



**HAL**  
open science

**Update of the BIPM comparison  
BIPM.RI(II)-K1.Cd-109 of activity measurements of the  
radionuclide  $^{109}\text{Cd}$  to include the 2012 result of the  
LNE-LNHB (France)**

C Michotte, S Courte, M Nonis, R Coulon, S Judge, G Ratel, Carole Fréchou,  
Cheick Thiam

► **To cite this version:**

C Michotte, S Courte, M Nonis, R Coulon, S Judge, et al.. Update of the BIPM comparison BIPM.RI(II)-K1.Cd-109 of activity measurements of the radionuclide  $^{109}\text{Cd}$  to include the 2012 result of the LNE-LNHB (France). *Metrologia*, 2021, 58 (1A), pp.06026. 10.1088/0026-1394/58/1A/06026 . cea-04127821

**HAL Id: cea-04127821**

**<https://cea.hal.science/cea-04127821v1>**

Submitted on 15 Jun 2023

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Update of the BIPM comparison BIPM.RI(II)-K1.Cd-109 of activity measurements of the radionuclide $^{109}\text{Cd}$ to include the 2012 result of the LNE-LNHB (France)

C. Michotte<sup>1</sup>, S. Courte<sup>1</sup>, M. Nonis<sup>1</sup>, R. Coulon<sup>1</sup>, S. Judge<sup>1</sup>,  
G. Ratel<sup>1</sup>, C. Fréchou<sup>2</sup>, C. Thiam<sup>2</sup>

<sup>1</sup> Bureau International des Poids et Mesures, Pavillon de Breteuil, F-92312 Sèvres  
Cedex, France.

<sup>2</sup> Laboratoire National de métrologie et d'Essais -Laboratoire National Henri  
Becquerel (LNE-LNHB), Gif-sur-Yvette, France.

E-mail: cmichotte@bipm.org

**Abstract** Since 1978, 10 laboratories have submitted 23 samples of  $^{109}\text{Cd}$  to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures (BIPM), with comparison identifier BIPM.RI(II)-K1.Cd-109. In 2012, the LNE-LNHB (France) participated in the comparison to update its degree of equivalence. A graphical presentation is also given.

## 1. Introduction

The SIR for activity measurements of  $\gamma$ -ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled with 3.6 g of the radioactive solution. For radioactive gases, a different standard ampoule is used. Each 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  $^{226}\text{Ra}$  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].

From its inception until 31 December 2020, the SIR has been used to measure 1021 ampoules to give 776 independent results for 72 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 standardizations. These comparisons are

described as BIPM continuous comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the Comité International des Poids et Mesures Mutual Recognition Arrangement (CIPM MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Cd-109 key comparison. The results of earlier participations in this key comparison were published previously [3,4].

## 2. Participants

Laboratory details are given in Table 1, with the earlier submissions being taken from [3,4]. The dates of measurement in the SIR given in Table 1 are used in the KCDB and all references in this report. The AECL (Atomic Energy of Canada Ltd) is not part of the NMI in Canada but was an invited participant in various SIR comparisons as, in the early years, J.G.V. Taylor of the AECL was a personal member of the predecessor to the CCRI(II).

Table 1: Details of the participants in the BIPM.RI(II)-K1.Cd-109.

| NMI or laboratory | Previous acronyms          | Full name  | Country        | RMO      | Date of SIR measurement<br>yyyy-mm-dd                    |
|-------------------|----------------------------|--|----------------|----------|--|
| AECL <sup>a</sup> | -                          | Atomic Energy of Canada Ltd  | Canada         | SIM      | 1982-03-31   |
| BIPM              | -                          | Bureau International des Poids et Mesures  |                |          | 1986-03-24   |
| CMI-IIR           | UVVVR                      | Czech Metrological Institute - Inspectorate for Ionizing Radiation                   | Czech Republic | EURAMET  | 1978-09-01<br><br>1986-04-08                             |
| IRA               | IER                        | Institut de Radiophysique  | Switzerland    | EURAMET  | 2000-12-05   |
| LNE-LNHB          | LMRI, LPRI, BNM-LNHB       | Laboratoire National de métrologie et d'Essais -Laboratoire National Henri Becquerel | France         | EURAMET  | 1980-07-02<br><br>1998-01-23<br>2012-02-29               |
| NIST              | NBS                        | National Institute of Standards and Technology                                       | United States  | SIM      | 2004-11-22   |
| NMIJ              | ETL                        | National Metrology Institute of Japan  | Japan          | APMP     | 1996-04-05   |
| NMISA             | NAC, CSIR-NML <sup>b</sup> | National Metrology Institute of South Africa   | South Africa   | AFRIMETS | 1978-04-18<br><br>1979-10-10<br>1981-03-24<br>1982-12-03 |
| NPL               | -                          | National Physical Laboratory   | United Kingdom | EURAMET  | 1980-10-09   |

... Continuation of Table 1.

| NMI or laboratory | Previous acronyms | Full name                             | Country | RMO     | Date of SIR measurement<br>yyyy-mm-dd |
|-------------------|-------------------|---------------------------------------|---------|---------|---------------------------------------|
| PTB               | -                 | Physikalisch-Technische Bundesanstalt | Germany | EURAMET | 1994-11-24<br>2004-09-24              |

<sup>a</sup> federal Crown corporation, not part of the NMI in Canada (see text)

<sup>b</sup> NAC is another institute of 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 appropriate correlations are taken into account.

A brief description of the standardization methods used by the laboratories, the activities submitted, the relative standard uncertainties and the half-life used by the participants are given in Table 2. The uncertainty budget for the new submission is given in Appendix D attached to this report; previous uncertainty budgets are given in the earlier K1 reports [3, 4]. The list of acronyms used to summarize the methods is given in Appendix E.

Since 2012, the half-life used by the BIPM is 461.9(4) days as published in BIPM Monographie 5 vol. 8 [5]. The half-life of 461.4(12) days [6] was used since 2004 and 464(1) days [7] for earlier results.

Table 2: Standardization methods of the participants for  $^{109}\text{Cd}$ .

| NMI or laboratory | Method used and the acronym   | Activity $A_i/\text{MBq}$ | Relative standard uncertainty / $10^{-2}$ |      | Reference date<br>yyyy-mm-dd | Half-life /d |
|-------------------|---|---------------------------|---|------|------------------------------|--------------|
|                   |   |                           | A   | B    |                              |              |
| AECL              | $\gamma$ efficiency tracing using $^{109}\text{Pd}$ (4P-PC-BP-GL-GR-ET) | 17 740                    | 1.2                                       | 0.6  | 1981-09-01<br>17:00 UT       | 462.3(7)     |
| BIPM              | $4\pi(\text{PPC})_{\text{ce}}$ (4P-PP-CE-00-00-00)                      | 21 547 <sup>f</sup>       | 0.01                                      | 0.26 | 1986-03-01<br>00:00 UT       | 462.6(4)     |
|                   |   | 21 708                    | 0.01                                      | 0.26 |                              |              |
| CMI-IIR           | $4\pi(\text{NaI}(\text{Tl}))_{\text{x}}$ (4P-NA-XR-00-00-00)            | 3802                      | 0.2                                       | 1.3  | 1978-07-27<br>11:00 UT       | 475          |
|                   | $4\pi(\text{NaI}(\text{Tl}))_{\text{x}}$ (4P-NA-XR-00-00-00)            | 17 670                    | 0.07                                      | 0.93 | 1986-03-19<br>12:00 UT       | 453.5        |

... Continuation of Table 2.

| NMI or laboratory | Method used and the acronym                                     | Activity $A_i$ /MBq  | Relative standard uncertainty / $10^{-2}$ |      | Reference date<br>yyyy-mm-dd | Half-life /d |
|-------------------|---|----------------------|---|------|------------------------------|--------------|
|                   |   |                      | A   | B    |                              |              |
| IRA               | Ionization chamber (4P-IC-GR-00-00-00) <sup>a</sup>             | 51 320               | 0.08                                      | 0.61 | 2000-12-01<br>12:00 UT       | 462.9(20)    |
| LNE-LNHB          | 4 $\pi$ (PPC)ce (4P-PP-CE-00-00-00)                             | 2716 <sup>f</sup>    | 0.03                                      | 0.14 | 1980-02-19<br>00:00 UT       | -            |
|                   |   | 2712                 | 0.03                                      | 0.14 |                              |              |
|                   | TDCR (4P-LS-MX-00-00-TD)<br>4 $\pi$ (PPC)ce (4P-PP-CE-00-00-00) | 6916 <sup>g</sup>    | 0.99                                      |      | 1997-10-01<br>12:00 UT       | -            |
|                   | 4 $\pi$ (NaI(Tl)) well-type counting (4P-NA-GR-00-00-00)        | 8731                 | 0.05                                      | 0.94 | 2011-06-13<br>12:00 UT       | 461.4(12)    |
| NIST              | Ionization chamber (4P-IC-GR-00-00-00) <sup>a</sup>             | 42 410               | 0.04                                      | 0.51 | 2004-11-15<br>12:00 UT       | 462(1)       |
| NMIJ              | 4 $\pi$ (PPC)ce (4P-PP-CE-00-00-00)                             | 13 903               | 0.19                                      | 0.4  | 1996-03-01<br>12:00 UT       | -            |
| NMISA             | 4 $\pi$ (LS)ce-x coincidence (4P-LS-CE-NA-XR-CO) <sup>b</sup>   | 99 200 <sup>f</sup>  | 0.11                                      | 2    | 1978-03-31<br>10:00 UT       |              |
|                   |   | 101 800              | 0.11                                      | 2    |                              |              |
|                   | 4 $\pi$ (LS)ce-x coincidence (4P-LS-CE-NA-XR-CO) <sup>b</sup>   | 176 400 <sup>f</sup> | 0.13                                      | 1.80 | 1979-09-28<br>13:00 UT       | -            |
|                   |   | 187 900              | 0.13                                      | 1.80 |                              |              |
|                   | 4 $\pi$ (LS)ce (4P-LS-CE-00-00-00) <sup>b</sup>                 | 176 800 <sup>f</sup> | 0.10                                      | 2.00 |                              |              |
|                   |   | 188 400              | 0.10                                      | 2.00 |                              |              |
|                   | 4 $\pi$ (LS)ce-x coincidence (4P-LS-CE-NA-XR-CO) <sup>c</sup>   | 209 400 <sup>f</sup> | 0.1                                       | 1.82 | 1981-02-10<br>12:00 UT       |              |
|                   |   | 212 100              | 0.1                                       | 1.82 |                              |              |
|                   | 4 $\pi$ (LS)ce-x coincidence (4P-LS-CE-NA-XR-CO) <sup>c</sup>   | 82 380 <sup>f</sup>  | 0.03                                      | 0.71 | 1982-10-29<br>10:00 UT       |              |
|                   |   | 79 930               | 0.03                                      | 0.71 |                              |              |
| NPL               | Ionization chamber (4P-IC-GR-00-00-00) <sup>d</sup>             | 39 300               | 0.09                                      | 1.64 | 1980-10-06<br>00:00 UT       |              |
| PTB               | Ionization chamber (4P-IC-GR-00-00-00) <sup>d</sup>             | 7405                 | 0.05                                      | 0.5  | 1994-10-01<br>00:00 UT       |              |
|                   | Ionization chamber (4P-IC-GR-00-00-00) <sup>e</sup>             | 14 962               | 0.06                                      | 0.29 | 2004-01-01<br>00:00 UT       | [10]         |

<sup>a</sup> calibrated in 1986 against 4 $\pi$ (NaI(Tl)) $\gamma$  counting and 4 $\pi$ (LS)ce counting in the frame of the CCRI(II) comparison

<sup>b</sup> see details in [8]

<sup>c</sup> see details in [9]

<sup>d</sup> calibrated by a primary method

<sup>e</sup> calibrated in March 2005 by a combination of 4 $\pi$ (PPC)ce counting 4P-PP-CE-00-00-HE, LS counting 4P-LS-CE-00-00-00 and HPGe spectrometry UA-GH-GR-00-00-00

<sup>f</sup> Several samples submitted

<sup>g</sup> The result is the mean of the different methods.

Details regarding the solutions 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.

Table 3: Details of each solution of  $^{109}\text{Cd}$  submitted.

| NMI or laboratory<br>/ SIR year | Chemical composition                     | Solvent conc.<br>/(mol dm <sup>-3</sup> ) | Carrier conc.<br>/( $\mu\text{g g}^{-1}$ ) | Density<br>/(g cm <sup>-3</sup> ) | Relative activity of any impurity <sup>b</sup>  |
|---------------------------------|--|---|--|-----------------------------------|---|
| AECL 1982                       | CdCl <sub>2</sub> in HCl                 | 0.1                                       | CdCl <sub>2</sub> : 20                     | 1                                 | -   |
| BIPM 1986 <sup>a</sup>          | CdCl <sub>2</sub> in HCl                 | 0.1                                       | CdCl <sub>2</sub> : 20                     | -                                 | <sup>110m</sup> Ag:<br>3.6(7)x10 <sup>-6</sup> %<br><sup>65</sup> Zn: 1.4(3)x10 <sup>-6</sup> %   |
| CMI-IIR<br>1978                 | Cd(NO <sub>3</sub> ) <sub>2</sub> in HCl | 0.08                                      | Cd(NO <sub>3</sub> ) <sub>2</sub> :<br>50  | -                                 | <0.1 %  |
| 1986                            | CdCl <sub>2</sub> in HCl                 | 0.08                                      | CdCl <sub>2</sub> : 50                     | -                                 | <0.1 %  |
| IRA 2000                        | CdCl <sub>2</sub> in HCl                 | 0.1                                       | CdCl <sub>2</sub> : 20                     | 1                                 | negligible  |
| LNE-LNHB<br>1980                | CdCl <sub>2</sub> in HCl                 | 1   | CdCl <sub>2</sub> : 25                     | 1.016                             | <sup>110m</sup> Ag: 0.082(2) %<br><br><sup>60</sup> Co: 0.0182(5) %<br><sup>65</sup> Zn: 0.0142(5) %<br><sup>152</sup> Eu: 0.0021(1) %<br><sup>134</sup> Cs: 0.0004(1) %<br><sup>115m</sup> Cd: 0.03(1) % |
| 1998                            | CdCl <sub>2</sub> in HCl                 | 1   | CdCl <sub>2</sub> : 10                     | 1.016                             | -   |
| 2012                            | CdCl <sub>2</sub> in HCl                 | 1   | CdCl <sub>2</sub> : 10                     | 1.016                             | None detected   |
| NIST 2004                       | CdCl <sub>2</sub> in HCl                 | 0.5                                       | CdCl <sub>2</sub> : 500                    | 1.007(1)                          | -   |
| NMIJ 1996                       | CdCl <sub>2</sub> in HCl                 | 0.1                                       | CdCl <sub>2</sub> : 200                    | 1                                 | <sup>65</sup> Zn: 0.0047(5) %   |
| NMISA 1978                      | CdCl in HCl                              | 1   | CdCl: 95                                   | 1.036                             | -   |
| 1979                            | CdCl <sub>2</sub> in HCl                 | 1   | CdCl <sub>2</sub> : 100                    | 1.036                             | -   |
| 1981                            | CdCl <sub>2</sub> in HCl                 | 1   | Cd <sup>++</sup> : 610                     | 1.037                             | -   |
| 1982                            | CdCl <sub>2</sub> in HCl                 | 1   | Cd <sup>++</sup> : 300                     | 1.037                             | -   |
| NPL 1980                        | CdCl <sub>2</sub> in HCl                 | 0.1                                       | CdCl <sub>2</sub> : 100                    | 1.001                             | -   |
| PTB 1994                        | CdCl <sub>2</sub> in HCl                 | 0.1                                       | CdCl <sub>2</sub> : 45                     | 1                                 | -   |
| 2004                            | CdCl in HCl                              | 0.1                                       | CdCl: 28                                   | 1                                 | -   |

<sup>a</sup> ampoules measured by the BIPM in the frame of the 1986 CCRI(II)-K2.Cd-109 and used to make the link for the CCRI(II) key comparison

<sup>b</sup> the ratio of the activity of the impurity to the activity of  $^{109}\text{Cd}$  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 "master-file". The latest submission has added 1 ampoule for the activity measurements for  $^{109}\text{Cd}$  giving rise to 23 ampoules in total. The SIR equivalent activity,  $A_{ei}$ , for each ampoule received from each NMI,  $i$ , including both previous and new results, is given in Table 4.

The relative standard uncertainties arising from the measurements in the SIR are

also shown. This uncertainty is additional to that declared by the NMI ( $u(A_i)$ ) for the activity measurement shown in Table 2. Although submitted activities are compared with a given source of  $^{226}\text{Ra}$ , all the SIR results are normalized to the radium source number 5 [1].

No recent submission has been identified as a pilot study so the most recent result of each NMI is normally eligible for inclusion on the KCDB platform of the CIPM MRA [2].

In principle, the chemical composition of the solutions could have an influence on the SIR measurements due to the intense x-ray emission from  $^{109}\text{Cd}$ . However, using the efficiency curve of the SIR [11], the contribution of the x rays to the ionization current is estimated to be negligible. In consequence, the influence of the chemical composition on the SIR measurements can also probably be neglected in this case although a more detailed study could be carried out.

Table 4: Results of SIR measurement of  $^{109}\text{Cd}$ .

| NMI or laboratory                              | $m_i$                 | $A_i$             | $^{226}\text{Ra}$ source | $A_{ei}$          | Relative uncert. from SIR | $u_{ci}$ | $A_{ei}$ for KCRV     |
|--|-----------------------|-------------------|--------------------------|-------------------|---------------------------|----------|-----------------------|
| / SIR year                                     | /g                    | /MBq              |                          | /MBq              | / $10^{-4}$               | /MBq     | /MBq                  |
| AECL 1982                                      | 1.613 23 <sup>a</sup> | 17 740            | 1                        | 8110              | 28                        | 110      | 8110(110)             |
| BIPM 1986                                      | 3.605 6               | 21 547            | 1                        | 8112              | 16                        | 25       | -                     |
|  | 3.632 5               | 21 708            | 1                        | 8114              | 16                        | 26       | -                     |
| CMI-IIR 1978<br>1986                           | 3.596 5               | 3802 <sup>c</sup> | 1                        | 7990              | 23                        | 110      | - <sup>b</sup>        |
|  | 3.615 4               | 17 670            | 1                        | 7913              | 25                        | 77       | - <sup>b</sup>        |
| IRA 2000                                       | 3.527<br>60(10)       | 51 320            | 1                        | 8065              | 14                        | 51       | 8065(51)              |
| LNE-LNHB<br>1980<br><br>1998<br>2012           | 3.685 56              | 2716 <sup>c</sup> | 1                        | 8128              | 240 <sup>d</sup>          | 200      | -                     |
|  | 3.679 70              | 2712              | 1                        | 8110              | 240                       | 200      | -                     |
|  | 3.594 4               | 6916 <sup>c</sup> | 1                        | 8187              | 42                        | 88       | -                     |
|  | 3.757                 | 8731 <sup>c</sup> | 1                        | 8042              | 23                        | 78       | -                     |
| NIST 2004                                      | 3.613 9(2)            | 42 410            | 1                        | 8168              | 16                        | 44       | - <sup>e</sup>        |
| NMIJ 1996                                      | 3.594 3               | 13 903            | 1                        | 8157              | 23                        | 41       | -                     |
| NMISA 1978<br><br>1979<br><br>1981<br><br>1982 | 3.600                 | 99 200            | 1                        | 8161              | 13                        | 160      | -                     |
|  | 3.599                 | 101 800           | 1                        | 8164              | 13                        | 160      | -                     |
|  | 3.604                 | 176 400           | 2                        | 8217              | 11                        | 150      | -                     |
|  | 3.605                 | 187 900           | 2                        | 8214              | 11                        | 150      | -                     |
|  | 3.604                 | 176 800           | 2                        | 8238 <sup>h</sup> | 11                        | 170      | -                     |
|  | 3.605                 | 188 400           | 2                        | 8236 <sup>h</sup> | 11                        | 170      | -                     |
| 1981   | 3.610                 | 209 400           | 2                        | 8186              | 11                        | 150      | -                     |
|  | 3.602                 | 212 100           | 2                        | 8187              | 11                        | 150      | -                     |
| 1982   | 3.596                 | 82 380            | 1                        | 8166              | 20 <sup>f</sup>           | 60       | 8171(60) <sup>g</sup> |
|  | 3.595                 | 79 930            | 1                        | 8175              | 21                        | 60       | -                     |

... Continuation of Table 4.

| NMI or laboratory | $m_i$      | $A_i$             | $^{226}\text{Ra}$ source | $A_{ei}$          | Relative uncert. from SIR | $u_{c,i}$ | $A_e$ for KCRV |
|-------------------|------------|-------------------|--------------------------|-------------------|---------------------------|-----------|----------------|
| / SIR year        | /g         | /MBq              |                          | /MBq              | /10 <sup>-4</sup>         | /MBq      | /MBq           |
| NPL 1980          | 3.589      | 39 300            | 1                        | 8130              | 16                        | 130       | 8130(130)      |
| PTB 1994          | 3.698 9    | 7405 <sup>c</sup> | 1                        | 8153 <sup>e</sup> | 37                        | 51        | -              |
| 2004              | 3.640 9(9) | 14 962            | 1                        | 8170              | 29                        | 34        | 8170(34)       |

<sup>a</sup> mass of solution before dilution<sup>b</sup> Result not included in the KCRV because of potential impurity in the solution (see details in [3].)<sup>c</sup> Activity in the ampoule significantly lower than the minimum required for the SIR (45 MBq)<sup>d</sup> the SIR uncertainty reflects the uncertainties of the impurity measurements reported by the NMI<sup>e</sup> Result not included in the KCRV because of the slow drift of the ampoule holder's height in the IC at the NIST discovered in 2012<sup>f</sup> changed from  $14 \times 10^{-4}$  to  $20 \times 10^{-4}$  and  $21 \times 10^{-4}$  due to the density effect in the SIR at low energy [12]<sup>g</sup> An average value and average uncertainty between all submitted samples is used for the KCDB [13].<sup>h</sup> unofficial result

#### 4.1. The key comparison reference value

In May 2013, the CCRI(II) decided to calculate the key comparison reference value (KCRV) by using the power-moderated weighted mean [14] rather than an unweighted mean, as had been the policy. This type of weighted mean is similar to a Mandel-Paule mean in that the NMIs' uncertainties may be increased until the reduced chi-squared value is one. In addition, it allows for a power  $\alpha$  smaller than two in the weighting factor. As proposed in [14],  $\alpha$  is taken as  $2 - 3/N$  where  $N$  is the number of results selected for the KCRV. Therefore, all SIR key comparison results can be selected for the KCRV with the following provisions:

- only results for solutions standardized by primary techniques are accepted, with the exception of radioactive gas standards (for which results from transfer instrument measurements that are directly traceable to a primary measurement in the laboratory may be included);
- each NMI or other laboratory may only use one result (normally the most recent result or the mean if more than one ampoule is submitted);
- results more than 20 years old are included in the calculation of the KCRV but are not included in data shown in the KCDB or in the plots in this report, as they have expired;
- possible outliers can be identified on a mathematical basis and excluded from the KCRV using the normalized error test with a test value of 2.5 and using the modified uncertainties;
- results can also be excluded for technical reasons; and



- (f) the CCRI(II) is always the final arbiter regarding excluding any data from the calculation of the KCRV.

The data set used for the evaluation of the KCRVs is known as the KCRV file and is a reduced data set from the SIR master-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 made only by the CCRI(II) during one of its biennial meetings, or by consensus through electronic means (e.g., email) as discussed at the CCRI(II) meeting in 2013.

The ampoules containing an activity significantly smaller than the minimum required for the SIR were not selected for the KCRV. See Table 4. In addition, the  $4\pi(\text{PPC})\text{ce}$  method was not considered as primary for  $^{109}\text{Cd}$ ; the corresponding SIR results were also not selected for the KCRV. Consequently, using the recent result produces an updated KCRV for  $^{109}\text{Cd}$  in 2012 of **8138(26) MBq** with the power  $\alpha = 1.4$  that has been calculated using the previously published results, selected as shown in Table 4, for the NPL (1980), AECL (1982), NMISA (1982), IRA (2000), and the PTB (2004) result. This can be compared with the previous KCRV values of 8136(44) MBq published in 2003 [3].

#### 4.2. Degrees of equivalence

Every participant in a comparison is entitled to have one result included in the KCDB as long as the NMI is a signatory or designated institute listed in the CIPM MRA and the result is valid (i.e., not older than 20 years). Normally, the most recent result is the one included. An NMI may withdraw its result only if all other participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the KCRV [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 result with the KCRV

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

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

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

$$U_i = 2u(D_i) \quad (2)$$

When the result of the NMI  $i$  is included in the KCRV with a weight  $w_i$ , then

$$u^2(D_i) = (1 - 2w_i)u_i^2 + u^2(\text{KCRV}) \quad (3)$$

However, when the result of the NMI  $i$  is not included in the KCRV, then

$$u^2(D_i) = u_i^2 + u^2(\text{KCRV}) \quad (4)$$

#### 4.2.2. Comparison between pairs of NMI results

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

$$D_{ij} = D_i - D_j = A_{ei} - A_{ej} \quad (5)$$

and the expanded uncertainty ( $k = 2$ ) of this difference,  $U_{ij} = 2u(D_{ij})$ , where

$$u^2(D_{ij}) = u_i^2 + u_j^2 - 2u(A_{ei}, A_{ej}) \quad (6)$$

where any obvious correlations between the NMIs (such as a traceable calibration, or correlations normally coming from the SIR or from the linking factor in the case of linked comparison) are subtracted using the covariance  $u(A_{ei}, A_{ej})$  (see [15] for more detail). However, the CCRI decided in 2011 that these pair-wise degrees of equivalence no longer need to be published as long as the methodology is explained.

Table B1 shows the matrix of all the degrees of equivalence as they will appear in 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 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 C1. This graphical representation indicates in part the degree of equivalence between the NMIs but obviously does not take into account the correlations between the different NMIs. It should be noted that the final data in this paper, while correct at the time of publication, will become out-of-date as NMIs make new comparisons. The formal results under the CIPM MRA [2] are those available in the KCDB.

## 5. Conclusion

The BIPM continuous key comparison for  $^{109}\text{Cd}$ , BIPM.RI(II)-K1.Cd-109, currently comprises 4 results. The results have been analyzed with respect to the updated KCRV, providing degrees of equivalence for 4 national metrology institutes. The degrees of equivalence have been approved by the CCRI(II) and are published in the BIPM key comparison database. Other results may be added when other NMIs contribute  $^{109}\text{Cd}$  activity measurements to this comparison or take part in other linked comparisons.

## 6. References

- [1] Ratel, G. The Système International de Référence and its application in key comparisons, *Metrologia*, 2007, **44**(4), S7-S16.
- [2] CIPM MRA: *Mutual recognition of national measurement standards and of calibration and measurement certificates issued by national metrology institutes*, International Committee for Weights and Measures, 1999, pp. 45.
- [3] Ratel G., Michotte C., BIPM comparison BIPM.RI(II)-K1.Cd-109 of activity measurements of the radionuclide  $^{109}\text{Cd}$  and links for the 1986 international comparison CCRI(II)-K2.Cd-109, *Metrologia*, 2003, **40**, Tech. Suppl., 06019.
- [4] Ratel G., Michotte C., Janssen, Kossert K., Lucas L. and Karam L., Activity measurements of the radionuclide  $^{109}\text{Cd}$  for the PTB, Germany and the NIST, USA in the ongoing comparison BIPM.RI(II)-K1.Cd-109, *Metrologia*, 2005, **42**, Tech. Suppl., 06011.
- [5] Bé M.-M., Chisté V., Dulieu C., Kellett M. A., Mougeot X., Arinc A., Chechev V.P., Kuzmenko N.K., Kibédi T., Luca A., Nichols A.L., *Table of radionuclides*, Monographie BIPM-5, 2016, Vol 8.
- [6] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Helmer R., Nichols A., Schönfeld E., Dersch R., *Table of radionuclides*, Monographie BIPM-5, 2004, Vol. 1.
- [7] NBS Radioactivity Section, Priv. Comm. (Nov. 1976).
- [8] Steyn J., Standardization of Cadmium-109, *CSIR Research Report* **281**, 1969.
- [9] Meyer B.R., Steyn J. and Conard F.M.P., Standardization of Cd-109 revisited, *CSIR Research Report* **588**, 1983.
- [10] Bé M.-M., Chisté V., Dulieu C., Browne E., Chechev V., Kuzmenko N., Helmer R., Nichols A., Schönfeld E., Dersch R., *Table of radionuclides*, Monographie BIPM-5, 2004, Vol 2.
- [11] Cox M.G., Michotte C., Pearce A.K., Measurement modelling of the International Reference System (IRS) for gamma-emitting radionuclides, *Monographie BIPM-7*, 2007, 48 pp.
- [12] Michotte C., Ratel G., Courte S., Dzeil T., Listkowska A., Csete I., Rozsa K., Szücs L., Zsinka A., Fréchou C., Bobin C. and Pierre S., Update of the BIPM comparison BIPM.RI(II)-K1.Am-241 of activity measurements of the radionuclide  $^{241}\text{Am}$  to include the 2009 results of the POLATOM (Poland), MKEH (Hungary) and the 2011 results of the LNE-LNHB (France), *Metrologia*, 2017, **54**, Tech. Suppl., 06010.
- [13] Woods M.J., Reher D.F.G. and Ratel G., Equivalence in radionuclide metrology, *Applied Radiation and Isotopes*, 2000, **52**(3), 313-318.
- [14] Pommé S. and Keightley J., Determination of a reference value and its uncertainty through a power-moderated mean, *Metrologia*, 2015, **52**(3), S200.
- [15] Michotte C. and Ratel G., Correlations taken into account in the KCDB, CCRI(II) working document, 2003, *CCRI(II)/03-29*.

## Appendix A. Introductory text for $^{109}\text{Cd}$ degrees of equivalence

### Key comparison BIPM.RI(II)-K1.Cd-109

#### MEASURAND: Equivalent activity of $^{109}\text{Cd}$

**Key comparison reference value:** the SIR reference value  $x_{\text{R}}$  for this radionuclide is 8138 MBq, with a standard uncertainty,  $u_{\text{R}}$  equal to 26 MBq (see Section 4.1 of the Final Report). The value  $x_i$  is taken as the equivalent activity for a laboratory  $i$ .

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms:  $D_i = (x_i - x_{\text{R}})$  and  $U_i$ , its expanded uncertainty ( $k = 2$ ), both expressed in MBq, and  $U_i = 2((1 - 2w_i)u_i^2 + u_{\text{R}}^2)^{1/2}$ , where  $w_i$  is the weight of laboratory  $i$  contributing to the calculation of  $x_{\text{R}}$ .

**Appendix B. Table of degrees of equivalence for BIPM.RI(II)-K1.Cd-109**

Table B1: The table of degrees of equivalence for BIPM.RI(II)-K1.Cd-109

| NMI $i$  | $D_i$ /MBq | $U_i$ /MBq |
|----------|------------|------------|
| IRA      | -73        | 90         |
| PTB      | 32         | 58         |
| NIST     | 30         | 100        |
| LNE-LNHB | -100       | 160        |

Appendix C. Graph of degrees of equivalence with the KCRV for  $^{109}\text{Cd}$  (as it appears in Appendix B of the MRA)

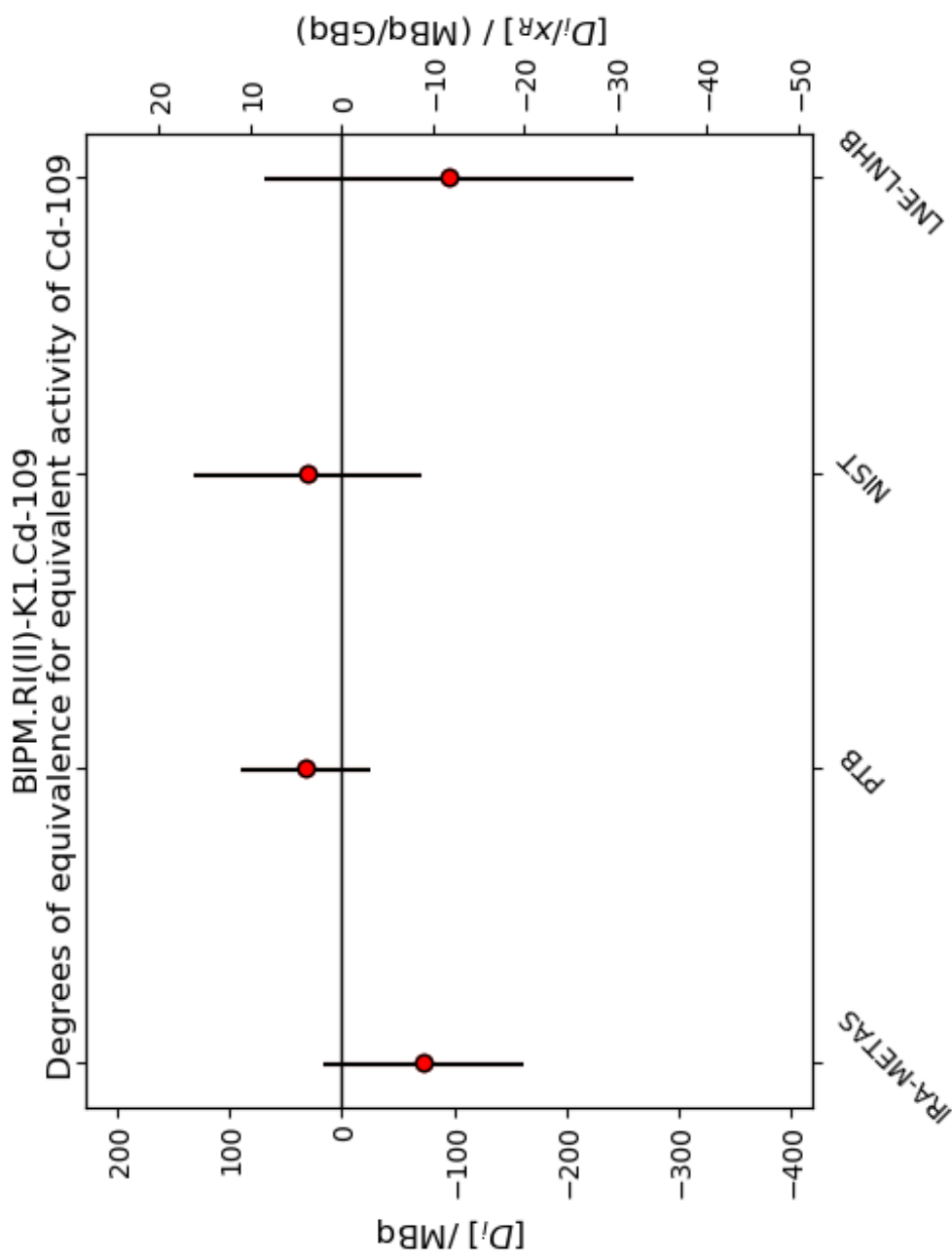


Figure C1. Degrees of equivalence for equivalent activity of  $^{109}\text{Cd}$ .

**Appendix D. Uncertainty budgets for the activity of  $^{109}\text{Cd}$  submitted to the SIR**

### Detailed Uncertainty Budget

Laboratory: **LNE-LNHB**; Radionuclide:  **$^{109}\text{Cd}$**  ; Ampoule number: **3**

Uncertainty components\*, in % of the activity concentration, due to

|  |             | Remarks                            | Evaluation<br>type (A or B) | Relative<br>Sensitivity<br>Factor |
|--|-------------|------------------------------------|-----------------------------|-----------------------------------|
| counting statistics  | <b>0.3</b>  | <b>10 sources measured</b>         | <b>B</b>                    | -----                             |
| weighing   | <b>0.1</b>  | <b>Pycnometer technique</b>        | <b>B</b>                    | -----                             |
| dead time  | <b>0.01</b> | <b>Live-time technique</b>         | <b>B</b>                    | -----                             |
| background   | <b>0.05</b> | -----                              | <b>A</b>                    | -----                             |
| pile-up  | -----       | -----                              | -----                       | -----                             |
| counting time  | -----       | -----                              | -----                       | -----                             |
| adsorption   | -----       | -----                              | -----                       | -----                             |
| impurities   | -----       | -----                              | -----                       | -----                             |
| tracer   | -----       | -----                              | -----                       | -----                             |
| input parameters<br>and statistical model                                  | -----       | -----                              | -----                       | -----                             |
| quenching  | -----       | -----                              | -----                       | -----                             |
| interpolation from<br>calibration curve                                    | -----       | -----                              | -----                       | -----                             |
| <b>Detection efficiency</b>  | <b>0.2</b>  | <b>GEANT4 MC code</b>              | <b>B</b>                    | -----                             |
| decay-scheme parameters  | <b>0.7</b>  | -----                              | <b>B</b>                    | -----                             |
| half life ( $T_{1/2}$ : <b>461.4 (12) d</b> )                              | <b>0.01</b> | <b>Decay correction</b>            | <b>B</b>                    | -----                             |
| self absorption  | -----       | -----                              | -----                       | -----                             |
| extrapolation of efficiency<br>curve                                       | <b>0.5</b>  | <b>Detection threshold setting</b> | <b>B</b>                    | -----                             |
| other effects (if relevant)<br>(geometry factor, retrodiffusion)           | -----       | -----                              | <b>B</b>                    | -----                             |
| combined uncertainty<br>( as quadratic sum of all<br>uncertainty component | <b>0.94</b> | -----                              | -----                       | -----                             |

\* The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, 17, 73 and *Guide to expression of uncertainty in measurement*, ISO, corrected and reprinted 1995).



### Appendix E. 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 |
|-----------------------|---------|-------------------------------|---------|
| $4\pi$                | 4P      | proportional counter          | PC      |
| defined solid angle   | SA      | press. Prop. Counter          | PP      |
| $2\pi$                | 2P      | liquid scintillation counting | LS      |
| undefined solid angle | UA      | NaI(Tl)                       | NA      |
|                       |         | Ge(HP)                        | GH      |
|                       |         | Ge(Li)                        | GL      |
|                       |         | Si(Li)                        | SL      |
|                       |         | CsI(Tl)                       | CS      |
|                       |         | ionization chamber            | IC      |
|                       |         | grid ionization chamber       | GC      |
|                       |         | Cerenkov detector             | CD      |
|                       |         | calorimeter                   | CA      |
|                       |         | solid plastic scintillator    | SP      |
|                       |         | PIPS detector                 | PS      |
|                       |         | CeBr3                         | CB      |

| 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      |
| mixed electrons              | ME      | coincidence                                       | CO      |
| bremsstrahlung               | BS      | anti-coincidence                                  | AC      |
| gamma rays                   | GR      | coincidence counting with efficiency tracing      | CT      |
| x-rays                       | XR      | anti-coincidence counting with efficiency tracing | AT      |
| photons ( $x + \gamma$ )     | PH      | triple-to-double coincidence ratio counting       | TD      |
| photons + electrons          | PE      | selective sampling                                | SS      |
| alpha particle               | AP      | high efficiency                                   | HE      |
| mixture of various radiation | MX      | digital coincidence counting                      | DC      |

| <b>Examples of methods</b>  | <b>acronym</b>    |
|---|-------------------|
| $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence counting                           | 4P-PC-BP-NA-GR-CO |
| $4\pi(\text{PPC})\beta\text{-}\gamma$ coincidence counting<br>eff. trac             | 4P-PP-MX-NA-GR-CT |
| defined solid angle $\alpha$ -particle<br>counting with a PIPS detector             | SA-PS-AP-00-00-00 |
| $4\pi(\text{PPC})\text{AX-}\gamma(\text{GeHP})\text{-}$<br>anticoincidence counting | 4P-PP-MX-GH-GR-AC |
| $4\pi\text{CsI-}\beta,\text{AX},\gamma$ counting                                    | 4P-CS-MX-00-00-HE |
| calibrated IC   | 4P-IC-GR-00-00-00 |
| internal gas counting   | 4P-PC-BP-00-00-IG |