

**CCRI comparison CCRI(II)-K2.I-125 of  
activity measurements of the radionuclide  $^{125}\text{I}$**

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**Abstract**

In 1988, an international comparison of  $^{125}\text{I}$  activity measurements was organized by the Bureau International des Poids et Mesures for the Consultative Committee for Ionizing Radiation Section II (CCRI(II)). Nineteen laboratories took part in this comparison and the details and results have been published elsewhere. The degrees of equivalence between each laboratory and the key comparison reference value (KCRV) have now been calculated and the results are given in the form of a matrix for sixteen laboratories. A graphical presentation is also given. The results of this comparison have been approved by Section II of the (CCRI(II)), comparison identifier CCRI(II)-K2.I-125.

**1. Introduction**

In 1988, an international comparison of  $^{125}\text{I}$  activity measurements was organized by the Bureau International des Poids et Mesures for the Consultative Committee for Ionizing Radiation Section II (CCRI(II)). Nineteen laboratories took part in this comparison and the details and results have been published elsewhere [1].

These results have been analyzed in accordance with the Mutual Recognition Arrangement (MRA) [2] so that they may be included in the BIPM key comparison database (KCDB).

**2. Participants**

Sixteen NMIs and three other laboratories took part in the comparison of  $^{125}\text{I}$  activity measurements in 1988. The laboratory details are given in Table 1. In cases where the laboratory has changed its name since the comparison, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB.

However, it should be noted that the AECL is not a designated laboratory of the NRC, Canada, and Indonesia (P3KRBiN) has not signed the MRA, therefore neither of these results is included in the KCDB. In addition, the KRIS result has been excluded from the KCDB with the agreement of the participants as a more recent standard is disseminated currently.

**Table 1. Details of the participants in the CCRI(II)-K2.I-125 comparison**

| <b>Original acronym</b> | <b>NMI</b> | <b>Full name</b>  | <b>Country</b>    | <b>Regional metrology organization</b> |
|-------------------------|------------|---|-------------------|--|
| AECL                    | –          | Atomic Energy of Canada Ltd   | Canada            | –                                      |
| BIPM                    | –          | Bureau International des Poids et Mesures   | –                 | –                                      |
| LMRI                    | BNM-LNHB   | Bureau national de métrologie-Laboratoire national Henri Becquerel                            | France            | EUROMET                                |
| UVVVR                   | CMI-IIR    | Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation | Czech Republic    | EUROMET                                |
| NAC*                    | CSIR-NML   | National Metrology Laboratory   | South Africa      | SADCMET                                |
| –                       | ENEA       | Ente per le Nuove Tecnologie, l'Energia e l'Ambiente  | Italy             | EUROMET                                |
| IRMM (CBNM)             | –          | Institute for Reference Materials and Measurements  | European Union    | EUROMET                                |
| KSRI                    | KRISS      | Korea Research Institute of Standards and Science   | Republic of Korea | APMP                                   |
| CNEN                    | LNMRI      | Laboratorio Nacional de Metrologia das Radiações Ionizantes                                   | Brazil            | SIM                                    |
| –                       | NIM        | National Institute of Metrology   | China             | APMP                                   |
| –                       | NIST       | National Institute of Standards and Technology  | United States     | SIM                                    |
| ETL                     | NMIJ       | National Metrology Institute of Japan   | Japan             | APMP                                   |
| –                       | NPL        | National Physical Laboratory  | United Kingdom    | EUROMET                                |
| –                       | NRC        | National Research Council   | Canada            | SIM                                    |
| –                       | OMH        | Országos Mérésügyi Hivatal  | Hungary           | EUROMET                                |
| PSPKR                   | P3KRBiN    | Pusat Penelitian & Pengembangan Keselamatan Radiasi & Biomedika Nuklir                        | Indonesia         | APMP                                   |
| IEA                     | RC         | Radioisotope Centre POLATOM   | Poland            | EUROMET                                |

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**Table 1 continued. Details of the participants in the CCRI(II)-K2.I-125 comparison**

| Original acronym | NMI   | Full name                              | Country            | Regional metrology organization |
|------------------|-------|--|--------------------|---------------------------------|
| –                | PTB   | Physikalisch-Technische Bundesanstalt  | Germany            | EUROMET                         |
| –                | VNIIM | D.I. Mendeleev Institute for Metrology | Russian Federation | COOMET                          |

\* another laboratory in the country

### 3. The comparison solution and NMI standardization methods

Each participant received a flame-sealed NBS-type ampoule containing a known mass of about 3.6 g of solution. The solution had a nominal activity concentration of 2.0 MBq g<sup>-1</sup> at the reference date of 1988-06-15 0 h UT. The aqueous solution consisted of  $5 \times 10^{-4}$  mol NaOH per dm<sup>3</sup>, with a carrier concentration of 50 µg as KI and 50 µg of Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> per gram of solution. Gamma-ray spectroscopy using Ge detectors identified that there was no <sup>126</sup>I contamination of the solution within the detection limit of some parts in 10<sup>-7</sup>.

The participants used eight different methods to measure the activity of the samples. Some laboratories used up to four different methods. Further details are given in [3] and [1, 4] in which the published references for the methods of each laboratory are also given. The list of acronyms in Table 2 used to summarize the methods is given in Appendix 2.

All measurements were made between May 3 and August 29 1988. Most laboratories measured close to the reference date in June to minimize the effect of the uncertainty in the half-life on the submitted results.

Four laboratories determined the half-life of <sup>125</sup>I during the comparison. A mean value deduced from these results is 59.37 (4) days [1] and this agrees well with the value of 59.4 (5) days [5] used by the participants. It is interesting to note that the value recommended by the IAEA since 1991 is 59.43 (6) days [6].

### 4. Results

The results [1] of the comparison are given in Table 2. Comments on the results were made at the time of the comparison and are given in [1 and 4]. The full uncertainty budgets are also given in [3, 4].

Although, normally, the degrees of equivalence would be linked to the International Reference System (SIR) for activity measurements, the low energy of the photons emitted by this radionuclide mitigate against the use of the SIR measurements for equivalence. (The SIR results are published in [7]). Consequently, the CCRI(II) has

decided that this comparison should stand alone in the KCDB until such time as a new comparison is made.

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

**Table 2. Results of the 1988 international comparison measurements of  $^{125}\text{I}$**

| NMI      | Measurement method and acronym (see Appendix 2) | Activity concentration measured $(A/m)_i$ / (kBq g <sup>-1</sup> ) | standard uncertainty $u_i$ / (kBq g <sup>-1</sup> ) | Mean activity concentration <sup>†</sup> $(A/m)_i$ / (kBq g <sup>-1</sup> ) | combined standard uncertainty <sup>†</sup> $u_{c,i}$ / (kBq g <sup>-1</sup> ) |
|----------|---|--|---|---|---|
| AECL     | UA-GH-MX-00-00-SC                               | 1413.35  | 2.54  | 1412.8  | 0.6   |
|          | UA-NA-MX-NA-MX-CO                               | 1413.85  | 0.78  |   |   |
|          | 4P-PC-CE-NA-MX-CO                               | 1412.05  | 0.65  |   |   |
| BIPM     | UA-NA-MX-00-00-SC                               | 1425.1   | 7.21  | 1421.9  | 4.0   |
|          | UA-NA-MX-NA-MX-CO                               | 1420.58  | 4.74  |   |   |
| BNM-LNHB | UA-NA-MX-NA-MX-CO                               | 1435.7   | 1.6   | 1435.7  | 1.6   |
| CMI-IIR  | 4P-NA-MX-00-00-SC                               | 1424.9   | 1.3   | 1428.5  | 2.9   |
|          | 4P-NA-MX-NA-MX-CO                               | 1429.95  | 0.35  |   |   |
| CSIR-NML | UA-NA-MX-00-00-SC                               | 1425.0   | 4.4   | 1435.0  | 2.4   |
|          | UA-NA-MX-NA-MX-CO                               | 1436.45  | 2.09  |   |   |
|          | UA-NA-MX-NA-MX-CO <sup>‡</sup>                  | 1434.83  | 2.29  |   |   |
|          | 4P-LS-MX-NA-MX-CO                               | 1447.6   | 7.6   |   |   |
| ENEA     | 4P-NA-MX-00-00-SC                               | 1446.9   | 3.8   | 1446.9  | 3.8   |
| IRMM     | 4P-NA-MX-00-00-SC                               | 1422.3   | 7.0   | 1430.2  | 4.4   |
|          | UA-NA-MX-NA-MX-CO                               | 1425.8   | 7.7   |   |   |
|          | UA-NA-MX-NA-MX-CO <sup>‡</sup>                  | 1443.2   | 6.3   |   |   |
|          | 4P-CS-MX-00-00-00                               | 1427.9   | 4.9   |   |   |
| KRISS    | UA-NA-MX-NA-MX-CO <sup>‡</sup>                  | 1358.0   | 7.6   | 1358.0  | 7.6   |
| LNMRI    | UA-NA-MX-00-00-SC                               | 1438.9   | 5.9   | 1434.2  | 2.8   |
|          | UA-NA-MX-NA-MX-CO                               | 1434.1   | 4.0   |   |   |
|          | UA-NA-MX-NA-MX-CO <sup>‡</sup>                  | 1430.8   | 5.2   |   |   |
| NIM      | UA-NA-MX-NA-MX-CO                               | 1433.0   | 9.0   | 1430.7  | 4.4   |
|          |   | 1430.0*  | 5.0   |   |   |
| NIST     | 4P-NA-MX-00-00-SC                               | 1436.0   | 3.9   | 1433.4  | 3.4   |
|          | UA-NA-MX-00-00-SC                               | 1429.0   | 5.0   |   |   |

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**Table 2 continued. Results of the 1988 international comparison measurements of  $^{125}\text{I}$** 

| NMI     | Measurement method and acronym (see Appendix 2) | Activity concentration measured $(A/m)_i$ / (kBq g <sup>-1</sup> ) | standard uncertainty $u_i$ / (kBq g <sup>-1</sup> ) | Mean activity concentration <sup>†</sup> $(A/m)_i$ / (kBq g <sup>-1</sup> ) | combined standard uncertainty <sup>†</sup> $u_{c,i}$ / (kBq g <sup>-1</sup> ) |
|---------|---|--|---|---|---|
| NMIJ    | 4P-NA-MX-00-00-SC                               | 1445.2   | 5.4   | 1448.8  | 6.6   |
|         | 4P-PC-CE-NA-MX-CO                               | 1461.0   | 10.0  |   |   |
| NPL     | 4P-PC-CE-GH-MX-CO                               | 1419.6   | 4.2   | 1419.6  | 4.2   |
| NRC     | 4P-PC-CE-NA-MX-AC                               | 1438.23  | 5.78  | 1434.8  | 4.1   |
|         | 4P-PC-CE-SL-MX-CO                               | 1431.54  | 5.71  |   |   |
| OMH     | 4P-NA-MX-00-00-SC                               | 1438.7   | 2.4   | 1438.7  | 2.1   |
|         |   | 1438.9 <sup>†</sup>  | 4.3   |   |   |
| P3KRBiN | 4P-NA-MX-00-00-SC                               | 1440.0   | 20.0  | 1435.6  | 6.7   |
|         | 4P-NA-MX-NA-MX-CO                               | 1440.0   | 10.0  |   |   |
|         | 4P-NA-MX-NA-MX-CO <sup>‡</sup>                  | 1430.0   | 10.0  |   |   |
| RC      | UA-NA-MX-NA-MX-CO                               | 1421.0   | 3.0   | 1421.0  | 3.0   |
| PTB     | UA-SL-MX-00-00-SC                               | 1429.0   | 11.0  | 1427.3  | 4.6   |
|         | UA-NA-MX-NA-MX-CO <sup>*</sup>                  | 1427.0   | 5.0   |   |   |
| VNIIM   | UA-NA-MX-NA-MX-CO                               | 1428.6   | 4.9   | 1432.2  | 5.7   |
|         | 4P-PC-CE-NA-MX-CO                               | 1441.2   | 7.7   |   |   |

<sup>†</sup> when two or more  $A/m$  values are given, a weighted mean and corresponding uncertainty has been attributed to an individual entry [1]

\* using different gate conditions

<sup>†</sup> an additional measurement further in time from the reference date

<sup>‡</sup> with efficiency extrapolation.

#### 4.1 The key comparison reference value

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

- only primary standardized solutions are accepted, or ionization chamber measurements that are directly traceable to a primary measurement in the laboratory;
- each NMI or other laboratory has only one result (normally the mean if more than one result is submitted);
- 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).

Consequently, the KCRV for  $^{125}\text{I}$  has been identified as 1431.5 (2.1) kBq g $^{-1}$  using the mean result of each laboratory except that of the KRISS.

#### 4.2 Degrees of equivalence

Every NMI that participates in a CCRI(II) comparison 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. However, an NMI may withdraw its result but only if all the participants agree. In view of the subsequent (2002) submission to the SIR of the KRISS, that agrees with the weighted mean of the SIR to within  $1.5 \times 10^{-2}$ , the participants have agreed that the 1988 KRISS result is excluded from the KCDB as this earlier standard is no longer disseminated. Results from a bilateral comparison undertaken by the KRISS, or from their participation in a regional or other international comparison for this radionuclide, will be included in any future evaluation for degrees of equivalence.

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 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

$$D_i = (A/m)_i - \text{KCRV} \quad (1)$$

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 (see Appendix 1).

##### 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

$$D_{ij} = D_i - D_j = (A/m)_i - (A/m)_j \quad (3)$$

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,  $f_k$ .

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 3 shows the matrix of all the degrees of equivalence as they 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  $(A/m)_i$  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 CCRI(II) key comparison for  $^{125}\text{I}$ , CCRI(II)-K2.I-125 comprises sixteen 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. These results will be updated when the CCRI(II) runs another activity comparison for this radionuclide.

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## References

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Table 3. Table of degrees of equivalence and introductory text for <sup>125</sup>I

Key comparison CCR(I)-K2.I-125

MEASURAND : Activity concentration of <sup>125</sup>I

Key comparison reference value: the reference value for this comparison is  $x_R = 1431.5 \text{ kBq g}^{-1}$  with a standard uncertainty,  $u_R = 2.1 \text{ kBq g}^{-1}$  (see Section 4.1 of the Final Report). The value  $x_i$  is the activity concentration 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  $\text{kBq g}^{-1}$ , and  $U_i = 2((1 - 2/n)u_i^2 + (1/n^2)\sum u_j^2)^{1/2}$  when each laboratory has contributed to the calculation of  $x_R$ , with  $n$  the number of laboratories in the KCRV.

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  $\text{kBq g}^{-1}$ .

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

Lab  $j$   $\longrightarrow$

Lab  $i$   $\Downarrow$

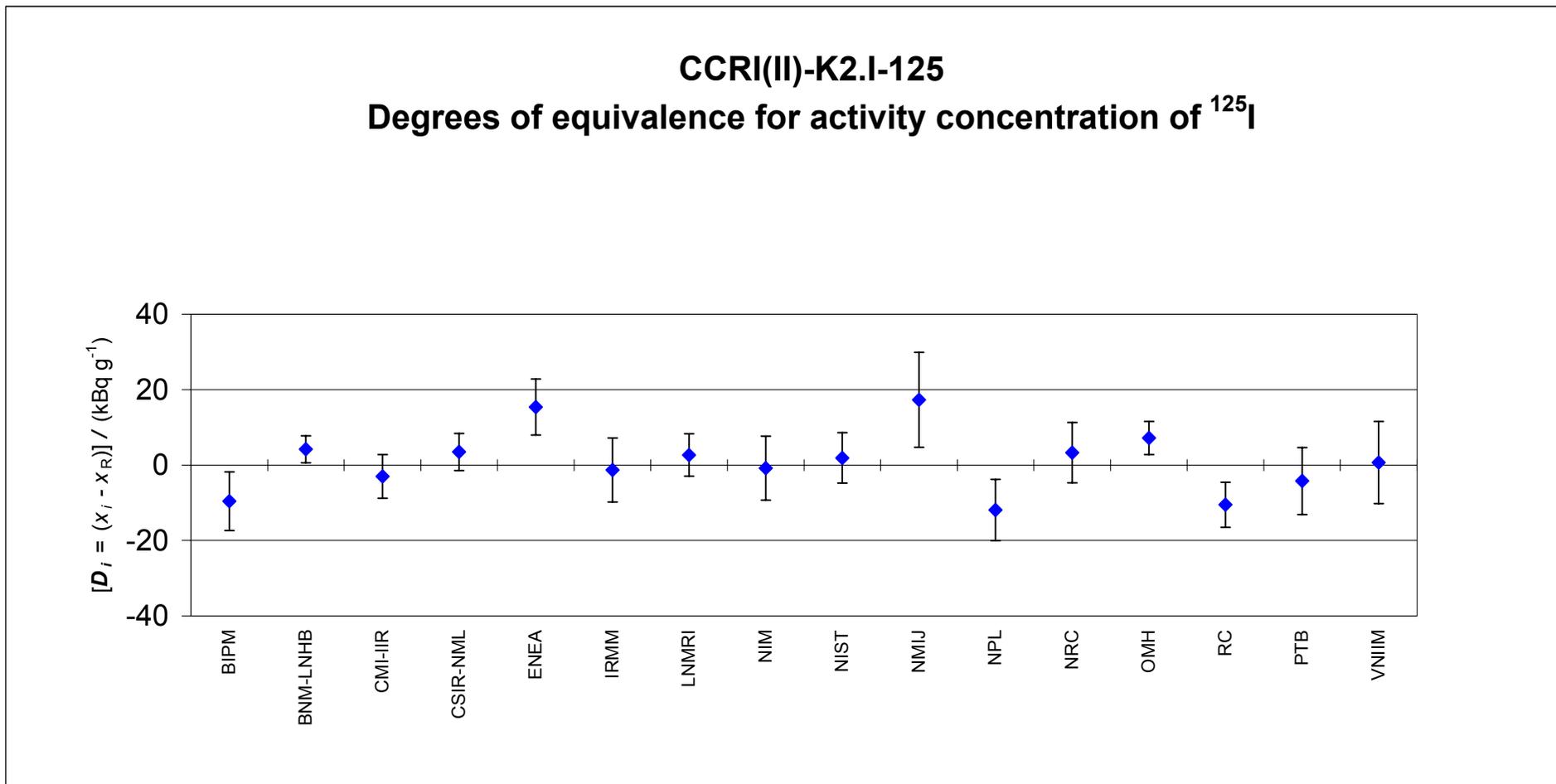
|          | $D_i$ $U_i$<br>/ $(\text{kBq g}^{-1})$ |       | BIPM     |          | BNM-LNHB |          | CMI-IIR  |          | CSIR-NML |          | ENEA     |          | IRMM     |          | LNMRI    |          | NIM      |          |
|----------|--|-------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
|          | $D_i$                                  | $U_i$ | $D_{ij}$ | $U_{ij}$ |
| BIPM     | -10                                    | 8     |          |          |          |          |          |          |          |          |          |          |          |          |          |          |          |          |
| BNM-LNHB | 4                                      | 4     | 14       | 9        |          |          |          |          |          |          |          |          |          |          |          |          |          |          |
| CMI-IIR  | -3                                     | 6     | 7        | 10       | -7       | 7        |          |          |          |          |          |          |          |          |          |          |          |          |
| CSIR-NML | 3                                      | 5     | 13       | 9        | -1       | 6        | 7        | 8        |          |          |          |          |          |          |          |          |          |          |
| ENEA     | 15                                     | 7     | 25       | 11       | 11       | 8        | 18       | 10       | 12       | 9        |          |          |          |          |          |          |          |          |
| IRMM     | -1                                     | 9     | 8        | 12       | -6       | 9        | 2        | 11       | -5       | 10       | -17      | 12       |          |          |          |          |          |          |
| LNMRI    | 3                                      | 6     | 12       | 10       | -2       | 6        | 6        | 8        | -1       | 7        | -13      | 9        | 4        | 10       |          |          |          |          |
| NIM      | -1                                     | 9     | 9        | 12       | -5       | 9        | 2        | 11       | -4       | 10       | -16      | 12       | 1        | 12       | -4       | 10       |          |          |
| NIST     | 2                                      | 7     | 12       | 10       | -2       | 8        | 5        | 9        | -2       | 8        | -14      | 10       | 3        | 11       | -1       | 9        | 3        | 11       |
| NMIJ     | 17                                     | 13    | 27       | 15       | 13       | 14       | 20       | 14       | 14       | 14       | 2        | 15       | 19       | 16       | 15       | 14       | 18       | 16       |
| NPL      | -12                                    | 8     | -2       | 12       | -16      | 9        | -9       | 10       | -15      | 10       | -27      | 11       | -11      | 12       | -15      | 10       | -11      | 12       |
| NRC      | 3                                      | 8     | 13       | 11       | -1       | 9        | 6        | 10       | 0        | 10       | -12      | 11       | 5        | 12       | 1        | 10       | 4        | 12       |
| OMH      | 7                                      | 4     | 17       | 9        | 3        | 5        | 10       | 7        | 4        | 6        | -8       | 9        | 9        | 10       | 5        | 7        | 8        | 10       |
| RC       | -11                                    | 6     | -1       | 10       | -15      | 7        | -8       | 8        | -14      | 8        | -26      | 10       | -9       | 11       | -13      | 8        | -10      | 11       |
| PTB      | -4                                     | 9     | 5        | 12       | -8       | 10       | -1       | 11       | -8       | 10       | -20      | 12       | -3       | 13       | -7       | 11       | -3       | 13       |
| VNIIM    | 1                                      | 11    | 10       | 14       | -4       | 12       | 4        | 13       | -3       | 12       | -15      | 14       | 2        | 14       | -2       | 13       | 2        | 14       |

Lab  $j$   $\longrightarrow$

Lab  $i$   $\Downarrow$

|          | $D_i$ $U_i$<br>/ $(\text{kBq g}^{-1})$ |       | NIST     |          | NMIJ     |          | NPL      |          | NRC      |          | OMH      |          | RC       |          | PTB      |          | VNIIM    |          |
|----------|--|-------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
|          | $D_i$                                  | $U_i$ | $D_{ij}$ | $U_{ij}$ |
| BIPM     | -10                                    | 8     | -12      | 10       | -27      | 15       | 2        | 12       | -13      | 11       | -17      | 9        | 1        | 10       | -5       | 12       | -10      | 14       |
| BNM-LNHB | 4                                      | 4     | 2        | 8        | -13      | 14       | 16       | 9        | 1        | 9        | -3       | 5        | 15       | 7        | 8        | 10       | 4        | 12       |
| CMI-IIR  | -3                                     | 6     | -5       | 9        | -20      | 14       | 9        | 10       | -6       | 10       | -10      | 7        | 8        | 8        | 1        | 11       | -4       | 13       |
| CSIR-NML | 3                                      | 5     | 2        | 8        | -14      | 14       | 15       | 10       | 0        | 10       | -4       | 6        | 14       | 8        | 8        | 10       | 3        | 12       |
| ENEA     | 15                                     | 7     | 14       | 10       | -2       | 15       | 27       | 11       | 12       | 11       | 8        | 9        | 26       | 10       | 20       | 12       | 15       | 14       |
| IRMM     | -1                                     | 9     | -3       | 11       | -19      | 16       | 11       | 12       | -5       | 12       | -9       | 10       | 9        | 11       | 3        | 13       | -2       | 14       |
| LNMRI    | 3                                      | 6     | 1        | 9        | -15      | 14       | 15       | 10       | -1       | 10       | -5       | 7        | 13       | 8        | 7        | 11       | 2        | 13       |
| NIM      | -1                                     | 9     | -3       | 11       | -18      | 16       | 11       | 12       | -4       | 12       | -8       | 10       | 10       | 11       | 3        | 13       | -2       | 14       |
| NIST     | 2                                      | 7     |          |          | -15      | 15       | 14       | 11       | -1       | 11       | -5       | 8        | 12       | 9        | 6        | 11       | 1        | 13       |
| NMIJ     | 17                                     | 13    | 15       | 15       |          |          | 29       | 16       | 14       | 16       | 10       | 14       | 28       | 14       | 22       | 16       | 17       | 17       |
| NPL      | -12                                    | 8     | -14      | 11       | -29      | 16       |          |          | -15      | 12       | -19      | 9        | -1       | 10       | -8       | 12       | -13      | 14       |
| NRC      | 3                                      | 8     | 1        | 11       | -14      | 16       | 15       | 12       |          |          | -4       | 9        | 14       | 10       | 8        | 12       | 3        | 14       |
| OMH      | 7                                      | 4     | 5        | 8        | -10      | 14       | 19       | 9        | 4        | 9        |          |          | 18       | 7        | 11       | 10       | 7        | 12       |
| RC       | -11                                    | 6     | -12      | 9        | -28      | 14       | 1        | 10       | -14      | 10       | -18      | 7        |          |          | -6       | 11       | -11      | 13       |
| PTB      | -4                                     | 9     | -6       | 11       | -22      | 16       | 8        | 12       | -8       | 12       | -11      | 10       | 6        | 11       |          |          | -5       | 15       |
| VNIIM    | 1                                      | 11    | -1       | 13       | -17      | 17       | 13       | 14       | -3       | 14       | -7       | 12       | 11       | 13       | 5        | 15       |          |          |

Figure 1. Graph of degrees of equivalence with the KCRV for  $^{125}\text{I}$   
(as it appears in Appendix B of the MRA)



## Appendix 1. Evaluation of the uncertainty of the degree of equivalence

Table 3 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 activity concentration  $(A/m)_i$  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 results for this radionuclide. Briefly at least four situations can occur depending on the consistency of the experimental data set :

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 activity concentrations  $(A/m)_i$  are not correlated. For the sake of coherence with the definition of the variables used in the text, the following notation is used :

$x_i = (A/m)_i$  and  $u_i = u_{(A/m)_i}$  its uncertainty.

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

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

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

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

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 3.

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 2. 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                           | 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      |
| bremstrahlung                                 | 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           |
|--|--------|-------------------|
| $4\pi$ (PC) $\beta$ - $\gamma$ -coincidence counting                 |        | 4P-PC-BP-NA-GR-CO |
| $4\pi$ (PPC) $\beta$ - $\gamma$ -coincidence counting eff. trac.     |        | 4P-PP-MX-NA-GR-CT |
| defined solid angle $\alpha$ -particle counting with a PIPS detector |        | SA-PS-AP-00-00-00 |
| $4\pi$ (PPC)AX- $\gamma$ (GeHP)-anticoincidence counting             |        | 4P-PP-MX-GH-GR-AC |
| $4\pi$ CsI- $\beta$ ,AX, $\gamma$ counting                           |        | 4P-CS-MX-00-00-00 |
| calibrated IC  |        | 4P-IC-GR-00-00-00 |
| internal gas counting  |        | 4P-PC-BP-00-00-IG |