APMP supplementary international comparison of activity measurement of Cs-134 and Cs-137 in brown rice

(APMP.RI(II)-S3.Cs-134.Cs-137)

Akira Yunoki[1)], Tsutomu Miura[1)], Yasushi Sato[1)], Pi-Fen Lee[2)], Mikael Hult[3)], Faidra Tzika[3)], Guillaume Lutter[3)], Gerd Marissens[3)], Jong-Man Lee[4)], K. B. Lee[4)], Svetlana Nour[5)], Jerome La Rosa[5)], Jun Saegusa[6)], Thongchai Soodprasert[7)], Hermawan Candra[8)], Holnisar[8)], Pujadi Marsoem[8)], Gatot Wurdiyanto[8)], Shioka Hamamatsu[9)] and Mayumi Hachinohe[9)]

[1)] National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan
[2)] Institute of Nuclear Energy Research, No.1000, Wenhua Rd., Jiaan Village, Longtan Township, Taoyuan County, 32546, Taiwan (R.O.C.)
[3)] European Commission, JRC-GEEL, Retieseweg 111, 2440 Geel, Belgium
[4)] Korea Research Institue of Standards and Science, 267 Gajeong-Ro, Yuseong-Gu, Daejeon 34113, Republic of Korea
[5)] National Institute of Standards and Technology, 100 Bureau Drive, MS 8462, Gaitherburg, MD 20899-8462, USA
[6)] National Physical Laboratory, Hampton RD, Teddington, Middleesex, UK, TW11 0LW
[7)] Ionizing Radiation Metrology Laboratory, Bureau of Technical Support for Safety Regulation, Office of Atoms for Peace, 16 Vibhavadi Rangsit Rd., Chatuchak, Bangkok 10900, Thailand
[8)] Pusat Teknologi Keselamatan Dan Metrologi Radiasi, Badan Tenaga Nuklir Nasional, Jl. Lebak Bulus Raya, Kotak Pos 7043 JKSKL, Jakarta Selatan, 12070, Indonesia
[9)] National Agriculture and Food Research Organization, 3-1-1 Kannondai, Tsukuba, Ibaraki 305-8517, Japan

Abstract

The supplementary international comparison of activity measurement of Cs-134 and Cs-137 in brown rice (APMP.RI(II)-S3.Cs-134.Cs-137) was carried out within the framework of the Asia-Pacific Metrology Programme (APMP). The matrix of the sample was brown rice grain harvested in autumn 2011, after the accident of the Fukushima Daiichi Nuclear Power Plant. No spiking was applied to the sample. Eight institutes reported their measurement results. Averages of the reported results and their associated uncertainties were obtained using the power-moderated mean. This report describes the supplemental comparison reference value, the degree of equivalence of the results, the procedure of the comparison and the sample preparation and measurements by the participants.

1. Introduction

After the accident of Fukushima Daiichi Nuclear Power Plant, many measurement institutes began to measure the activities of various foods. Consequently, the importance of reliable measurement of radionuclides in food increased significantly. National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology (NMIJ/AIST) and Institute of

Food Research, the National Agriculture and Food Research Organization (NARO) developed a certified reference material of brown rice grain containing Cs-134 and Cs-137 (NMIJ CRM 7541-a/b) [1-3] to provide a tool to validate their measuring methods. The NMIJ/AIST and NARO conducted several domestic comparisons of activity measurement using the brown rice samples [4]. To confirm the equivalence of NMIJ/AIST's measurements with internationally obtained values, NMIJ/AIST proposed a regional international comparison to the Asia-Pacific Metrology Programme Technical Committee for Ionizing Radiation (APMP/TCRI) during its workshop held on 24 December 2012. The proposal was approved and the NMIJ/AIST was assigned as a pilot laboratory. The comparison was registered to the BIPM key comparison database (KCDB) as APMP.RI(II)-S3.Cs-134.Cs-137. NARO prepared the sample using rice harvested in 2011 after the accident of the Fukushima Daiichi Nuclear Power Plant. The sample contained Cs-134 and Cs-137 released from the Fukushima Daiichi Nuclear Power Plant at the accident, and their activities were approximately the same. In 2013, each participant received two packages of the sample whose amount was 81 g each. Eight institutes received the sample packages and reported their measurement results. This report provides the supplemental comparison reference values (SCRVs) and the degrees of equivalence of the results. The procedure for this comparison, the sample preparation and measurements by the participants are also described.

2. Technical protocol

2.1 Measurement

Each participant received two vessels of the brown rice grain sample whose net mass was listed in Table 2. The participants were asked to report activities (in Bq) of Cs-134 and Cs-137 in the sample. They actually reported an average activity of two samples, an activity of each sample separately or an activity of either sample. Some of them reported an activity per unit mass. Uncertainty estimation was performed according to the Guide to the expression of uncertainty in measurement (GUM) [5]. The reference date of the measurement was 1st August 2013, 0:00:00 UTC. The participants also received two empty vessels identical to those that contained the sample for the convenience of efficiency calibration.

2.2 Time Schedule

The samples were shipped to the participants in July 2013. In some cases, it took several months for samples to reach the participants. The participants reported their results to the pilot laboratory (NMIJ/AIST) by January 2014. The NMIJ/AIST prepared draft A of this report in June 2017. After circulating it among participants of the comparison, the NMIJ/AIST revised and submitted it to the BIPM via APMP/TCRI in 2022. A work-flow of the comparison is in accordance with Appendix B of CIPM MRA-G-11 "Measurement comparisons in the CIPM MRA - Guidelines for organizing, participating and reporting".

2.3 Evaluation of the comparison results

The result of each participant was evaluated by the degree of the equivalence (DoE) with the supplementary comparison reference value (SCRV). The SCRV was obtained by the power moderated mean [6]. In addition, the result of each participant was also evaluated by using the E_n score (see Appendix 3).

3. Participants

Table 1 lists the acronyms, their full names, countries and Regional Metrological Organizations (RMOs) of the participants.

Table 1. Acronyms, full names, countries, and RMOs of participants

Acronym	Full name of participant	Country	RMO
NRSL/INER	Institute of Nuclear Energy Research, Atomic Energy Council, Executive Yuan	Chinese Taipei	APMP
JRC-Geel	European Commission, Joint Research Centre in Geel	European Union	EURAMET
KRISS	Korea Research Institute of Standards and Science	Korea	APMP
NIST	National Institute of Standards and Technology	United States of America	SIM
NPL	National Physical Laboratory	United Kingdom	EURAMET
OAP	Office of Atoms for Peace	Thailand	APMP
PTKMR-BATAN	Pusat Teknologi Keselamatan dan Metrologi Radiasi, Badan Tenaga Nuklir Nasional	Indonesia	APMP
NMIJ/AIST	National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology	Japan	APMP

Unfortunately, the China National Institute of Metrology did not receive the sample because Chinese customs did not permit importation of the rice sample. The India Bhabha Atomic Research Centre did not report its measurement result.

4. Sample preparation

NARO prepared the raw sample material using brown rice grain originated from rice (*Oryza sativa* L.) harvested a half year after the accident at the Fukushima Daiichi Nuclear Power Plant. The brown rice grain was stored in six paper bags weighing 30 kg each at 15 °C to prevent insect damage and spoilage. To prevent removal of Cs-134 and Cs-137 from the surface of the brown rice, the grains were homogenized manually using a stainless-steel scoop with a capacity of 1800 cm^3. The homogenization procedure was as follows:

1) Brown rice grains from the six paper bags were placed on a thoroughly cleaned polyethylene sheet in six blocks placed in a circle at equal intervals.
2) One scoop of the brown rice grain from each block was gathered to the center of the circle and mixed each time by stirring vertically with scoop. This process was repeated until all six blocks of the brown rice were mixed.
3) The gathered mixed brown rice grains were spread out in a cone shape and radially divided into eight blocks of the same amount. Diagonally opposed blocks were combined and mixed by

stirring vertically with the scoop. As a result, four blocks of thoroughly homogenized brown rice grains were obtained [7].

Then, the homogenized brown rice grains were packaged in polypropylene vessels with a volume of approximately 100 cm^3 [8]. They were not spiked with Cs-134 or Cs-137. Their amounts (net wet mass of the rice grain) were controlled to 81.00 ± 0.02 g per package at a bulk density of approximately 0.94 g cm^{-3}. Packages were sterilized by gamma irradiation of 17.6 kGy - 21.8 kGy to facilitate their transport across national borders. Each participant received two vessels containing the rice grain sample of 81.00 g to 81.01 g in each vessel (see Table 2). Their net wet masses were measured when they were packaged in the pilot laboratory. Special attention was paid to packages having the same mass. An acrylic plate and a polystyrene foam plate were inserted between the rice grains and cap of each vessel to avoid rice grains from moving and rubbing against each other.

Table 2. Identification numbers and net wet masses of samples

Sample No.	Sample net wet mass (g)	Participant
3	81.00	NRSL/INER
17	81.00	
48	81.00	JRC-Geel
79	81.01	
7	81.00	KRISS
22	81.00	
30	81.00	NIST
68	81.01	
32	81.00	NPL
70	81.01	
8	81.01	OAP
23	81.00	
14	81.00	PTKMR-BATAN
41	81.00	

The NMIJ/AIST measured activities of 27 randomly selected 81 g samples using gamma spectrometry with a high-purity germanium detector. The averages of the measurement results were 3.35 Bq for Cs-134 and 6.99 Bq for Cs-137 on the reference date of 1 August 2013, 0:00 UTC.

Relative standard uncertainty due to bottle-to-bottle homogeneities (s_{bb}) was estimated using the following formula:

$$s_{bb}^2 = s_{b+r}^2 - s_r^2,$$

where s_{b+r} is the relative standard deviation of the measurement results and s_r is repeatability, which is considered to be a measure of uncertainty due to counting statistics. The s_{b+r}, s_r and s_{bb} values are listed in Table 3.

Table 3. Relative uncertainty components due to bottle-to-bottle homogeneity (10^{-2})

Nuclide	s_{b+r}	s_r	s_{bb}
Cs-134	3.11	1.68	2.62
Cs-137	1.44	0.94	1.10

5. Sample treatment by the participants

NRSL/INER, KRISS, NPL, OAP, PTKMR-BATAN and the NMIJ/AIST measured the activity of the samples as they were received. JRC-Geel measured the activity of the one sample (No. 48) as received, while the other sample (No. 79) was transferred to one of JRC-Geel's standard sample containers. NIST measured the activity of the samples by both non-destructive and destructive methods. The non-destructive method was direct measurement of one 81 g brown rice sample as received. The destructive method measured the activity of the sample treated as follows: combustion of both 81 g brown rice samples at 450 °C, dissolution of the remaining ash and preparation of 1 M HNO_3 solutions (100 cm^3 each) of dissolved ash.

6. Method of efficiency calibration

NRSL/INER calibrated peak count efficiencies (counts per photon, s^{-1}/s^{-1}) for 569 keV, 605 keV and 796 keV photons from Cs-134 and 662 keV photons from Cs-137. NRSL/INER used a standard solution containing Eu-152, Cs-134 and Cs-137, and a rice sample spiked with Cs-134 and Cs-137.

JRC-Geel set up an experimental efficiency curve (counts per photon, s^{-1}/s^{-1}) using a multinuclide reference solution from NPL that was poured into one of the empty containers (identical to the container in which the rice was shipped) that was sent to the participants by the pilot laboratory. Efficiency transfer was calculated using the Monte Carlo code EGSnrc with a model of the detector that was set up and validated using point sources from PTB. Peak count efficiencies for Cs-134 were determined using the Monte Carlo code, which included the complete decay scheme of Cs-134, and properly accounted for the true summing coincidences. A peak count efficiency for Cs-137 was determined using the Monte Carlo code, which included only tracing the 662 keV gamma rays.

KRISS calibrated the peak counting efficiencies (counts per photon, s^{-1}/s^{-1}) for 605 keV photons from Cs-134 and 662 keV photons from Cs-137 using a standard solution source with Monte Carlo calculations.

NIST calibrated peak counting efficiencies (counts per decay, s^{-1}/Bq^{-1}) for 605 keV and 796 keV photons from Cs-134 and 662 keV photons from Cs-137. The peak counting efficiency was obtained by dividing the net photopeak rate (s^{-1}) by the activity of the radionuclide (Cs-134 or Cs-137, Bq). NIST prepared two calibration sources of different dimensions; one was for the non-destructive method and the other was for the destructive method. The source for the non-destructive method was

prepared by spiking brown rice with a calibrated solution containing Cs-134 and Cs-137. The source for the destructive method was prepared by spiking 100 cm^3 of 1 M HNO_3 solutions of blank rice ash with a calibrated solution containing Cs-134 and Cs-137. The same spiked materials (rice and solutions), containing both Cs-134 and Cs-137, were used for counting efficiency calibrations of the 605, 796 and 662 keV gamma ray photopeaks.

NPL obtained the activity of each sample by comparing the net counts of the sample and a reference source of known activity while considering self-attenuation correction using MCNP5 and decay correction. The photopeaks of 605 and 796 keV photons were used for Cs-134 and that of 662 keV photons was used for Cs-137. NPL prepared two reference sources of Cs-134 and two reference sources of Cs-137, whose dimensions were the same as those of the distributed samples, and the activity was approximately 0.75 kBq per package for Cs-134 and 0.5 kBq per package for Cs-137, respectively.

OAP calibrated the peak count efficiencies (counts per decay, s^{-1}/Bq^{-1}) for photons from Cs-134 and Cs-137. The photopeak of 796 keV photons was used for Cs-134 and that of 662 keV photons was used for Cs-137. OAP prepared a point source using brown Thai rice and placed it at various positions to simulate the sample.

PTKMR-BATAN calibrated the peak count efficiencies (counts per photon, s^{-1}/s^{-1}) for 796 keV photons from Cs-134 and 662 keV photons from Cs-137. PTKMR-BATAN prepared the calibration source of rice spiked with Cs-134 and Cs-137 using the same containers and filling them with the same amount of rice as that of the samples.

NRSL/INER, JRC-Geel, KRISS, OAP and PTKMR-BATAN used DDEP or BIPM/Monograph-5 as references of the nuclear data of the photon emission rate. NRSL/INER initially reported results using LARA [9] and NuDat [10]; however, it revised its value using DDEP according to a requirement from the pilot laboratory. The result for Cs-134 did not change, while that for Cs-137 increased 0.2%, which is negligibly smaller than the associated uncertainty in this comparison.

The NMIJ/AIST calibrated counting efficiencies (counts per decay, s^{-1}/Bq^{-1}) of 796 keV and 804 keV photons for Cs-134 and that of 662 keV photons for Cs-137. The counting efficiencies were calibrated using standard solutions of Cs-134 and Cs-137. The solutions were contained in the same type of vessel as that used for the sample. A difference of efficiency due to differences in sample height and self-attenuation were adjusted using EGS5.

7. Detector details

All participants used a high-purity germanium detector. NRSL/INER used an ORTEC GEM 30 coaxial detector with dimensions of 58.8 mm in diameter and 69.6 mm in length. The gap from the end cap to the detector was 3 mm. Signals from the detector were processed by an ORTEC digital signal processor (DSP).

JRC-Geel used an ultra-low-level gamma-ray spectrometer with a high-purity germanium detector (HPGe) in the HADES laboratory located 225 m underground. JRC-Geel used three types of detectors; (1) Canberra Ge-detector (P-type, coaxial with 0.7 mm dead layer) whose diameter and height were 59 mm and 31 mm, respectively. Aluminium endcap was used. Relative efficiency was 20%, (2) Canberra Ge-detector (P-type, coaxial with 0.7 mm dead layer) whose diameter and height were 62 mm and 66 mm, respectively. Aluminium endcap was used. Relative efficiency was 46%, (3) Canberra Ge-detector (P-type, planar with 0.0003 mm dead layer) whose diameter and height were 61 mm and 25.8 mm, respectively. Aluminium endcap was used. Relative efficiency was 19%.

Signals from the detector were processed by a Canberra DSA-2000 type DSP with Genie-2000 software.

KRISS used p-type detectors whose relative efficiencies were 70% and 120%.

NIST used a Canberra model GR7023, a reverse electrode and closed-end coaxial type detector whose Ge crystal dimensions were 74.5 mm in diameter and 66.5 mm in length. It had 70% relative efficiency and a vertical dipstick.

NPL used an ORTEC GEM-FX8530-S cooled by LN_2; its dimensions were 85.1 mm in diameter and 33.2 mm in length. It was mounted onto a copper crystal cup and installed in a lead shield. The sample was set on a sample holder that covered a light-shield acrylic detector cap with a thickness of 0.76 mm.

OAP used an EG&G Model GWL-120230 well type detector whose volume was 120 cm^3. The signal from the detector was processed by DSPEC.

PTKMR-BATAN used a Canberra model GC 1018 N-type coaxial detector whose crystal was 47.5 mm in diameter and 28.5 mm thick. It was operated with a depletion voltage of +4500 V. It had a relative efficiency of 10.3% with an energy resolution of 1.6 keV full width at half maximum at 1332.5 keV. The signal from the detector was fed to a model 2002CSL pre-amplifier and a Canberra model 2020 amplifier. GENIE 2000 software was used for its spectrum analysis.

The NMIJ/AIST measured activities of samples using ORTEC GEM-130225 HPGe detectors whose dimension were 90 mm in diameter and 100 mm in length.

8. Measurement methods

All participants measured the activity of a sample using gamma-ray spectrometry with high-purity germanium detector(s). Its code is UA-GH-GR-00-00-00 (see Appendix 2).

NRSL/INER, KRISS, NPL, PTKMR-BATAN and the NMIJ/AIST set the sample in a distributed vessel on the detector during sample activity measurement.

JRC-Geel also measured it according to a conventional configuration; however, JRC-Geel used two types of vessels when measuring the activity of samples: the vessel the NMIJ/AIST distributed was used for measurement of sample No. 48, and sample No. 79 was poured into the Teflon container that JRC-Geel normally uses. The average value of the measurements on the three detectors were used. The sample to detector distance was 20 mm for detectors (1) and (2), and 2 mm for the detector (3).

NIST measured the activity of the sample by applying both non-destructive and destructive methods. When applying the non-destructive method, the activity of the sample was measured in a reproducible geometry by gamma spectrometry. When applying the destructive method, the activity of a solution of ash originated from the brown rice sample was measured by gamma spectrometry (see Appendix 1). Figure A.1 depicts the sample–detector configuration. Combination of the non-destructive and destructive results for Cs-134 and for Cs-137 was carried out according to a procedure recommended by Dr. Ronald Collé of NIST's Radioactivity Group and based on references [11-12].

OAP set the sample 15 cm away from the detector surface when it measured the sample activity.

The NMIJ/AIST reported the activities obtained by measuring the 27 samples for homogeneity evaluation, as described in Section 4. Their associated uncertainties were estimated for measurement of one sample.

9. Results

The measurement results are listed in Table 4 for Cs-134 and Table 5 for Cs-137. Although the technical protocol of the comparison required measurement of the activity (Bq) of the sample, NRSL/INER, NIST and PTKMR-BATAN reported activity per unit mass (Bq kg^{-1}) of the sample. For these cases, the coordinator calculated the activity of the sample by multiplying the reported value by the mass of each sample measured by the NMIJ/AIST before shipping. The NRSL/INER and NPL reported the result of each sample. Therefore, the coordinator calculated the average value of their two samples by weighted mean. The uncertainty component (σ_{ave}) of counting statics of NRSL/INER, and C_{sample} and $C_{reference}$ of NPL were averaged by $\sigma_{ave} = \sigma_1\sigma_2/\sqrt{\sigma_1^2 + \sigma_2^2}$, where σ_1 and σ_2 are the uncertainty components of samples, respectively. The other uncertainty components were averaged by $\sigma_{ave} = (A_1\sigma_1 + A_2\sigma_2)/(A_1 + A_2)$, where A_1 and A_2 are the activities of samples.

The uncertainties budgets of the average values are listed in Table 10 for NSRL/INER and Table 11 for NPL. OAP's results were very different from the others, then the NMIJ/AIST asked OAP to confirm the correctness of the results in January 2016. The NMIJ/AIST received the revised report from OAP in April 2016. The NMIJ/AIST also asked JRC-Geel to give additional information about the uncertainty budgets in 2021. JRC-Geel revised its report and sent to the NMIJ/AIST in December 2021. The half-life of Cs-134 and Cs-137 used for estimating the activities at the reference date are 2.0644 (14) years and 30.05 (8) years, respectively [13-14].

The final laboratory results, their expanded uncertainties and their relative expanded uncertainties are summarised in Table 6 for Cs-134 and Table 7 for Cs-137. The standard uncertainties used in the calculation of the power moderated mean [6] are also listed in Tables 6 and 7. Figures 1 and 2 show the final laboratory results with their expanded uncertainties listed in Tables 6 and 7, respectively. The calculation of the SCRVs and their associated standard uncertainties for Cs-134 and Cs-137 were performed using the PMM [6], following the decision of CCRI(II) in its meeting of May 2013. The PMM [6] of the final results of the participants for Cs-134 was 3.39 Bq and its standard uncertainty was 0.04 Bq at the reference date. In calculation of the PMM for Cs-134, the outlier result of OAP was not used because the coverage factor used for determining rejection was more than 2.5 [6]. In calculation of the PMM for Cs-137, the results of all the participants were used. The PMM for Cs-137 was 6.89 Bq and its standard uncertainty was 0.10 Bq. Standard uncertainties obtained by dividing the reported expanded uncertainties by 2, which was the value of coverage factor (k) used for obtaining the expanded uncertainties, were used as input quantities in calculating the PMM. They are listed in Tables 6 and 7.

The reported uncertainty budgets of each participant are listed in Tables 8-1 to 8-4 for Cs-134 and Tables 9-1 to 9-4 for Cs-137. Considering the uncertainties due to the bottle-to-bottle homogeneities, the agreement of participant's results was good.

Table 4. The measurement results for Cs-134

Participant	Sample number	Activity per unit mass (Bq kg^{-1})[*1]	Expanded uncertainty (Bq kg^{-1})[*1] (k = 2)	Relative expanded uncertainty (%) (k = 2)	Mass of the rice grain (g)	Activity (Bq)[*1]	Expanded uncertainty (Bq)[*1] (k = 2)	Relative expanded uncertainty (%) (k = 2)	Mean date of measurements
NRSL/INER	3	44.4	3.9	8.6	81.00	3.60[*2]	0.32[*2]	8.6	6 Nov. 2013
	17	43.7	3.8	8.6	81.00	3.54[*2]	0.31[*2]	8.6	
	average	-	-	-	81.00	3.57[*2]	0.27[*2]	7.6[*2]	
JRC-Geel	48	-	-	-	81.00	3.31	0.21	6.3	25 Jan. 2014
	79	-	-	-	81.01	3.51	0.19	5.4	15 Jan. 2015
	Final	-	-	-	81.01	3.41	0.24	7.1	-
KRISS	Final	-	-	-	81.00	3.347	0.081	2.42	30 Sept. 2013
NIST	non-destructive	42.5	-	6.0	-	-	-	-	21 Sept. 2013
	destructive	43.5	-	7.0	-	-	-	-	17 Dec. 2013
	Final	43.0	2.3	5.5	81.01	3.48[*2]	0.19[*2]	5.5	-
NPL	32	-	-	-	81.00	3.38	0.31	9.0	15 Dec. 2013
	70	-	-	-	81.01	3.40	0.31	9.0	8 Dec. 2013
	average	-	-	-	81.01	3.39[*2]	0.30[*2]	8.7[*2]	-
OAP	final	-	-	-	81.01	2.5	0.3	10	24 Dec. 2013
PTKMR-BATAN	final	41.74	2.45	5.87	81.00	3.38[*2]	0.20[*2]	5.9	30 Oct. 2013
NMIJ/AIST	-	-	-	-	81.00	3.35	0.28	8.2	1 May 2013

*1 Reference date = 1 Aug. 2013, 0:00 UTC. *2 Calculated by the coordinator.

Half-life of Cs-134 = 2.0644 year, u = 0.0014 year [13]; half-life of Cs-137 = 30.05 year, u = 0.08 year [14].

Table 5. The measurement results for Cs-137

Participant	Sample number	Activity per unit mass (Bq kg^{-1})[*1]	Expanded uncertainty (Bq kg^{-1})[*1] (k = 2)	Relative expanded uncertainty (%) (k = 2)	Mass of the rice grain (g)	Activity (Bq)[*1]	Expanded uncertainty (Bq)[*1] (k = 2)	Relative expanded uncertainty (%) (k = 2)	Mean date of measurements
NRSL/INER	3	86.7	8.0	9.2	81.00	7.02[*2]	0.65[*2]	9.2	6 Nov. 2013
	17	87.4	8.1	9.2	81.00	7.08[*2]	0.66[*2]	9.2	
	average	-	-	-	81.00	7.05[*2]	0.62[*2]	8.7[*2]	
JRC-Geel	48	-	-	-	81.00	6.86	0.47	6.9	25 Jan. 2014
	79	-	-	-	81.01	7.14	0.45	6.3	15 Jan. 2014
	final	-	-	-	81.01	7.00	0.49	7.0	-
KRISS	final	-	-	-	81.00	6.54	0.17	2.6	30 Sept. 2013
NIST	non-destructive	87.5	-	5.4	-	-	-	-	21 Sept. 2013
	destructive	88.4	-	3.6	-	-	-	-	17 Dec. 2013
	final	87.9	3.9	4.4	81.01	7.12[*2]	0.32[*2]	4.4	-
NPL	32	-	-	-	81.00	6.91	0.59	8.6	15 Dec. 2013
	70	-	-	-	81.01	6.97	0.60	8.6	8 Dec. 2013
	average	-	-	-	81.01	6.94[*2]	0.59[*2]	8.5[*2]	-
OAP	final	-	-	-	81.01	6.5	0.7	10	24 Dec. 2013
PTKMR-BATAN	final	88.06	4.97	5.64	81.00	7.13[*2]	0.41[*2]	5.64	30 Oct. 2013
NMIJ/AIST	-	-	-	-	81.00	6.99	0.44	6.2	1 May 2013

*1 Reference date = 1 Aug. 2013, 0:00 UTC. *2 Calculated by the coordinator.

Half-life of Cs-134 = 2.0644 year, u = 0.0014 year [13]; half-life of Cs-137 = 30.05 year, u = 0.08 year [14]

Table 6. Final laboratory results, expanded uncertainties and standard uncertainties for Cs-134

Participant	Activity (Bq)	Expanded uncertainty (Bq) (k = 2)	Relative expanded uncertainty (%) (k = 2)	Standard uncertainty (Bq)
NRSL/INER	3.57	0.27	7.6	0.135
JRC-Geel	3.41	0.24	7.1	0.12
KRISS	3.347	0.081	2.42	0.041
NIST	3.48	0.19	5.5	0.095
NPL	3.39	0.30	8.7	0.15
OAP*1	2.5	0.3	10	0.15
PTKMR-BATAN	3.38	0.20	5.9	0.10
NMIJ/AIST	3.35	0.28	8.2	0.14
Power moderated mean	3.39	0.07	---	0.04

*1 The OAP result was not used for calculation of the PMM.

Table 7. Laboratory final results, expanded uncertainties and standard uncertainties for Cs-137

Participant	Activity (Bq)	Expanded uncertainty (Bq) (k = 2)	Relative expanded uncertainty (%) (k = 2)	Standard uncertainty (Bq)
NRSL/INER	7.05	0.62	8.7	0.31
JRC-Geel	7.00	0.49	7.0	0.245
KRISS	6.54	0.17	2.6	0.085
NIST	7.12	0.32	4.4	0.16
NPL	6.94	0.59	8.5	0.295
OAP	6.5	0.7	10	0.35
PTKMR-BATAN	7.13	0.41	5.64	0.205
NMIJ/AIST	6.99	0.44	6.2	0.22
Power moderated mean	6.89	0.19	---	0.10

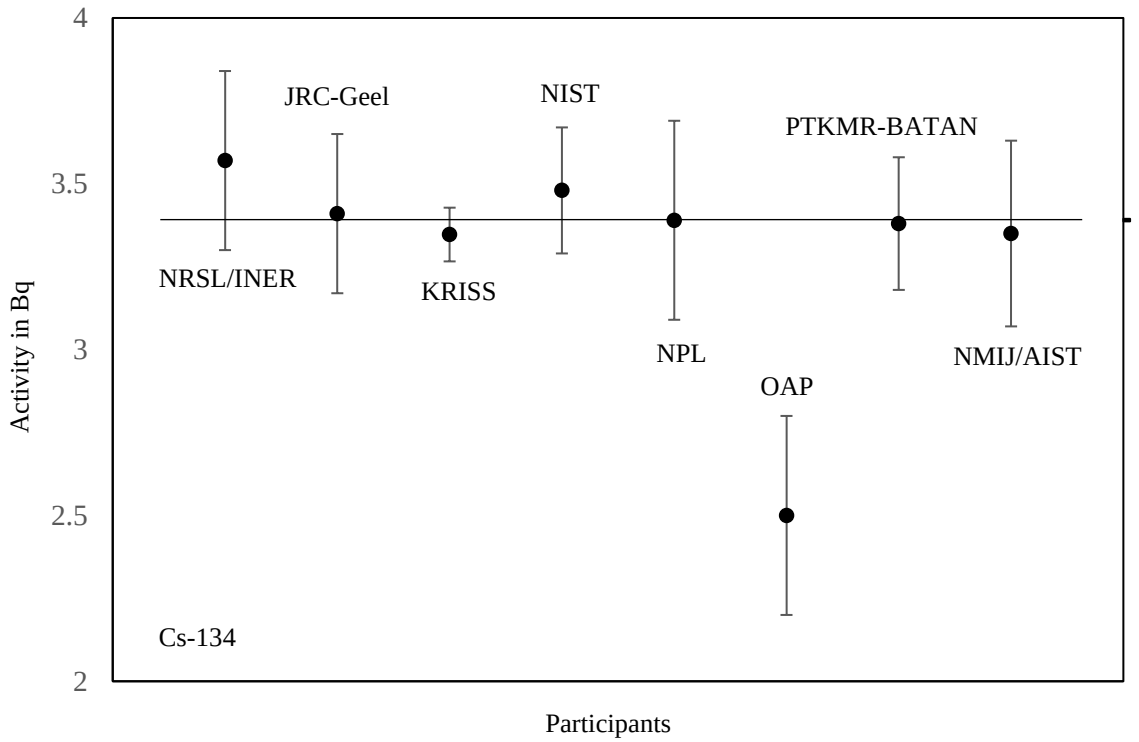

Figure 1. Laboratory results for Cs-134 with the expanded uncertainties (k=2) listed in Table 6. The solid horizontal line indicates the PMM [6] of the final results of the participants except for OAP.

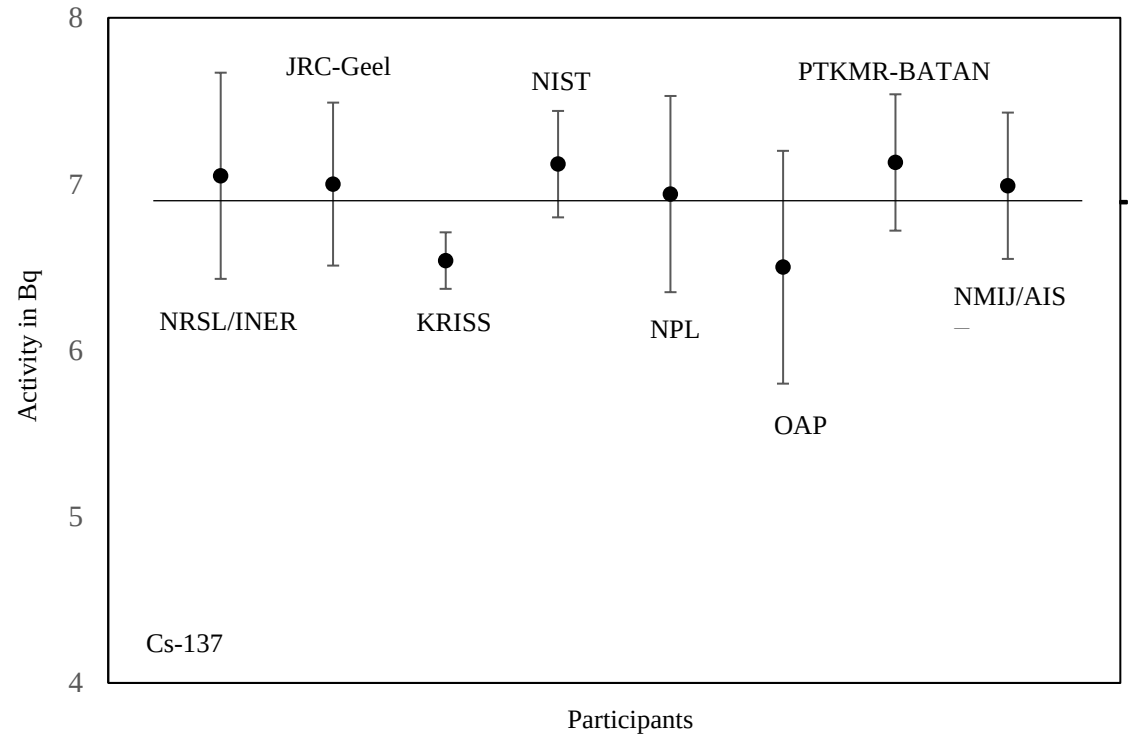

Figure 2. Laboratory results for Cs-137 with the expanded uncertainties (k=2) listed in Table 7. The solid horizontal line indicates the PMM [6] of the final results of the participants.

Table 8-1. Uncertainty components and reported relative values for Cs-134 (10^{-2}).

Item No.	Function	NRSL/ INER	JRC-Geel No. 48	JRC-Geel No. 79	KRISS
1	Counting statistics	2.7	0.7	0.6	0.37
2	Calibration factor	1.9	-	-	0.60
3	Decay correction to reference date	0.1	<0.1	<0.1	0.005
4	Live time correction	0.1	<0.1	<0.1	0.01
5	Sample geometry reproducibility		1.8	1.7	0.10
6	Sample-calibration source self-absorption difference	1.5	1.5	1.5	0.97
7	Instrument stability	2.2	0.1	0.1	0.10
8	Extra model and coincidence summing	-	2	1	-
9	Transfer of sample	-	-	1	-
10	Background	-	-	-	0.00
11	Decay correction for accumulation time	-	-	-	0.0006
12	Original rice weight	-	-	-	-
Combined standard uncertainty		4.3	3.16	2.74	1.21

Table 9-1. Uncertainty components and reported relative values for Cs-137 (10^{-2}).

Item No.	Function	INER	JRC-Geel No. 48	JRC-Geel No. 79	KRISS
1	Counting statistics	1.7	0.7	0.42	0.23
2	Calibration factor	2.1	-	-	0.77
3	Decay correction to reference date	0.1	<0.1	<0.1	0.001
4	Live time correction	0.1	<0.1	<0.1	0.01
5	Sample geometry reproducibility	-	2.7	2.55	0.10
6	Sample-calibration source self-absorption difference	3.5	1.5	1.5	0.98
7	Instrument stability	1.0	0.1	0.1	0.10
8	Extra model	-	1	-	-
9	Transfer of sample	-	-	1	-
10	Background	-	-	-	0.0003
11	Decay correction for accumulation time	-	-	-	0.0002
12	Original rice weight	-	-	-	-
Combined standard uncertainty		4.6	3.43	3.15	1.28

Table 8-2. Uncertainty components and reported relative values for Cs-134 (10^{-2}).

Item No.	Function	NIST*1	NIST*2	OAP	PTKMR-BATAN
1	Counting statistics	0.7	0.7	2	1.33
2	Calibration factor	2.37	1.5	2	1.9
3	Decay correction to reference date	<0.011	<0.011	0.0014	0.039
4	Live time correction	-	-	1.9	0.05
5	Sample geometry reproducibility	1.61	3.1	1.5	1
6	Sample-calibration source self-absorption difference	-	-	1	1
7	Instrument stability	-	-	3	1
8	Extra model and coincidence summing	-	-	-	-
9	Transfer of sample	-	-	-	-
10	Background	-	-	-	-
11	Decay correction for accumulation time	-	-	-	-
12	Original rice weight	0.12	0.12	-	0.5
Combined standard uncertainty		3.0	3.5	4.9	2.94

*1 Non-destructive method.

*2 Destructive method.

Table 9-2. Uncertainty components and reported relative values for Cs-137 (10^{-2}).

Item No.	Function	NIST*1	NIST*2	OAP	PTKMR-BATAN
1	Counting statistics	0.7	0.7	2	1.04
2	Calibration factor	2.07	1.5	2	1.9
3	Decay correction to reference date	<0.003	<0.003	0.0014	0.15
4	Live time correction	-	-	1.9	0.05
5	Sample geometry reproducibility	1.64	0.73	1.5	1
6	Sample-calibration source self-absorption difference	-	-	1	1
7	Instrument stability	-	-	3	1
8	Extra model	-	-	-	-
9	Transfer of sample	-	-	-	-
10	Background	-	-	-	-
11	Decay correction for accumulation time	-	-	-	-
12	Original rice weight	0.12	0.12	-	0.5
Combined standard uncertainty		2.7	1.8	4.9	2.82

*1 Non-destructive method.

*2 Destructive method.

Table 8-3. Uncertainty components and reported relative values for Cs-134 (10^{-2}).

Item No.	Function	NPL No. 32	NPL No. 70
1	A_0: activity of reference source at the reference time	6.9×10^{-1}	6.9×10^{-1}
2	$LT_{reference}$: live time of the reference measurement	2.0×10^{-3}	2.0×10^{-3}
3	LT_{sample}: live time of a sample measurement	4.2×10^{-4}	3.9×10^{-4}
4	C_{sample}: peak counts of a sample	1.6×10^{0}	1.6×10^{0}
5	$C_{reference}$: peak counts of the reference source	1.7×10^{-1}	1.7×10^{-1}
6	λ: decay constant	6.8×10^{-2}	6.8×10^{-2}
7	$t_{m,sample} - t_{m,reference}$: time difference between mid-time of a measurement of the sample and that of the reference	6.7×10^{-4}	3.7×10^{-4}
8	S: self-attenuation correction factor	4.1×10^{0}	4.1×10^{0}
Combined standard uncertainty		4.5×10^{0}	4.5×10^{0}

Table 9-3. Uncertainty components and reported relative values for Cs-137 (10^{-2}).

Item No.	Function	NPL No. 32	NPL No. 70
1	A_0: activity of reference source at the reference time	7.2×10^{-1}	7.2×10^{-1}
2	$LT_{reference}$: live time of the reference measurement	2.0×10^{-3}	2.0×10^{-3}
3	LT_{sample}: live time of a sample measurement	4.2×10^{-4}	3.9×10^{-4}
4	C_{sample}: peak counts of a sample	8.3×10^{-1}	8.5×10^{-1}
5	$C_{reference}$: peak counts of the reference source	2.2×10^{-1}	2.2×10^{-1}
6	λ: decay constant	2.7×10^{-1}	2.7×10^{-1}
7	$t_{m,sample} - t_{m,reference}$: time difference between mid-time of a measurement of the sample and that of the reference	6.7×10^{-4}	3.7×10^{-4}
8	S: self-attenuation correction factor	4.1×10^{0}	4.1×10^{0}
Combined standard uncertainty		4.3×10^{0}	4.3×10^{0}

Table 8-4. Uncertainty components of sample activity determination for Cs-134 (10^{-2}).

Item No.	Function	NMIJ/AIST
Gamma ray spectrometry		-
1	Peak net counts	1.68
2	Background count	0.32
3	Peak counting efficiency	2.25
4	Stability of instrument	0.25
5	Decay	0.0014
6	Measurement time	0.10
7	Effect of sample height	1.29
8	Effect of sample density on self-attenuation	0.52
Bottle-to-bottle homogeneities (s_{bb})		2.62
Combined relative standard uncertainty		4.11
Relative expanded uncertainty (k = 2)		8.2

Table 9-4. Uncertainty components of sample activity determination for Cs-137 (10^{-2}).

Item No.	Function	NMIJ/AIST
Gamma ray spectrometry		-
1	Peak net counts	0.94
2	Background count	0.060
3	Peak counting efficiency	2.25
4	Stability of instrument	0.13
5	Decay	0.00036
6	Measurement time	0.10
7	Effect of sample height	1.50
8	Effect of sample density on self-attenuation	0.14
Bottle-to-bottle homogeneities (s_{bb})		1.10
Combined relative standard uncertainty		3.08
Relative expanded uncertainty (k = 2)		6.2

Table 10. Uncertainty associated to the average value of NSRL/INER (10^{-2}).

Item No.	Function	Cs-134	Cs-137
1	Counting statistics	1.91	1.2
2	Calibration factor	1.9	2.1
3	Decay correction to reference date	0.1	0.1
4	Live time correction	0.1	0.1
5	Sample geometry reproducibility	-	-
6	Sample-calibration source self-absorption difference	1.5	3.5
7	Instrument stability	2.2	1.0
8	Extra model and coincidence summing	-	-
9	Transfer of sample	-	-
10	Background	-	-
11	Decay correction for accumulation time	-	-
12	Original rice weight	-	-
Combined standard uncertainty		3.8	4.4

Table 11. Uncertainty associated to the average value of NPL (10^{-2}).

Item No.	Function	Cs-134	Cs-137
1	A_0: activity of reference source at the reference time	6.9×10^{-1}	7.2×10^{-1}
2	$LT_{reference}$: live time of the reference measurement	2.0×10^{-3}	2.0×10^{-3}
3	LT_{sample}: live time of a sample measurement	4.0×10^{-4}	4.0×10^{-4}
4	C_{sample}: peak counts of a sample	1.13×10^{0}	5.9×10^{-1}
5	$C_{reference}$: peak counts of the reference source	1.2×10^{-1}	1.6×10^{-1}
6	λ: decay constant	6.8×10^{-2}	2.7×10^{-1}
7	$t_{m,sample} - t_{m,reference}$: time difference between mid-time of a measurement of the sample and that of the reference	5.2×10^{-4}	5.2×10^{-4}
8	S: self-attenuation correction factor	4.1×10^{0}	4.1×10^{0}
Combined standard uncertainty		4.4×10^{0}	4.3×10^{0}

10. Preliminary degrees of equivalence

The degree of equivalence of the result of a particular participant, *i*, with the SCRV is expressed as the difference D_i as follows:

$$D_i = x_i - x_{ref},$$

where x_i and x_{ref} are each participant's result and the SCRV obtained by the PMM [6], respectively. The expanded uncertainty (k = 2) of this difference U_i is known as the equivalence uncertainty; therefore,

$$U_i = 2u(D_i).$$

When the result of the NMI *i* is included in the SCRV with a weighting w_i, then

$$u^2(D_i) = (1\text{-}2w_i)\, u_i^2 + u^2(x_{ref}),$$

where u_i is the combined standard uncertainties reported by each laboratory. However, when the result of the NMI *i* is not included in the SCRV, then

$$u^2(D_i) = u_i^2 + u^2(x_{ref}).$$

This was applied to the result of OAP for Cs-134.

The weighting, w_i, obtained by the PMM calculation was listed in Table 12. The preliminary degrees of equivalence for each participant in the comparison are presented in Table 13 for Cs-134 and Table 14 for Cs-137.

Table 12. weighting, w_i, obtained in the PMM calculation

Institute	Cs-134	Cs-137
NRSL/INER	0.0716	0.0820
JRC-Geel	0.0862	0.1088
KRISS	0.4749	0.2338
NIST	0.1244	0.1643
NPL	0.0607	0.0873
OAP	not defined	0.0700
PTKMR-BATAN	0.1147	0.1315
NMIJ/AIST	0.0676	0.1223

Table 13. Degree of equivalence for Cs-134

Participant	Difference, D_i (Bq)	Expanded uncertainty (k = 2) of the difference, U_{Di} (Bq)
NRSL/INER	0.18	0.26
JRC-Geel	0.02	0.23
KRISS	-0.04	0.07
NIST	0.09	0.18
NPL	0.00	0.29
OAP	-0.89	0.31
PTKMR-BATAN	-0.01	0.19
NMIJ/AIST	-0.04	0.27

Table 14. Degree of equivalence for Cs-137

Participant	Difference, D_i (Bq)	Expanded uncertainty (k = 2) of the difference, U_{Di} (Bq)
NRSL/INER	0.16	0.60
JRC-Geel	0.11	0.47
KRISS	-0.35	0.23
NIST	0.23	0.32
NPL	0.05	0.57
OAP	-0.39	0.68
PTKMR-BATAN	0.24	0.40
NMIJ/AIST	0.10	0.43

11. Conclusion

The APMP supplementary comparison of radionuclide activity measurements (APMP.RI(II)-S3.Cs-134.Cs-137) was successfully completed. In total, eight institutes measured sample activity and reported their results. The SCRV for Cs-134 for the reference date of 1st August 2013, 0:00 UTC, was 3.39 Bq, and its standard uncertainty was 0.04 Bq based on the power moderated mean. The SCRV for Cs-137 was 6.89 Bq, and its standard uncertainty was 0.10 Bq.

Acknowledgements

The authors would like to thank all participants in this comparison, especially JRC-Geel, NIST and NPL, which participated from other RMOs. We also appreciate Dr. Yasuhiro Unno of the NMIJ/AIST for his cooperation in preparing the sample and measuring the activity of the sample.

References

[1] T. Miura, A. Yunoki, Y. Unno, S. Hamamatsu, M. Hachinohe and S. Todoriki, The Development of SI Traceable Brown Rice Certified Reference Material for Caesium 134 and Caesium 137 Measurement, RADIOISOTOPES (in Japanese), Vol. 65, 157-167 (2016).

[2] M. Hachinohe, S. Todoriki, Y. Unno, T. Miura, A. Yunoki and S. Hamamatsu, Preparation of Certified Reference Materials for Radioactivity Measurement Using Brown Rice, RADIOISOTOPES (in Japanese), Vol. 65, 169-180 (2016).

[3] Y. Unno, M. Hachinohe, S. Hamamatsu, S. Todoriki, A. Yunoki and T. Miura, Characterization of brown rice as a certified reference material for Fukushima accident-related radioactivity measurements, Applied Radiation and Isotopes 87 (2014) 485-488.

[4] Y. Unno, A. Yunoki, T. Miura, S. Hamamatsu, M. Hachinohe and S. Todoriki, Proficiency Test of Radioactivity Measurement Using Samples of Brown Rice, RADIOISOTOPES (in Japanese), Vol. 65, 181-190 (2016).

[5] JCGM, *Evaluation of measurement data – Guide to the expression of uncertainty in measurement*, JCGM 100:2008

[6] S. Pommé and J Keightley, Determination of a reference value and its uncertainty through a power-moderated mean, *Metrologia* 52 (2015) S200-S212.

[7] Ministry of Agriculture, Forestry and Fisheries, Inspection of agricultural products, Basic guidelines for inspection of agricultural products, Basic guideline I-4, Available at: https://www.maff.go.jp/j/seisan/syoryu/kensa/attach/pdf/hourei-194.pdf (4 November 2021). (In Japanese.)

[8] T. Yamada and Y. Nakamura, Examination of the U8 type polypropylene container used for radioactivity standard volume source, RADIOISOTOPES (in Japanese), Vol. 54, 105-110 (2005).

[9] http://www.nucleide.org/Laraweb/index.php.

[10] https://www.nndc.bnl.gov/nudat2/chartNuc.jsp.

[11] P. Meier, Variance of a Weighted Mean, Biometrics, Vol. 9, No. 1 (1953), 59 – 73.

[12] W. G. Cochran and S. P. Carroll, A Sampling Investigation of the Efficiency of Weighting Inversely as the Estimated Variance, Biometrics, Vol. 9, No. 4 (1953), 447 – 459.

[13] M. –M. Be *et al.*, Monographie BIPM-5, 2013, **Vol. 7** – A = 14 to 245, 73-79.

[14] M. –M. Be *et al.*, Monographie BIPM-5, 2006, **Vol. 3** – A = 3 to 244, 91-98.

[15] ISO, Statistical methods for use in proficiency testing by interlaboratory comparisons, ISO 13528:2022.

Appendix 1 Detector configuration of NIST's destructive sample measurement

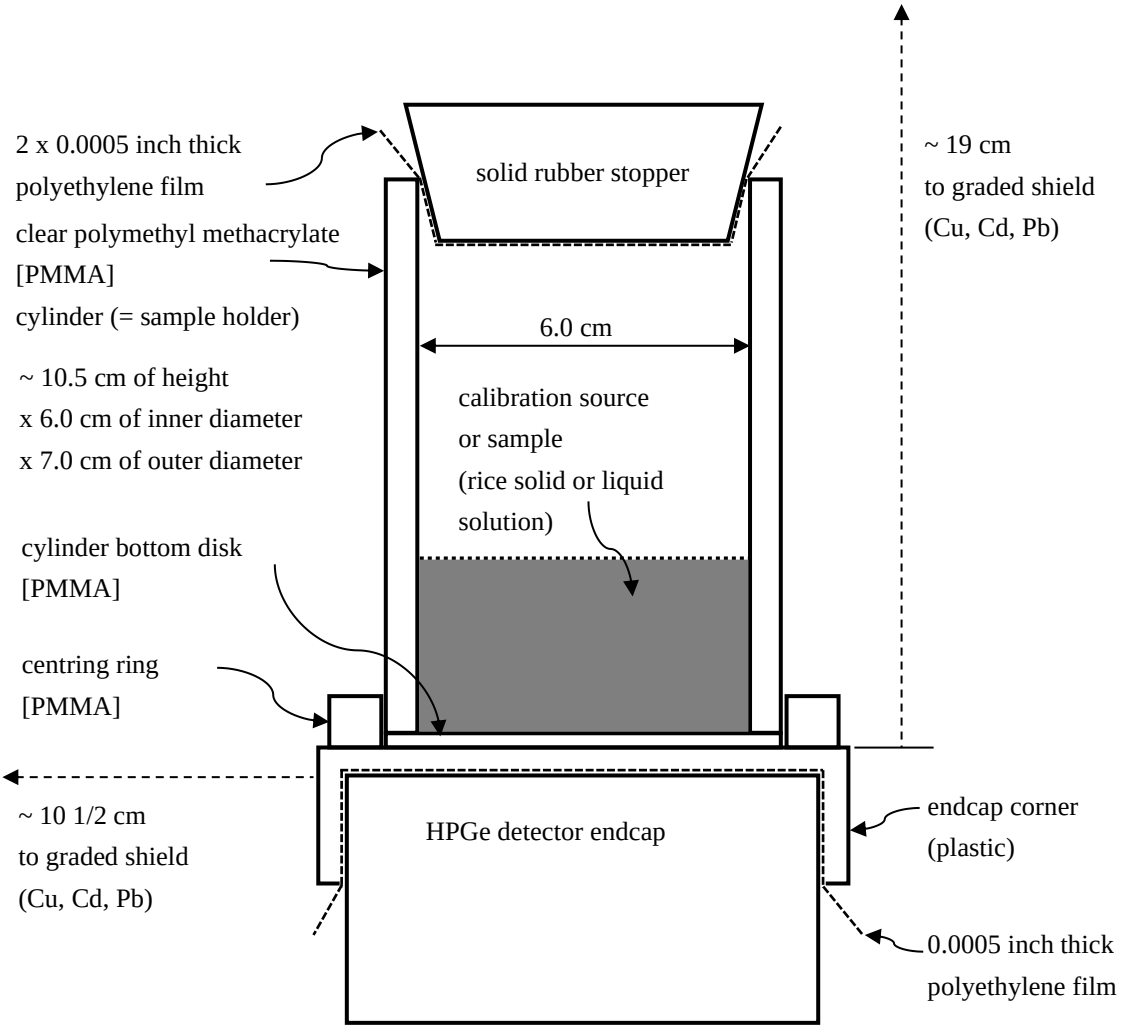

Figure A.1. NIST gamma-ray spectrometry measurement experimental arrangement (scale is approximate)

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**
4π	4P	proportional counter	PC
defined solid angle	SA	press. prop counter	PP
2π	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
Bremsstrahlung	BS	coincidence	CO
gamma ray	GR	anti-coincidence	AC
X-ray	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 rays	MX	triple-to-double coincidence ratio counting	TD
		selective sampling	SS

Example method	**Acronym**
4π(PC)β–γ-coincidence counting	4P-PC-BP-NA-GR-CO
4π(PPC)β–γ-coincidence counting eff. tracing	4P-PP-MX-NA-GR-CT
defined solid angle α-particle counting with a PIPS detector	SA-PS-AP-00-00-00
4π(PPC)AX-γ(GeHP)-anticoincidence counting	4P-PP-MX-GH-GR-AC
4π CsI-β, AX, γ counting	4P-CS-MX-00-00-00
calibrated IC	4P-IC-GR-00-00-00
internal gas counting	4P-PC-BP-00-00-IG

Appendix 3 Evaluation of the comparison results using the E_n score

The result of each participant was evaluated by using the E_n score obtained according to the following formula [15]:

$$E_n = \frac{A_{lab} - A_{ref}}{\sqrt{U_{lab}^2 + U_{ref}^2}}$$

where A_{lab} and U_{lab} are the activity and expanded uncertainty (k=2) obtained by each participant, respectively. A_{ref} and U_{ref} are usually the activity and an expanded uncertainty obtained by the reference laboratories, respectively. However, in this comparison, the A_{ref} and U_{ref} are the SCRV of this comparison and twice its associated standard uncertainty obtained by the power-moderated mean (PMM) [6] for each nuclide, respectively. The $|E_n|$ equal to or larger than 1 could indicate a need to review the uncertainty estimates or to correct a measurement issue [15].

Table B.2. E_n scores for Cs-134 and Cs-137

Participant	E_n score[*1] for Cs-134	E_n score[*2] for Cs-137
NRSL/INER	0.63	0.25
JRC-Geel	0.08	0.21
KRISS	-0.40	-1.37
NIST	0.47	0.62
NPL	0.00	0.08
OAP	-2.89	-0.54
PTKMR-BATAN	-0.04	0.55
NMIJ/AIST	-0.14	0.21

*1 The SCRV for Cs-134 = 3.39 Bq, its standard uncertainty = 0.04 Bq (see Table 6), and its expanded uncertainty (= twice the standard uncertainty) = 0.07 Bq.

*2 The SCRV for Cs-137 = 6.89 Bq, its standard uncertainty = 0.10 Bq, (see Table 7), and its expanded uncertainty (= twice the standard uncertainty) = 0.19 Bq.