12.2	21.2	3.4	8.8		

Figure 1. Graph of degrees of equivalence with the KCRV for ^{201}Tl
(as it appears in Appendix B of the MRA)



Appendix 1. Uncertainty budgets for the activity of ^{201}Tl submitted to the SIR

The detailed uncertainty budget of the BNM-LNHB is as follows:

BNM-LNHB (2003)

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
Contributions due to		
counting statistics	12	-
weighing	-	5
live time	-	< 0.5
counting time	-	1
accidental coincidences (LS channel)	-	< 2
half-life	-	15
extrapolation technique	35	-
decay scheme correction	-	5
radionuclide impurities	-	10
Quadratic summation	37	20
Relative combined standard uncertainty, u_c	42	

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 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 $u_i = u_{A_{ei}}$ its uncertainty.

Case 1. All n laboratories contribute to the reference value, and appear in Table 5.

In this case obviously we have

$$x_{\text{ref}} = \bar{x} = \frac{\sum_{j=1}^n x_j}{n} \quad (\text{A-1})$$

$$D_i = x_i - x_{\text{ref}} \quad (\text{A-2})$$

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

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 .

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

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

Then the total combined uncertainty becomes

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

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

or, after recombination

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

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

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

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

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

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

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

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.

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

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

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

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

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

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
		high efficiency	HE
		digital coincidence counting	DC

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