International comparison of activity measurements of a solution of $^{238}$Pu

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An international comparison of activity measurements of a solution of $^{238}$Pu was organised by the BIPM under the auspices of the Comité Consultatif pour les Rayonnements Ionisants (CCRI(II)) in 2001. The importance of $^{238}$Pu in environmental studies was the main reason for choosing this radionuclide. However, this exercise is also being used to widen the scope of the comparisons organised by the BIPM and to extend the SIR (Système International de Référence pour la Mesure d’Activité d’Émetteurs $\gamma$) to $\alpha$-emitting radionuclides.

A reporting form (see Appendix 1) agreed by the Key Comparison Working Group (KCWG) members was issued by the BIPM and sent in December 2000 to the participating laboratories. Originally it was proposed to start the comparison in January 2001 with a deadline of 01 April 2001. For reasons principally related to customs problems and consequent difficulties for some laboratories to receive the ampoules, the deadline was postponed by a few months until 17 September 2001. Submissions up to 09 October 2001 were therefore accepted.

The solution of $^{238}$Pu was supplied, prepared and dispensed by the NPL. The solution was dispensed to NBS-type ampoules that were supplied by the BIPM. As no attempt was made to measure the ampoules in the SIR chambers, the ampoules were dispatched directly to the twelve participants by the NPL in the first semester of 2001. Each participant listed in Table 1 received a flame-sealed ampoule, containing about 3.0 g to 3.2 g of the solution. The $^{238}$Pu nominal activity concentration was 400 kBq/g of PuCl$_4$ in an aqueous solution of 1 M HCl. The solution contained high purity $^{238}$Pu; some traces of $^{241}$Pu (0.0121 % of the total activity), $^{240}$Pu (0.0004 %) and $^{239}$Pu (0.0027 %) were quoted by the NPL.

In order to harmonise the measurements, the same reference date 2001-04-01, 0h UTC was used for all reported measurements, the preliminary activity concentration measurements before opening of the ampoule, the impurity checks and the final activity concentration. All participants agreed to use the half-life value $T_{1/2} = (3203.0 \times 10^2 \text{ d}; \ u = 1.1 \times 10^2 \text{ d})$. The decay scheme of $^{238}$Pu from Lagoutine et al.\textsuperscript{1} was simplified following G. Triscone, and is shown in Fig. 1.

The twelve participating laboratories and the names of the persons who carried out the measurements are listed in Table 1.

Table 2 provides the list of the methods used by the laboratories. The method acronyms and codes that are used in Tables 3 and 4 and in Fig. 2 are also given. An overview of the codes is given in Appendix 2. For clarity the methods have been grouped into four categories characterized by their detection devices: liquid-scintillation counters, proportional counters pressurized or working at atmospheric pressure, defined solid angle counters and CsI(Tl) sandwich spectrometer.

Table 3 summarises the relative standard uncertainty components (1\(\sigma\)) as stated by the laboratories for the different methods applied. The uncertainties range from 0.12 % to 0.61 %, with most of the estimates around 0.30 %. There is no obvious link between the method used and the uncertainty stated.

The final results are presented in Table 4 and shown in Figure 2. Most of the results are enclosed in a band of about 0.5 % (+ 0.65 % and – 0.45 % respectively) at either side of the mean value of 360.40(0.29) kBq/g. The value assessed by the KRISS is slightly lower when compared with the other results but remains acceptable considering the quoted uncertainties. The first result sent by the OMH (355.6(2.2) kBq/g) was identified as being discrepant with the other values and the OMH asked for the possibility to carry out further measurements after the deadline. These additional measurements gave a value of 356.3(1.1) kBq/g in agreement with the first result obtained by this laboratory.

The degrees of equivalence are shown in Figure 3.

\textsuperscript{1}Nuclear and Atomic Decay Data, CD version : 1-98 – 19/12/98, BNM – CEA/DTA/DAMRI/LPRI
$T_{1/2} = 320,300 \text{ d}; \ u = 110 \text{ d}$

$\Sigma \alpha$-emission probabilities = 100 %

$Q^\alpha = 5593.22 \text{ keV}$

All energies are given in keV

$0^\alpha; 0$

Fig. 1 - Simplified decay scheme of $^{238}_{94}\text{Pu}_{144}$.
Table 1 - List of participants

| Organization       | Description                                                                 |
|--------------------|-----------------------------------------------------------------------------|
| BIPM               | Bureau International des Poids et Mesures, Sèvres, France (C. Colas, C. Michotte and G. Ratel) |
| BNM-LNHB           | Bureau National de Métrologie - Laboratoire National Henri Becquerel, Saclay, France (Ph. Cassette, F. Dayras, J. de Sanoit, C. Collin and B. Leprince) |
| CIEMAT             | Centro de Investigaciones Energéticas Medioambientales y Tecnológicas, Madrid, Spain (M. Teresa Crespo and E. Garcia-Toro) |
| CNEA               | Comision Nacional de Energia Atomica, Laboratorio de Metrologia de Radioisótopos, Buenos Aires, Argentina (J. Aguiar, P. Arenillas and M. Lobbe) |
| IRA                | Institut Universitaire de Radiophysique Appliquée, Lausanne, Suisse (G. Triscone and J.-Ch. Gostely) |
| IRMM               | Institute for Reference Materials and Measurements, Geel, Belgium (S. Pommé, T. Altitzoglou, L. Johansson, G. Sibbens and B. Denecke, measurements; T. Altitzoglou, G. Sibbens, S. Pommé, L. Johansson, source preparation; T. Altitzoglou, adsorption tests; T. Altitzoglou, G. Sibbens, impurity tests) |
| KRISS              | Korea Research Institute of Standards and Science, Taejon, Korea (Jong Man Lee, Pil Jae Oh and Tae Soon Park) |
| NPL                | National Physical Laboratory, Teddington, United Kingdom (A. Stroak, D. Woods, A. Woodman and A. Pearce) |
| OMH                | Országos Mérésügyi Hivatal, Budapest, Hungary (L. Szücsi, Gy. Hegyi and K. Rózsa) |
| PTB                | Physikalisch-Technische Bundesanstalt, Braunschweig, Germany (E. Günther, H. Janßen and R. Klein) |
| RC                 | Radioisotope Center “POLATOM”, Świerk, Poland (R. Broda and T. Terlikowska) |
| VNIIM              | D.I. Mendeleyev Institute for Metrology, St. Petersburg, Russia (T.E. Sazonova, M.A. Rasko and V. Zanevsky) |

Note: Since the comparison was carried out, the names and acronyms of some participating organizations have changed. These are: BNM-LNHB (now LNE-LNHB), IRMM (now JRC), OMH (now MKEH) and RC (now POLATOM).
| Method acronym | Description of the method | Laboratories using this method |
|----------------|---------------------------|--------------------------------|
| 4π(LS)α       | 4πα counting with a liquid-scintillation spectrometer | BIPM, BNM-LNHB, CIEMAT, CNEA, IRA, IRMM, RC, PTB |
| 4P-LS-AP-00-00 |                           | NPL                            |
| 4π(LS)α-γ tracer technique | 4πα-γ coincidence tracer technique method using the liquid-scintillation technique and $^{241}$Am as a tracer | RC                             |
| 4P-LS-AP-GR-CT-00 |                         |                                 |
| 4π(LS)α TDCR method | 4πα detection using the liquid-scintillation technique and the Triple-to-Double Coincidence Ratio method |                                 |
| 4P-LS-TD-AP-00-00 |                           |                                 |
| 4π(PC)α       | 4πα counting using a proportional counter at atmospheric pressure | BIPM, OMH, PTB |
| 4P-PC-AP-00-00 |                           | IRMM                           |
| 4π(PC)α-γ tracer technique | 4πα-γ coincidence tracer technique method using a proportional counter at atmospheric pressure for α-detection, a NaI(Tl) scintillator for γ-ray detection and $^{241}$Am as a tracer | NPL |
| 4P-PC-AP-GR-CT-00 |                         |                                 |
| 4π(PC)α-γ tracer technique | 4πα-γ coincidence tracer technique method using a pressurized proportional counter for α-detection, a NaI(Tl) scintillator for γ-ray detection and $^{241}$Am as a tracer | NPL |
| 4P-PP-AP-GR-CT-00 |                         |                                 |
| 4π(PC)α-Lx coincidence | 4πα-Lx coincidence counting using a proportional counter at a pressure slightly above the atmospheric pressure for α-detection and a NaI(Tl) scintillator for x-ray detection | VNIIM |
| 4P-PC-AP-XR-CO-00 |                         |                                 |
| α(DSA)         | α counting under defined solid angle | BNM-LNHB, VNIIM |
| SA-AP-00-00-00  |                           | CNEA, IRMM, PTB                 |
| α(DLSA)        | α counting under defined low solid angle | KRISS                          |
| SA-PS-AP-00-00  |                           | CIEMAT                         |
| α-x(DSA) coincidence | α-x coincidence counting under defined solid angle |                                 |
| SA-AP-XR-CT-00  |                           |                                 |
| 2π(GIC)α       | α counting using a grid ionization chamber with a 2π geometry |                                 |
| 2P-GC-AP-00-00  |                           |                                 |
| 4πCsI(Tl)α     | α counting with a 4π-CsI(Tl) sandwich-spectrometer | IRMM                           |
| 4P-CS-AP-00-00  |                           |                                 |
| Laboratory Method | BIPM $4\pi(\text{LS})\alpha$ | BIPM $4\pi(\text{PC})\alpha$ | BNM-LNHB $4\pi(\text{LS})\alpha$ | BNM-LNHB $\alpha(\text{DSA})$ | CIEMAT $2\pi(\text{GIC})\alpha$ | CIEMAT $4\pi(\text{LS})\alpha$ |
|-------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| **Components due to:** | | | | | | |
| counting statistics | ~ 0.07 0.12 3) | 0.12 3) | 0.04 0.03 | 0.11 0.04 | 0.08 3) | 0.01 |
| weighing | 0.28 # | # | < 0.001 0.001 | 0.01 | 0.01 | 0.01 |
| dead time | ~ 0.02 negligible | negligible | < 0.001 < 0.001 | 0.01 | 0.01 | 0.01 |
| background | | | | | | |
| pile-up | | | | | | |
| timing | | | | | | |
| adsorption | | | | | | |
| impurities | | | | | | |
| tracer | | | | | | |
| input parameters and statistical model | | | | | | |
| quenching | | | | | | |
| interpolation from calibration curve | | | | | | |
| decay-scheme parameters | < 0.0001 0.001 0.0002 0.0001 | 0.0001 | 0.3 | 0.0001 | 0.3 | 0.1 |
| self absorption | | | | | | |
| extrapolation of efficiency curve | | | | | | |
| spread of the measurements | 0.15 | 0.05 | 0.056 | 0.2 | 0.2 | 0.2 |
| wall effect | | | | | | |
| geometry factor | | | | | | |
| tail extrapolation and backscattering | | | | | | |
| differences between counters and series of measurements | | | | | | |
| uncertainty of method | | | | | | |
| combined uncertainty (as quadratic sum of all uncertainty components) | 0.33 | 0.32 | 0.12 | 0.12 | 0.39 | 0.24 |

* 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).

2) with efficiency extrapolation using the VYNS film addition technique
3) includes items marked by #
4) estimated from the bias observed when measuring $^{241}$Am sources of known activities with the same method including dilution
5) no visible effect on alpha counting
6) no visible effect on alpha counting
Table 3 - Uncertainty components of the activity concentration (in %) (continued)

| Laboratory Method | CNEA $4\pi(\text{LS})\alpha$ | IRA $4\pi(\text{LS})\alpha$ | IRMM $4\pi(\text{PPC})\alpha$ | IRMM $4\pi(\text{LS})\alpha$ |
|-------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| Components due to:|                               |                               |                               |                               |
| counting statistics | 0.25                          | 0.05                          | 0.1                           | 0.06                          |
| weighing           | 0.01                          | 0.06                          | 0.05                          | 0.08                          |
| dead time          | 0.01                          | 0.02                          | 0.1                           | 0.15                          |
| background         | < 0.01                        | 0.001                         | 0.01                          | 0.005                         |
| pile-up            |                               | 0.04                          |                               | 0.05                          |
| adsorption         | 0.08                          | 0.008                         | 0.008                         | 0.008                         |
| impurities         |                               | 0.007                         |                               |                               |
| tracer             | 0.48                          |                               |                               |                               |
| input parameters and statistical model | 1.0 | 0.1                          |                               |                               |
| quenching         |                               |                               |                               |                               |
| interpolation from calibration curve | 0.25 | 0.25                          |                               |                               |
| decay-scheme parameters | < 0.01 | 0.001                        | 0.001                         | 0.001                         |
| half life ($T_{1/2} = 320.3 \times 10^2$ d; $\mu = 1.1 \times 10^5$ d) | 0.9 |                               |                               |                               |
| self absorption    |                               | 0.3                           |                               |                               |
| degradation of the cocktail quality with time |                               |                               |                               |                               |
| foil absorption    |                               |                               | 0.2                           |                               |
| low energy tail extrapolation |                               |                               | 0.1                           |                               |
| variation amongst sources |                               |                               | 0.25                          |                               |
| combined uncertainty (as quadratic sum of all uncertainty components) | 0.58 | 0.4                           | 0.35                          | 0.29                          |

7) solid angle
8) standard deviation of the experimental data
9) $\Delta m = 22 \, \mu g$ and $m$ is the mean distributed mass of the four sources used
10) propagation of the background uncertainty on the result for the source having the lowest activity
11) $\Delta t$: $\Delta t = 0.01 \, \text{min}$ and $t$ is the mean measuring time of the four sources used
12) estimated from the content of the ten first channels divided by the total counting for the source of lowest activity
13) propagation of the uncertainty of the half life on the activity concentration
14) included in foil absorption
15) automatically corrected for by the instrument
16) 50 % of the adsorption
17) 50 % of the impurities
18) assumption of a 100 % efficiency
Table 3 - Uncertainty components of the activity concentration (in %) (continued)

| Laboratory Method | IRMM $\alpha$(DLSA) | KRISS $4\pi$CsI(Tl)$\alpha$ (coincidence) | NPL $4\pi$PC$\alpha-\gamma$ tracer technique | NPL $4\pi$LS$\alpha-\gamma$ tracer technique |
|-------------------|----------------------|----------------------------------------|------------------------------------------|------------------------------------------|
| counting statistics | 0.05 | 0.1 | 0.45 $^{21}$ | 0.04 | 0.092 |
| weighing | 0.05 | 0.05 | 0.03 | 0.03 | 0.017 |
| dead time | 0.01 | 0.02 | 0.03 $^{22}$ | 0.09 | $\alpha$ 0.116; $\gamma$ 0.002 |
| background | 0.05 (0.05) $^{19}$ | 0.1 $^{23}$ | $< 0.01$ | 0.041 |
| pile-up | 0.0 | 0.0 | $< 0.01$ | 0.1 |
| timing | 0.0005 | 0.0005 | 0.001 $^{24}$ | $< 0.01$ | 0.16 |
| adsorption | 0.008 | 0.008 | 1.0 | 0.52 |
| impurities | 0.007 | 0.007 | 0.05 | 0.050 |
| tracer | 0.0 | 0.0 | 0.16 | 0.175 |
| input parameters and statistical model | 0.0 | 0.0 | 0.0 | 0.0 |
| quenching | 0.0 | 0.0 | 0.0 | 0.0 |
| interpolation from calibration curve | 0.0 | 0.0 | 0.0 | 0.0 |
| decay-scheme parameters | 0.001 | 0.001 | 0.002 $^{25}$ | $< 0.01$ | 0.003; tracer 0.003 |
| half life ($T_{1/2} = 320.3 \times 10^2$ d; $u = 1.1 \times 10^3$ d ) | 0.23 $^{26}$ | 0.13 | 0.272 |
| self absorption extrapolation of efficiency curve | 0.09-0.02 | 0.1 | 0.016 |
| foil absorption | 0.25 | 0.25 | 0.25 |
| low energy tail extrapolation | 0.005 | 0.03 | 0.25 |
| variation amongst sources | 0.01 | 0.01 | 0.01 |
| detector efficiency | 0.01 | 0.01 | 0.01 |
| scattering at diaphragm & wall solid angle | 0.1 | 0.03 | 0.16 |
| evaporation during source preparation | 0.09-0.02 | 0.03 | 0.028 $^{27}$ |
| choice of fit to efficiency function | 0.1 | 0.1 | 0.1 |
| uncertainty in fit | 0.016 | 0.28 | 0.29 |
| combined uncertainty (as quadratic sum of all uncertainty components) | 0.30 | 0.36 | 0.52 | 0.28 |

$^{19}$ included in counting statistics
$^{20}$ included in foil absorption
$^{21}$ standard deviation of the means obtained for 8 sources
$^{22}$ calculated from measured uncertainty
$^{23}$ estimated from background data set
$^{24}$ estimated from time distribution
$^{25}$ calculated from the given data
$^{26}$ evaluated from residuals of the linear extrapolation
$^{27}$ effect of using Cox-Isham formula instead of Campion formula for dead-time correction
Table 3 - Uncertainty components of the activity concentration (in %) (continued)

| Laboratory Method | NPL $4\pi$(PPC)α-γ tracer technique | OMH $4\pi$(PC)α | PTB $4\pi$(LS)α | PTB α(DLSA) | PTB $4\pi$(PC)α |
|-------------------|------------------------------------|----------------|----------------|-------------|---------------|
| Components due to: |                                    |                |                |             |               |
| counting statistics | 0.17                              | 0.085          | 0.05           | 0.15        | 0.1           |
| weighing          | 0.016                              | 0.005          | 0.08           | 0.15        | 0.15          |
| dead time         | 0.046                              | < 0.001        | < 0.03         | < 0.01      | 0.01          |
| background        | 0.049                              | < 0.03         | < 0.03         | 0.03        | 0.03          |
| pile-up           | 0.086                              | 0.005          | 0.05           | < 0.01      | < 0.01        |
| timing            | 0.018                              | 0.02           | < 0.03         | < 0.03      | 0.20          |
| adsorption        |                                    |                |                |             |               |
| impurities        |                                    |                |                |             |               |
| tracer            |                                    |                |                |             |               |
| input parameters and statistical model |                    |                |                |             |               |
| quenching         |                                    |                |                |             |               |
| interpolation from calibration curve |                    |                |                |             |               |
| decay-scheme parameters |                    |                |                |             |               |
| half life ($T_{1/2} = 320.3 \times 10^2 \text{ d}; \mu = 1.1 \times 10^3 \text{ d}$) | < 0.001          | < 0.001        | < 0.001      | < 0.001     |
| self absorption   |                                    |                |                |             |               |
| extrapolation of efficiency curve |                    |                |                |             |               |
| scattering at diaphragm & wall tracer mixing ratio |                    |                |                |             |               |
| energy transfer from the α and the scintillator |                    |                |                |             |               |
| scattering of α in the spectrometer |                    |                |                |             |               |
| combined uncertainty (as quadratic sum of all uncertainty components) | 0.28               | 0.61           | 0.18           | 0.35        | 0.27          |

28) self absorption was estimated from a $^{241}$Am standardization carried out using $4\pi\alpha-\gamma$ coincidence counting under the same conditions as those used for the pure $\alpha$ measurements
29) negligible contribution
30) weights of about 40 mg
31) count rates approximately 1700 s$^{-1}$
32) sample spread 0.07 %
33) efficiency 100 %, no influence
34) no influence
35) for a time difference of 12 days between the date of reference and the date of measurements
36) for the set of sources
37) count rates lower than 20 s$^{-1}$
38) live-time correction of the multichannel analyser
39) geometry factors
40) time of measurement
41) not investigated
42) from efficiency measurements
43) negligible
Table 3 - Uncertainty components of the activity concentration (in %) (continued)

| Laboratory Method | RC | VNIIM |
|-------------------|----|-------|
|                   | 4π(LS)α | 4π(LS)α | 4π(PC)α-Lx coincidence | α(DSA) |
| TDCR method       | 0.2  | 0.2    | 0.02                    | 0.03   |
|                   | 0.19 | 0.19   | 0.05                    | 0.05   |
| Components due to:|     |        |                         |        |
| counting statistics | 0.03 | 0.02   | 0.15                    | 0.05   |
| weighing           | 0.03 | 0.001  |                         |        |
| dead time          | 0.01 |       |                         |        |
| background         | 0.1  | 0.02   |                         |        |
| pile-up            |      |        |                         |        |
| timing             |      |        |                         |        |
| adsorption         |      |        |                         |        |
| impurities         |      |        |                         |        |
| tracer             |      |        |                         |        |
| input parameters and statistical model | 0.1  |       |                         |        |
| quenching          |      |        |                         |        |
| interpolation from calibration curve |      |        |                         |        |
| decay-scheme parameters | 0.14 | 0.14  | 0.001                   | 0.001  |
| half life (T_{1/2} = 320.3 \times 10^2 \text{ d}; u = 1.1 \times 10^3 \text{ d}) | 0.001| 0.001 |                         |        |
| dilution           |      |        | 0.03                    | 0.03   |
| resolving time     |      |        | 0.003                   | 0.07   |
| G- factor          |      |        |                         | 0.1    |
| discrimination level |      |        |                         |        |
| combined uncertainty | (as quadratic sum of all uncertainty components) | 0.33  | 0.31    | 0.16   | 0.15   |
| Laboratory Method | Activity concentration (kBq g⁻¹) | Combined uncertainty (kBq g⁻¹) | Relative uncertainty / % |
|-------------------|----------------------------------|--------------------------------|--------------------------|
| **BIPM**          |                                  |                                |                          |
| 4π(LS)α           | 359.1*                           | 1.2                            | 0.33                     |
| 4π(PC)α           | 361.1                            | 1.2                            | 0.32                     |
| **BNM-LNHB**      |                                  |                                |                          |
| 4π(LS)α           | 361.77*                          | 0.40                           | 0.11                     |
| α(DSA)            | 362.05                           | 0.43                           | 0.12                     |
| **CIEMAT**        |                                  |                                |                          |
| 2π(GIC)α          | 359.4                            | 1.4                            | 0.39                     |
| 4π(LS)α           | 361.7*                           | 0.9                            | 0.25                     |
| **CNEA**          |                                  |                                |                          |
| 4π(LS)α           | 358.80*                          | 0.97                           | 0.27                     |
| α(DLSA)           | 359.4                            | 1.7                            | 0.48                     |
| **IRA**           |                                  |                                |                          |
| 4π(LS)α           | 360.9*                           | 1.5                            | 0.4                      |
| **IRMM**          |                                  |                                |                          |
| 4π(PC)α           | 360.2¹                           | 1.1                            | 0.3                      |
| 4π(LS)α           | 360.4                            | 1.1                            | 0.35                     |
| α(DLSA)           | 360.3                            | 1.1                            | 0.3                      |
| 4πCsI(Tl)α        | 359.6                            | 1.3                            | 0.35                     |
| **KRISS**         |                                  |                                |                          |
| α-x (DSA) coincidence | 358.1*                        | 1.9                            | 0.52                     |
| **NPL**           |                                  |                                |                          |
| 4π(PC)α-γ tracer technique | 362.7*                        | 1.0                            | 0.28                     |
| 4π(LS)α-γ tracer technique | 361.2                         | 1.7                            | 0.46                     |
| 4π(PC)α-γ tracer technique | 362.0                         | 1.1                            | 0.28                     |
| **OMH**           |                                  |                                |                          |
| 4π(PC)α           | 355.6*                           | 2.2                            | 0.61                     |
| **PTB**           |                                  |                                |                          |
| 4π(LS)α           | 360.8²                           | 0.7                            | 0.18                     |
| α(DLSA)           | 360.7                            | 1.3                            | 0.35                     |
| 4π(PC)α           | 360.8                            | 1.0                            | 0.27                     |
| **RC**            |                                  |                                |                          |
| 4π(LS)α TDCR method | 360.3                         | 1.2                            | 0.33                     |
| 4π(LS)α           | 360.3*                           | 1.1                            | 0.31                     |
| **VNIIM**         |                                  |                                |                          |
| 4π(PC)α-Lx coincidence | 361.5                        | 0.6                            | 0.16                     |
| α(DSA)            | 361.3*                           | 0.5                            | 0.15                     |
Notes

* Value used in estimating the KCRV and the degrees of equivalence

\(^1\) Value to be used was given by IRMM as the mean of the four results: 360.1(10) kBq/g

\(^2\) Value to be used was given by PTB as 360.8(10) kBq/g
Figure 2 – Final results of the $^{238}$Pu international comparison of activity concentration.

Notes

Further details of the methods used are given in Table 2. The result using the CIEMAT/NIST method by PTB has been reclassified as $4\pi(LS)\alpha$. 
Figure 3 - Degrees of equivalence for activity concentration for $^{238}$Pu

Notes:

1) The measurand is the activity concentration of $^{238}$Pu
2) The Key Comparison Reference Value is the power moderated weighted mean of the results ($x_R = 360.51 \text{ kBq.g}^{-1}$ with a standard uncertainty $u_R = 0.48 \text{ kBq.g}^{-1}$)
3) The value $x_i$ is the activity concentration for laboratory $i$.
4) The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_R)$ and $U_i$, its expanded uncertainty ($k = 2$), both expressed in kBq.g$^{-1}$, and $U_i = 2((1 - 2w_i)u_i^2 + u_R^2)^{1/2}$, where $w_i$ is the weight of laboratory $i$ contributing to the calculation of $x_R$.
5) The right-hand axis shows approximate values only.
6) Since the comparison was carried out, the names and acronyms of some participating organizations have changed. These are: BNM-LNHB (now LNE-LNHB), IRMM (now JRC), OMH (now MKEH) and RC (now POLATOM).
Appendix 1 – Agreed reporting form for the $^{238}$Pu international comparison

BUREAU INTERNATIONAL DES POIDS ET MESURES

Comparison of activity measurements of a solution of $^{238}$Pu
(January 2001)

Participating laboratory:

\[ T_{1/2} = (320.3 \times 10^2 \text{ d}; \quad u = 1.1 \times 10^2 \text{ d})^* \]

Ampoule number _ _ _ _ _ _

Mass of solution, according to distributing laboratory _ _ _ _ _ g

Name(s) of the person(s) who carried out the measurements:

Date:

Please send the filled-in form and any additional information to the BIPM
not later than April 1st, 2001.

* Ulrich Schötzig und Heinrich Schrader, Halbwertszeiten und Photonen-Emissionswahrscheinlichkeiten von häufig verwendeten Radionukliden, PTB-Bericht PTB-Ra-16/5, Braunschweig, September 1998.
A. Preliminary measurements

A.1. Method used for preliminary measurements

- calibrated ionization chamber [ ] YES [ ] NO
- well crystal [ ] YES [ ] NO
- other method [ ] YES [ ] NO

A.2. Results obtained

Radioactivity concentration, in kBq g\(^{-1}\) (2001-04-01, 00 h UTC)

- before opening the original ampoule
  date of this measurement

- after transfer to another ampoule
  date of this measurement

Total mass of solution found in the ampoule _ _ _ _ _ g.

A.3. Adsorption tests

Please take into account the adsorption tests in the evaluation of the final results.

A.3.1. Adsorption tests carried out with liquid-scintillation counting

Please keep in mind the following:
- No rinsings are necessary,
- Use a water immiscible cocktail to measure the residual activity.

Activity remaining in the "empty" original ampoule _ _ _ _ _ Bq.

Date of this test _ _ _ _ _ _ _ _ _ _ _ _ _ _ .

Please explain the measuring procedure used:
A.3.2. Adsorption tests carried out with proportional counting

Please rinse the ampoule with an aggressive solution to remove most of the activity and prepare solid source(s) to measure this residual activity.

Activity remaining in the "empty" original ampoule _ _ _ _ _ _ Bq.
Date of this test _ _ _ _ _ _ _ _ _ _ _ _.

A.4. Impurity checks*

Method of measurement _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _
B. Source preparation

B.1. Methods used for source preparation:
Possible remarks about drying, precipitation, foils used (gold-coated or not, number, etc.), type of balance used…
B.2. Solutions, sources

B.2.1. For beta counting and photon counting (if relevant)

Diluent:

| dilution number |
|-----------------|
| 1               | 2 3 | ... |
| - dilution factor |     |     |
| possible remarks     |     |     |
| - number of sources prepared |     |     |
| - dispensed mass of solution (approx.) |     |     |

B.2.2. For liquid-scintillation counting

Diluent
Dilution factor
Scintillator used to prepare the sources
Volume of scintillator used cm³
Chemicals used to stabilize the solution
Substances used as quenching agent
Type of vials used
C. Procedures used for the activity measurements

**Method of measurement used**
(e.g. $2\pi$(PC)$\alpha$ counting, $4\pi$(PC)$\alpha$ counting, absolute counting with defined solid angle, $\alpha$-x coincidence counting with defined solid angle, liquid-scintillation counting or other)

Please list the values for all the decay-scheme parameters (branching ratios, transition intensities, internal conversion coefficients, etc.) relevant to your measurements.
In case you used more than one method, please assemble the relevant information on separate sheets.

D. Detectors, counting equipment

D.1. Alpha counting (channel 1)

D.1.1. Proportional counter

| Solid angle | _ _ _ _ _ sr |
| Wall material | _ _ _ _ _ | Height of each half | _ _ _ _ _ mm |
| Anode | | |
| Nature | _ _ _ _ _ |
| Wire diameter | _ _ _ _ _ mm | Wire length | _ _ _ _ _ mm |
| Distance from source | _ _ _ _ _ mm |
| Voltage applied | _ _ _ _ _ kV |
| Gas | | |
| Nature | _ _ _ _ _ |
| Pressure | (above atmospheric pressure) | _ _ _ _ _ MPa |

Remarks
D.1.2. Liquid-scintillation equipment

D.1.2.1. CIEMAT/NIST method

D.1.2.1.1. Characterization of the liquid-scintillation counter (LSC)

Type of the counter
Age
Quench parameter
Nuclide used for the determination of the quench parameter
Efficiency obtained with an unquenched standard of $^3$H
Background (unquenched standard in toluene scintillator, 0 keV to 2000 keV or more)
Options used (e.g. low-level counting)

D.1.2.1.2. Characterization of the tracer (e.g. $^3$H)

Standard used and its origin
Uncertainty on the standard
Date of preparation of the tracer samples
Chemical composition of the tracer samples

D.1.2.2. TDCR method

D.1.2.2.1. Characteristics of the experimental equipment

Type of phototubes
Operating temperature
Discrimination level
Coincidence resolving time

Type of dead time extending □ non-extending □
Minimum dead-time length _ _ _ _ μs
Efficiency variation method:
- defocusing
- grey filters
- chemical quenching
- other ones (please describe)

External standard ($^3$H or other) used for the determination of the figure of merit
D.1.2.3. *Other method(s)*
### D.2. Photon counting (channel 2)

#### D.2.1. Scintillator detector

| Parameter                              | Value           | Parameter                              | Value           |
|----------------------------------------|-----------------|----------------------------------------|-----------------|
| Crystal material                       |                 | Solid angle                            | _ _ _ _ _ _ _ _ sr |
| Number of crystals                     |                 | Well type                               | YES □           |
| Crystal diameter                       | _ _ _ _ _ _ _ _ mm | Crystal height                          | _ _ _ _ _ _ _ _ mm |
| Well diameter                          | _ _ _ _ _ _ _ _ mm | Well depth                              | _ _ _ _ _ _ _ _ mm |
| Window material                        |                 | Window thickness                        | _ _ _ _ _ _ _ _ mm |
| Distance between photon counter and source |                 |                                         | _ _ _ _ _ _ _ _ mm |
| Resolution at                          | _ _ _ _ _ _ _ _ keV, | FWHM*                                   | _ _ _ _ _ _ _ _ %, _ _ _ _ _ keV |

Please add a typical pulse-height spectrum.

#### D.2.2. Semiconductor detector

| Parameter                              | Value           | Parameter                              | Value           |
|----------------------------------------|-----------------|----------------------------------------|-----------------|
| Nature                                 |                 | Solid angle                            | _ _ _ _ _ _ _ _ sr |
| Type                                   |                 | Coaxial                                | □ Planar □      |
| Number of detectors                    |                 | Well type                               | YES □           |
| Diameter                               | _ _ _ _ _ _ _ _ mm | Volume                                 | _ _ _ _ _ _ _ _ cm³ |
| Window material                        |                 | Window thickness                        | _ _ _ _ _ _ _ _ mm |
| Distance between photon counter and source |                 |                                         | _ _ _ _ _ _ _ _ mm |
| Resolution at                          | _ _ _ _ _ _ _ _ keV, | FWHM*                                   | _ _ _ _ _ _ _ _ %, _ _ _ _ _ keV |

Please add a typical pulse-height spectrum and an efficiency curve.

---

* full width at half maximum
Radionuclides used for an efficiency determination (if relevant)

| $E_\gamma$ (keV) | $P_\gamma$ (%) |
|------------------|--------------|
| -----------------|--------------|
| -----------------|--------------|
| -----------------|--------------|
| -----------------|--------------|

D.2.3. *Other detectors used (e.g. ionization chamber)*
D.3. Parameters of counting equipment

(Give a brief description and/or a block diagram of the experimental arrangement.)

D.3.1. Channel 1 (alphas)

| a) Discrimination level | _ _ _ _ keV (or window) |
|-------------------------|-------------------------|
| b) Dead times and their uncertainties (standard deviation) |
| Dead time $\tau_1 = _ _ _ _ \mu s; u = _ _ _ _ \mu s$ |
| Type of dead time | extending ☐ non-extending ☐ |
| Method used for measurement | ____________________________ |
| Live time clock | Yes ☐ No ☐ |
| Pulser technique | Yes ☐ No ☐ |
| Loss free counting | Yes ☐ No ☐ |
| c) Pile-up rejector | Yes ☐ No ☐ |

D.3.2. Channel 2 (photons) (if relevant)

| a) Discrimination level | _ _ _ _ keV (or window) |
|-------------------------|-------------------------|
| b) Dead times and their uncertainties (standard deviation) |
| Dead time $\tau_2 = _ _ _ _ \mu s; u = _ _ _ _ \mu s$ |
| Type of dead time | extending ☐ non-extending ☐ |
| Method used for measurement | ____________________________ |
D.3.3. *Coincidence unit (if relevant)*

Coincidence resolving time \( \tau_R = _-_ \text{ } \mu\text{s} \); \( \sigma = _-_ \text{ } \mu\text{s} \)

Method used for measurement


D.3.4. *Other modules used*

(for LSC see section D.1.2.)
E. Relevant data, corrections and uncertainties

E.1. Date of measurement
(Mean date on which your measurements were performed)

E.2. Measuring data

E.2.1. Channel 1 (alphas)
Dead time \(_ \_ \_ \_ \_ \_ \mu s \) Number of sources measured \(_ \_ \_ \_ \) Background rate \(_ \_ \_ \_ \_ s^{-1} \) Typical count rate \(_ \_ \_ \_ \_ s^{-1} \)
Typical time for one measurement \(_ \_ \_ \_ \_ s \) Discrimination threshold or window \(_ \_ \_ \_ \_ keV \)

E.2.2. Channel 2 (photons) (if relevant)
Dead time \(_ \_ \_ \_ \_ \_ \mu s \) Number of sources measured \(_ \_ \_ \_ \) Background rate \(_ \_ \_ \_ \_ s^{-1} \) Typical count rate \(_ \_ \_ \_ \_ s^{-1} \)
Typical time for one measurement \(_ \_ \_ \_ \_ s \) Discrimination threshold or window \(_ \_ \_ \_ \_ keV \)

E.2.3. Extrapolation of efficiency function (coincidence method)
Maximum achieved efficiency \(_ \_ \_ \_ \_ \_ \% \)
Method used for varying the efficiency

Number of degrees of freedom \(_ \_ \_ \_ \_ \)
Please add a figure, if possible.
E.2.4. *Liquid-scintillation parameters*

Numerical codes used

kB value  

Formula used to calculate the ionization quenching correction factor $Q(E)$

Are M, N, ... captures taken into account?

Are M, N, ... x-ray and Auger electrons taken into account?

Model used to evaluate the interaction probability of the photons with the scintillator

Values used for cross section of interaction

E.2.5. *Calculated data for the liquid-scintillation method*

Total efficiency for $^{238}\text{Pu}$

E.2.6. *Corrections applied*
E.2.7. *Uncertainty components*, in % of the activity concentration, due to

| Remark                        | Remarks                                      |
|-------------------------------|----------------------------------------------|
| counting statistics           | ---                                          |
| weighing                      | ---                                          |
| dead time                     | ---                                          |
| background                    | ---                                          |
| pile-up                       | ---                                          |
| timing                        | ---                                          |
| adsorption                    | ---                                          |
| impurities                    | ---                                          |
| tracer                        | ---                                          |
| input parameters and statistical model | ---                                      |
| quenching                     | ---                                          |
| interpolation from calibration curve | ---                                      |
| decay-scheme parameters       | ---                                          |
| half life (= 320.3 × 10^2 d;  
  \(u = 1.1 \times 10^2 d\)) | ---                                          |
| self absorption               | ---                                          |
| extrapolation of efficiency curve (coincidence method) | ---                                      |
| other effects (if relevant) (explain) | ---                                      |
| combined uncertainty (as quadratic sum of all uncertainty components) | ---                                      |

* 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).
F. Combination of individual results
(obtained from the individual dilutions, source preparation, etc.)

How have the individual results been used for arriving at the final result (statistical weights, coverage factor, etc.)?

G. Final result

The radioactivity concentration of the $^{238}$Pu solution on the reference date$^*$ (2001-04-01, 00 h UTC) is

\[ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \text{kBq g}^{-1}, \]

and the combined uncertainty is

\[ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \_ \text{kBq g}^{-1}, \_ \_ \_ \_ \_ \_ \% . \]

Remarks

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* To adjust your result to the reference date, please use the half-life value given on page 1.
Appendix 2 - KCWG proposal for acronyms used to identify different measurement methods

| Geometry                | acronym | Detector                    | acronym |
|-------------------------|---------|-----------------------------|---------|
| $4\pi$                  | 4P      | proportional counter        | PC      |
| defined solid angle     | SA      | pressurized proportional counter | PP      |
| $2\pi$                  | 2P      | liquid scintillation counting | LS      |
|                         |         | Nal(Tl)                     | NA      |
|                         |         | Ge(HP)                      | GH      |
|                         |         | Ge-Li                       | GL      |
|                         |         | Si-Li                       | SL      |
|                         |         | CsI                          | CS      |
|                         |         | ionisation chamber           | IC      |
|                         |         | bolometer                    | BO      |
|                         |         | calorimeter                  | CA      |
|                         |         | PIPS detector                | PS      |
|                         |         | Grid ionisation chamber      | GC      |

| Radiation               | acronym | Mode                           | acronym |
|-------------------------|---------|--------------------------------|---------|
| positron                | PO      | efficiency tracing             | ET      |
| beta particle           | BP      | internal gas counting          | IG      |
| Auger electron          | AE      | CIEMAT/NIST                    | CN      |
| conversion electron     | CE      | sum counting                   | SC      |
| bremsstrahlung          | BS      | coincidence                    | CO      |
| gamma ray               | GR      | anti-coincidence               | AC      |
| x - rays                | XR      | coincidence counting with      | CT      |
|                         |         | efficiency tracing             |         |
| alpha - particle        | AP      | anti-coincidence counting with | AT      |
|                         |         | efficiency tracing             |         |
| mixture of various      | MX      | triple-to-double coincidence   | TD      |
| radiation e.g. x         |         | ratio counting                 |         |
| and gamma               |         | selective sampling             | SA      |

**Examples**

| method                                         | acronym            |
|------------------------------------------------|--------------------|
| $4\pi$(PC)$\beta-\gamma$-coincidence counting | 4P-PC-BP-NA-GR-CO  |
| $4\pi$(PPC)$\beta-\gamma$-coincidence counting efficiency, tracing | 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  |