

Activity measurements of the radionuclide ^{60}Co for the RC, Poland

in the ongoing comparison BIPM.RI(II)-K1.Co-60

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Abstract

In 2003, the Radioisotope Centre, (RC, POLATOM, Poland) submitted a sample of known activity of ^{60}Co to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures. The value of the activity submitted was about 180 kBq. The result of this comparison has been approved by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), comparison identifier BIPM.RI(II)-K1.Co-60 and has been added to the matrix of degrees of equivalence that now contains twenty-one results.

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, the SIR has measured over 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) that was set up under the Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is part of the BIPM.RI(II)-K1.Co-60 key comparison and the results are displayed in the KCDB.

2. Participants

As well as the RC, twenty other NMIs, two other laboratories and the BIPM have submitted sixty-one ampoules for the comparison of ^{60}Co activity measurements since 1976. The date of the RC submission is given in Table 1 and those of the other submissions are given in [3 and 4].

Table 1. Details of the RC participation in the BIPM.RI(II)-K1.Co-60

NMI	Full name	Country	Regional metrology organization	Date of measurement at the BIPM
RC	Radioisotope Centre, POLATOM	Poland	EUROMET	2003-06-17

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 method for the laboratory, the activities submitted and the relative standard uncertainties ($k = 1$) are given in Table 2. The uncertainty budgets are given in Appendix 1.

Table 2. Standardization methods of the RC for ^{60}Co

NMI	Method used and acronym [3]	Half-life / d	Activity A_i / kBq	Reference date YYYY-MM-DD	Relative standard uncertainty $\times 100$ by method of evaluation	
					A	B
RC	$4\pi\beta(\text{LS})-\gamma$ coincidence and anti-coincidence [5] 4P-LS-BP-NA-GR-CO and 4P-LS-BP-NA-GR-AC	1925.3 [6]	177.4	2003-04-03 12 h UT	0.27	0.50

Details regarding the solution submitted are shown in Table 3, including any impurities, when present, as identified by the laboratory. When given, the standard uncertainties on these evaluations are given. The BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [7]. The CCRI(II) agreed in 1999 [8] that this method should be followed according to

the protocol described in [9] when an NMI makes such a request or when there appear to be discrepancies. No supplementary measurements were made for this latest result.

Table 3. Details of the solution of ^{60}Co submitted

NMI	Chemical composition	Solvent conc. / (mol dm ⁻³)	Carrier: conc. / (μg g ⁻¹)	Density / (g cm ⁻³)	Relative activity of impurity [†]
RC	CoCl ₂ in HCl	0.1	Co : 25	1.00	< 0.1 %

[†] the ratio of the activity of the impurity to the activity of ^{60}Co at the reference date.

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The previous activity measurements for ^{60}Co arise from sixty ampoules and the SIR equivalent activity, A_{ei} , for each ampoule is given in [3, 4] for each NMI, i . The activity measurement for the RC is given in Table 4. The relative standard uncertainty arising from the measurements in the SIR is 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 half-life currently used by the BIPM is that recommended by the IAEA, 1925.5 (5) d [10]. The date of measurement in the SIR is given in Table 1.

Table 4. Results of SIR measurements of ^{60}Co for the RC

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
RC	3.699 87	177.4	2	7040	8×10^{-4}	40

4.1 The key comparison reference value

The key comparison reference value for ^{60}Co is 7064.6 (3.8) kBq as given in [3]. 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.

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

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

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.

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

$$D_{ij} = D_i - D_j = A_{e_i} - A_{e_j} \quad (3)$$

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. 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 appear in Appendix B of the KCDB including the present RC result. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with A_{e_i} 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 ^{60}Co , BIPM.RI(II)-K1.Co-60 currently comprises twenty-one results, including the 2003 result for the RC. 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 ^{60}Co activity measurements to this comparison.

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Table 5. Table of degrees of equivalence and introductory text for ⁶⁰Co

Key comparison BIPM.RI(II)-K1.Co-60

MEASURAND : Equivalent activity of ⁶⁰Co

Key comparison reference value: the SIR reference value x_R for this radionuclide is 7065 kBq, with a standard uncertainty u_R of 4 kBq [Metrologia, 2003, 40, Tech. Suppl., 06007].

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 kBq, and $U_j = 2((1 - 2/n)u_i^2 + (1/n^2)\sum u_j^2)^{1/2}$ when each laboratory has contributed to the calculation of x_R , with n the number of laboratories [Metrologia, 2003, 40, Tech. Suppl., 06007].

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 kBq.

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

Lab j →

Lab i ↓

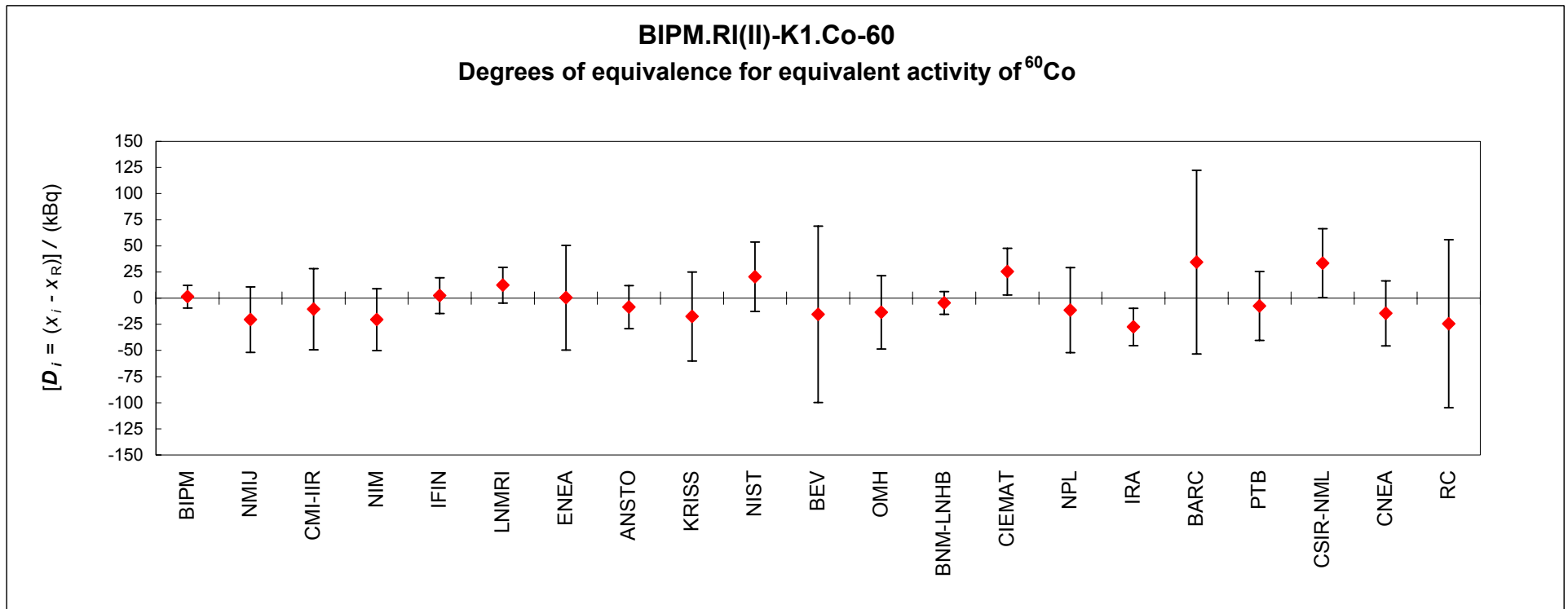
	D_i / kBq		U_i / kBq		BIPM		NMIJ		CMI-IIR		NIM		IFIN		LNMRI		ENEA		ANSTO		KRISS		NIST		BEV			
	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq
BIPM	1	11																										
NMIJ	-21	31	-22	33																								
CMI-IIR	-11	39	-12	41	10	51																						
NIM	-21	30	-22	31	0	44	-10	51	0	44																		
IFIN	2	17	1	18	23	36	13	43	23	34																		
LNMRI	12	17	11	18	33	36	23	43	33	34	10	23																
ENEA	0	50	-1	53	21	61	11	66	21	60	-2	54	-12	54														
ANSTO	-9	21	-10	22	12	38	2	45	12	36	-11	26	-21	26	-9	56												
KRISS	-18	43	-19	45	3	54	-7	59	3	53	-20	47	-30	47	-18	68	-9	48										
NIST	20	33	19	35	41	47	31	52	41	45	18	38	8	38	20	62	29	39	38	56								
BEV	-16	84	-17	84	5	90	-5	93	5	89	-18	86	-28	86	-16	99	-7	86	2	95	-36	91						
OMH	-14	35	-15	37	7	48	-3	54	7	47	-16	39	-26	39	-14	63	-5	41	4	57	-34	50	2	91				
BNM-LNHB	-5	11	-6	11	16	33	6	41	16	31	-7	18	-17	18	-5	53	4	22	13	45	-25	35	11	84				
CIEMAT	25	22	24	23	46	39	36	46	46	37	23	27	13	27	25	56	34	30	43	49	5	40	41	87				
NPL	-12	41	-13	43	9	53	-1	58	9	52	-14	45	-24	45	-12	67	-3	47	6	61	-32	54	4	77				
IRA	-28	18	-29	18	-7	36	-17	43	-7	34	-30	23	-40	23	-28	54	-19	26	-10	47	-48	38	-12	86				
BARC	34	88	33	92	55	97	45	100	55	97	32	93	22	93	34	106	43	94	52	102	14	98	50	125				
PTB	-8	33	-9	33	13	45	3	51	13	44	-10	36	-20	36	-8	61	1	38	10	54	-28	47	8	90				
CSIR-NML	33	33	32	33	54	45	44	51	54	44	31	36	21	36	33	61	42	38	51	54	13	47	49	90				
CNEA	-15	31	-16	31	6	44	-4	50	6	42	-17	34	-27	34	-15	60	-6	36	3	53	-35	45	1	89				
RC	-25	80	-26	80	-4	86	-14	89	-4	85	-27	82	-37	82	-25	95	-16	82	-7	91	-45	87	-9	116				

Lab j →

Lab i ↓

	D_i / kBq		U_i / kBq		OMH		BNM-LNHB		CIEMAT		NPL		IRA		BARC		PTB		CSIR-NML		CNEA		RC		
	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	D_{ij} / kBq	U_{ij} / kBq	
BIPM	1	11	15	37	6	11	-24	23	13	43	29	18	-33	92	9	33	-32	33	16	31	26	80			
NMIJ	-21	31	-7	48	-16	33	-46	39	-9	53	7	36	-55	97	-13	45	-54	45	-6	44	4	86			
CMI-IIR	-11	39	3	54	-6	41	-36	46	1	58	17	43	-45	100	-3	51	-44	51	4	50	14	89			
NIM	-21	30	-7	47	-16	31	-46	37	-9	52	7	34	-55	97	-13	44	-54	44	-6	42	4	85			
IFIN	2	17	16	39	7	18	-23	27	14	45	30	23	-32	93	10	36	-31	36	17	34	27	82			
LNMRI	12	17	26	39	17	18	-13	27	24	45	40	23	-22	93	20	36	-21	36	27	34	37	82			
ENEA	0	50	14	63	5	53	-25	56	12	67	28	54	-34	106	8	61	-33	61	15	60	25	95			
ANSTO	-9	21	5	41	-4	22	-34	30	3	47	19	26	-43	94	-1	38	-42	38	6	36	16	82			
KRISS	-18	43	-4	57	-13	45	-43	49	-6	61	10	47	-52	102	-10	54	-51	54	-3	53	7	91			
NIST	20	33	34	50	25	35	-5	40	32	54	48	38	-14	98	28	47	-13	47	35	45	45	87			
BEV	-16	84	-2	91	-11	84	-41	87	-4	77	12	86	-50	125	-8	90	-49	90	-1	89	9	116			
OMH	-14	35			-9	37	-39	42	-2	55	14	39	-48	99	-6	48	-47	48	1	47	11	88			
BNM-LNHB	-5	11	9	37			-30	23	7	43	23	18	-39	92	3	33	-38	33	10	31	20	80			
CIEMAT	25	22	39	42	30	23			37	47	53	27	-9	95	33	39	-8	39	40	37	50	83			
NPL	-12	41	2	55	-7	43	-37	47			16	45	-46	101	-4	53	-45	53	3	52	13	90			
IRA	-28	18	-14	39	-23	18	-53	27	-16	45			-62	93	-20	36	-61	36	-13	34	-3	82			
BARC	34	88	48	99	39	92	9	95	46	101	62	93			42	97	1	97	49	97	59	122			
PTB	-8	33	6	48	-3	33	-33	39	4	53	20	36	-42	97			-41	45	7	44	17	86			
CSIR-NML	33	33	47	48	38	33	8	39	45	53	61	36	-1	97	41	45			48	44	58	86			
CNEA	-15	31	-1	47	-10	31	-40	37	-3	52	13	34	-49	97	-7	44	-48	44			10	85			
RC	-25	80	-11	88	-20	80	-50	83	-13	90	3	82	-59	122	-17	86	-58	86	-10	85					

Figure 1. Graph of degrees of equivalence with the KCRV for ^{60}Co
 (as it appears in Appendix B of the MRA)



Appendix 1. Uncertainty budget for the activity of ^{60}Co submitted to the SIR**Uncertainty evaluation of the activity of RC ampoule 767a (2003)**

Relative standard uncertainties	$u_i \times 10^4$ evaluated by method	
	A	B
counting statistics, $\nu = 11^\dagger$	11	–
weighing	24	–
dead-time	1	–
coincidence resolving time	2	–
counting method	–	50
Quadratic summation	26.5	50
Total relative combined uncertainty u_c	57	

† number of degrees of freedom