# BIPM comparison BIPM.RI(II)-K1.Ce-144 of activity measurements of the radionuclide $^{144}$Ce

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## Abstract

Since 1978, five national metrology institutes and one other laboratory have submitted eight samples of known activity of $^{144}$Ce to the International Reference System (SIR) for activity comparison at the Bureau International des Poids et Mesures. The activities ranged from about 2 MBq to 8 MBq. The degrees of equivalence between each equivalent activity measured in the SIR and the key comparison reference value (KCRV) have been calculated and the results are given in the form of a matrix. A graphical presentation is also given. The results of this comparison have been approved by Section II of the Consultative Committee for Ionizing Radiation (CCRI(II)), comparison identifier BIPM.RI(II)-K1.Ce-144.

## 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 (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 818 ampoules to give 590 independent results for 62 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key comparison database (KCDB) of the Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM.RI(II)-K1.Ce-144 key comparison.

## 2. Participants

Five NMIs and one other laboratory (AECL) have submitted eight ampoules for the comparison of $^{144}$Ce activity measurements since 1978. The laboratory details are given in Table 1. In cases where the laboratory has changed its name since the original submission, both the earlier and the current acronyms are given, as it is the latter that are used in the KCDB. The AECL 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 precursor to the CCRI(II).

**Table 1. Details of the participants in the BIPM.RI(II)-K1.Ce-144**

| Original acronym | NMI | Full name | Country | Regional metrology organization | Date of measurement at the BIPM |
|---|---|---|---|---|---|
| UVVVR | CMI-IIR | Český Metrologický Institut/Czech Metrological Institute, Inspectorate for Ionizing Radiation | Czech Republic | EUROMET | 1978-06-20 |
| – | NPL | National Physical Laboratory | United Kingdom | EUROMET | 1984-06-21 |
| AECL | – | Atomic Energy of Canada Ltd | Canada | – | 1985-12-03 |
| LMRI | BNM-LNHB | Bureau national de métrologie-Laboratoire national Henri Becquerel | France | EUROMET | 1988-10-12 |
| – | NIST | National Institute of Standards and Technology | United States | SIM | 1990-01-23 |
| – | OMH | Országos Mérésügyi Hivatal | Hungary | EUROMET | 1990-06-11 |

## 3. NMI standardization methods

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

A brief description of the standardization methods for each laboratory, the activities submitted and the relative standard uncertainties (*k* = 1) are given in Table 2. Full uncertainty budgets have been requested as part of the comparison protocol only since 1998. Consequently, no uncertainty budgets were provided in these six cases.

The half-life used by the BIPM is 284.6 (0.8) days [3].

**Table 2. Standardization methods of the participants for $^{144}$Ce**

| NMI or laboratory | Method used | Half-life / d | Activity / kBq | Reference date YYYY-MM-DD | Relative standard uncertainty × 100 | |
|---|---|---|---|---|---|---|
| | | | | | Type A | Type B |
| CMI-IIR | 4πβ-γ coincidence | 284.8 | 3246 | 1978-05-23<br>11 h GMT | 0.10 | 0.73 |
| NPL | High pressure ionization chamber calibrated by 4πβ(PC)-γ coincidence | - | 7371 | 1984-06-12<br>8 h UT | 0.04 | 0.50 |
| AECL † | 4πβ-γ coincidence | - | 2989.9<br>2798.0 | 1985-10-01<br>17 h UT | 0.09 | 0.12 |
| BNM-LNHB † | 4πβ-γ coincidence | 285.0 (2) [4] | 6977.9<br>6994.7 | 1988-07-13<br>12 h UT | 0.07 | 0.34 |
| NIST | High pressure ionization chamber calibrated by 4πβ(PPC)-γ anti-coincidence | - | 7683 | 1990-01-09<br>17 h UT | 0.12 | 0.42 |
| OMH | 4πβ-γ coincidence | 285.0 (2) | 4613 | 1990-06-15<br>12 h UT | 0.07 | 0.55 |

† two ampoules submitted (see Table 4)

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

**Table 3. Details of the solution of $^{144}$Ce submitted**

| NMI or laboratory | Chemical composition | Solvent conc. / (mol dm$^{-3}$) | Carrier: conc. /(μg g$^{-1}$) | Density /(g cm$^{-3}$) | Relative activity of impurity† |
|---|---|---|---|---|---|
| CMI-IIR | $CeCl_3$ and $PrCl_3$ in HCl | 0.08 | $CeCl_3$: 20<br>$PrCl_3$ : 20 | – | $^{137}$Cs:<br>0.11 (0.01) % |
| NPL | $CeCl_3$ in HCl | 0.1 | Ce : 63 | 1.001 | – |
| AECL | $CeCl_3$ in HCl | 1 | – | 1.01 | $^{154}$Eu:<br>0.01 (0.01) % |
| BNM-LNHB | $CeCl_3$ and $PrCl_3$ in HCl | 1 | $CeCl_3$: 10<br>$PrCl_3$ : 10 | 1.018 | $< 10^{-5}$ |
| NIST | $CeCl_3$ in HCl | 0.65 | $CeCl_3$: 4680 | 1.0158 | – |
| OMH | $CeCl_3$ in HCl | 0.1 | – | – | – |

† the ratio of the activity of the impurity to the activity of $^{144}$Ce 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 activity measurements for $^{144}$Ce arise from eight ampoules and the SIR equivalent activity for each ampoule, $A_{ei}$, is given in Table 4 for each NMI, $i$. The assumption is made that the daughter radionuclides $^{144}$Pr and $^{144}$Pr$^{m}$ ($T_{1/2}$ of 17.29 (3) min and 7.2 (3) min respectively [4]) were in equilibrium with the parent at the reference dates. The dates of measurement in the SIR are given in Table 1.

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

Measurements repeated at the BIPM after periods of up to one and a half years later produced the same comparison results for the AECL. These measurements confirm the validity of the half-life value used and the evaluation of the impurity corrections.

As no submissions were withdrawn and no recent submission has been identified as a pilot study, the most recent result of each NMI is normally eligible for Appendix B of the MRA. However, the result from the AECL is not included as it is not a designated laboratory of the NRC, Canada.

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

**Table 4. Results of SIR measurements of $^{144}$Ce**

| NMI | Mass of solution /g | Activity submitted/ kBq | N° of Ra source used | SIR $A_e$ /kBq | Relative uncertainty from SIR | Total standard uncertainty $u_i$ / kBq |
|---|---|---|---|---|---|---|
| CMI-IIR | 3.563 56 | 3246 | 1 | 284 100 | $17 \times 10^{-4}$ | 2200 |
| NPL | 3.662 6 | 7371 | 2 | 280 780 | $11 \times 10^{-4}$ | 1430 |
| AECL † | 0.306 06 *<br>0.286 42 * | 2989.9<br>2798.0 | 1 | 279 350<br>283 530‡ | $28 \times 10^{-4}$ | 890<br>910 |
| BNM-LNHB † | 3.665 21<br>3.674 09 | 6977.9<br>6994.7 | 1 | 285 790<br>281 730 | $18 \times 10^{-4}$ | 1100#<br>1100 |
| NIST | 3.656 60 | 7683 | 2 | 284 940 | $10 \times 10^{-4}$ | 1280 |
| OMH | 3.607 1 | 4613 | 1 | 281 850 | $14 \times 10^{-4}$ | 1600 |

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

# the uncertainty (Type B) has been increased from the original estimation of 580 kBq to account for the difference between the two results

* mass of solution before dilution

‡ measurements of the two ampoules at the AECL before submission to the SIR and after being returned 18 months later showed differences in the response of < 0.1 % per mg of undiluted solution

## 4.1 The key comparison reference value

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

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

b) each NMI has only one result (normally the mean if more than one ampoule is submitted);

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

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

The reduced data set used for the evaluation of the KCRVs is known as the KCRV file and is the reduced data set from the SIR mother-file. Although the KCRV may be modified when other NMIs participate, on the advice of the Key Comparison Working Group of the CCRI(II), such modifications are only made by the CCRI(II), normally during one of its biennial meetings.

Consequently, the KCRV for $^{144}$Ce has been identified as 282 810 (680) kBq using the results from the CMI-IIR, NPL, AECL, BNM-LNHB, NIST and the OMH.

## 4.2 Degrees of equivalence

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

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

### 4.2.1 *Comparison of a given NMI with the KCRV*

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

$$D_i = A_{ei} - \mathrm{KCRV} \qquad (1)$$

and the expanded uncertainty ($k = 2$) of this difference, $U_{\mathrm{NMI}}$, known as the equivalence uncertainty, hence

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

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

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

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

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

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

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

Table 5 shows the matrix of all the degrees of equivalence as they will appear in Appendix B of the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with $A_{ei}$ replaced by $x_i$. The introductory text is that agreed for the comparison. The graph of the first column of results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV, 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 correlations into account as appropriate.

## Conclusion

The BIPM ongoing key comparison for $^{144}$Ce, BIPM.RI(II)-K1.Ce-144 currently comprises results from five NMIs. 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 will be published in the BIPM key comparison database. Other results may be added as and when other NMIs contribute $^{144}$Ce activity measurements to this comparison.

## Acknowledgements

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## References

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[8] Woods M.J., Reher D.F.G. and Ratel G., Equivalence in radionuclide metrology, *Applied Radiation and Isotopes*, **52**, (2000) 313-318.

Table 5. Table of degrees of equivalence and introductory text for $^{144}$Ce

Key comparison BIPM.RI(II)-K1.Ce-144

MEASURAND : Equivalent activity of $^{144}$Ce

Key comparison reference value: the SIR reference value for this radionuclide is 282.8 MBq, with an uncertainty $u_R$ of 0.7 MBq.
$x_R$ is computed from the mean of the participants' results including a result from Canada (see the Report)

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

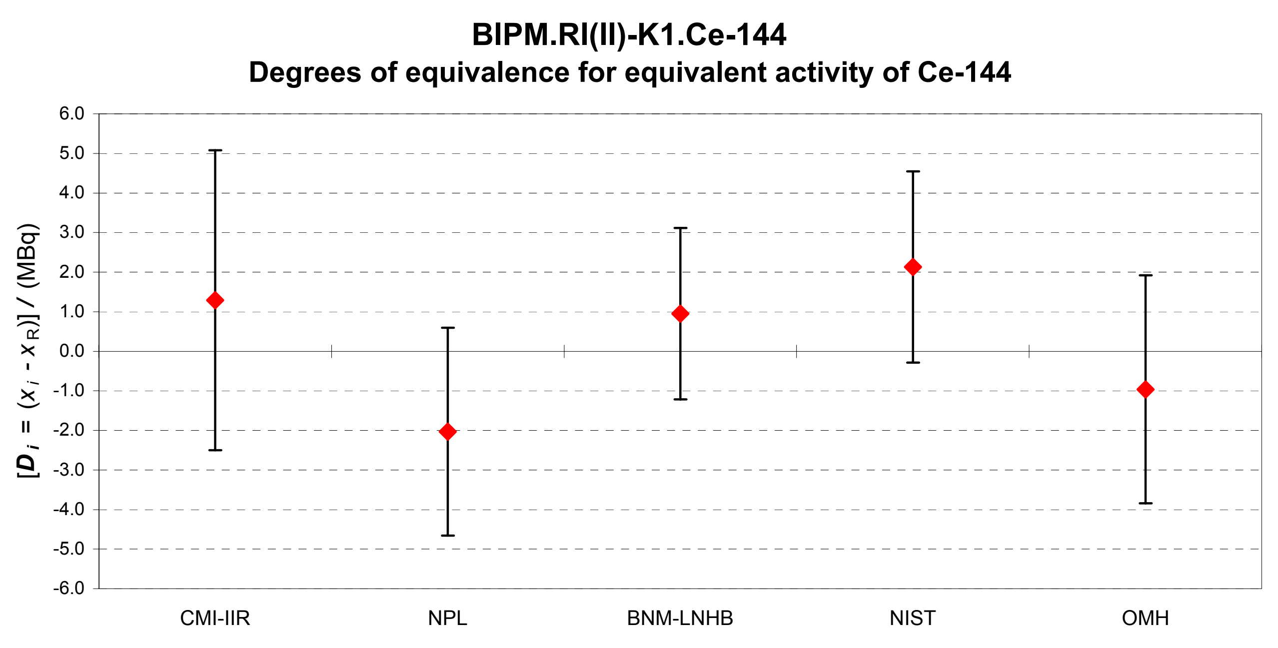
The degree of equivalence between two laboratories is given by a pair of terms:
$D_{ij} = D_i - D_j = (x_i - x_j)$ and $U_{ij}$, its expanded uncertainty ($k = 2$), both expressed in MBq.

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

Lab $j$

| Lab $i$ | $D_i$ | $U_i$ | CMI-IIR | | NPL | | BNM-LNHB | | NIST | | OMH | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $D_i$ /MBq | $U_i$ | $D_{ij}$ /MBq | $U_{ij}$ | $D_{ij}$ /MBq | $U_{ij}$ | $D_{ij}$ /MBq | $U_{ij}$ | $D_{ij}$ /MBq | $U_{ij}$ | $D_{ij}$ /MBq | $U_{ij}$ |
| CMI-IIR | **1.3** | 3.8 | | | **3.3** | 5.2 | **0.3** | 4.9 | **-0.8** | 5.1 | **2.3** | 5.4 |
| NPL | **-2.0** | 2.6 | **-3.3** | 5.2 | | | **-3.0** | 3.6 | **-4.2** | 3.8 | **-1.1** | 4.3 |
| BNM-LNHB | **0.9** | 2.2 | **-0.3** | 4.9 | **3.0** | 3.6 | | | **-1.2** | 3.4 | **1.9** | 3.9 |
| NIST | **2.1** | 2.4 | **0.8** | 5.1 | **4.2** | 3.8 | **1.2** | 3.4 | | | **3.1** | 4.1 |
| OMH | **-1.0** | 2.9 | **-2.3** | 5.4 | **1.1** | 4.3 | **-1.9** | 3.9 | **-3.1** | 4.1 | | |

**Figure 1.** **Graph of degrees of equivalence with the KCRV for $^{144}$Ce**
(as it appears in Appendix B of the MRA)



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

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

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

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

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

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

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

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

$x_i = A_{\mathrm{e}i}$ and $u_i = u_{A\mathrm{e}i}$ its uncertainty.

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

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

$$D_i = x_i - x_{\mathrm{ref}} \qquad (\mathrm{A-2})$$

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

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 $D_\mathrm{i}$ with respect to the $x_i$.

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

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

Then the total combined uncertainty becomes

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

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

or, after recombination

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

similar to (A-11).