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Reference Material for Radionuclides in Sediment, IAEA-384 (Fangataufa Lagoon Sediment)

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Introduction

The IAEA Marine Environment Laboratory (IAEA-MEL) in Monaco has conducted intercomparison exercises on radionuclides in marine samples for many years as part of its contribution to the IAEA's programme of Analytical Quality Control Services (AQCS)[1-3]. An important part of the AQCS programme has been a production of Reference Materials (RMs) and their provision to radioanalytical laboratories. The RMs have been developed for different marine matrices (sediment, water, biota), with accuracy and precision required for the present state of the art of radiometrics and mass spectrometry methods[4-7].

The RMs have been produced as the final products of world-wide intercomparison exercises organised during last 30 years. A total of 44 intercomparison exercises were undertaken and 39 RMs were produced for radionuclides in the marine environment. All required matrices (sewater, biota, sediment) have been covered with radionuclide concentrations ranging from typical environmental levels to elevated levels affected by discharges from nuclear reprocessing plants.

The long-term availability of RMs (over 10 years) requires the use of very specific techniques to collect and pretreat large quantities of material (e.g., in excess of 100 kg) in order to ensure sample stability and homogenization of any analytes of interest. The production of a RM is therefore a long process, covering the identification of needs, sample collection, pre-treatment, homogenisation, bottling, distribution to laboratories, evaluation of data, preliminary reporting, additional analyses in expert laboratories, certification of the material, and finally issuing the RM.

In this paper we describe a reference material IAEA-384, Fangataufa lagoon sediment, designed for determination of anthropogenic and natural radionuclides in the marine environment. This RM has been prepared with the aim of testing the performance of analytical laboratories to measure the activity of these radionuclides in a sediment sample contaminated by elevated levels of fallout from nuclear weapons tests. Participating laboratories were requested to determine as many radionuclides as possible by radiometric (alpha, beta and gamma-spectrometry) as well as by mass spectrometry methods (e.g., ICPMS – Inductively Coupled Plasma Mass Spectrometry, TIMS – Thermal Ionisation Mass Spectrometry, AMS – Accelerator Mass Spectrometry).

Materials and Methods

Description of the material

About 158 kg of sediment was collected by IAEA-MEL in July 1996 in Fangataufa lagoon (French Polynesia), where nuclear weapons testing had been carried out. It was expected therefore that the sediment would contain elevated levels of radionuclides formed during nuclear weapons tests, such as ^{60}Co, ^{90}Sr, ^{137}Cs, ^{155}Eu, ^{238}Pu, $^{239+240}$Pu, ^{241}Pu and ^{241}Am.

After box coring, the sediment was frozen for 2 hours at -40º C, then freeze-dried with a +5º C increase in temperature per hour. Secondary drying was done over 24 hours under a pressure of 0.02 mbar at a constant temperature of +40º C. The sediment was then ground into powder, sieved at 250 µm, homogenized under nitrogen gas and bottled in flasks under nitrogen gas (100 g per bottle).

A particle size analysis has shown that about 58% of the sediment was below 63 µm and 84% below 125 µm. The composition of the sediment is almost 100% Ca(Mg, Sr)CO_3. The final density of the material was 2.64 ± 0.06 g cm^{-3}. The moisture content of the sediment was determined by drying several aliquots in an oven at 80 ºC to constant weight (1-2 days) and was found to be 1.3% at the time of sample preparation. It is recommended that the water content be checked prior to use and that all results be reported on a dry-weight basis.

Sample dispatch and data return

The sample aliquots were distributed to participating laboratories during 1997-1998. There were 110 laboratories world-wide that agreed to participate. Each participant received 100 g of sediment sample. For each radionuclide analysed, the following information was requested: (i) the average weight of sample used for analysis; (ii) the number of analyses performed; (iii) the net massic activity (Bq kg^{-1} dry weight) (iv) an estimate of the total uncertainty for each measurement provided to include counting, and other randon and systematicerrors; (v) a brief description of chemical procedure and counting equipment used; (vi) a list of reference standard solutions used during the analysis; (vii) an estimate of chemical recovery; and (viii) information on count times and decay corrections used in the data reduction.

A total of 94 sets of results were received from participants and included in the evaluation report of the intercomparsison exercise.[6] High precision data from the intercomparison exercise and some additional data received from expert laboratories were included in the certification process, results of which are reported in the present paper.

Data treatment

The massic activities of 15 anthropogenic and 21 natural radionuclides were reported. Laboratory means were calculated when necessary from individual results and are given as weighted means with weighted uncertainties. All values have been rounded off to the most significant figure keeping in mind the necessity of maintaining the uniformity of presentation. The statistical programmes used for data evaluation were the same as described in previous IAEA-MEL AQCS reports.[5] Briefly, the data treatment consists of identifying and eliminating the outlying values, calculating the median and the confidence intervals. Calculations are based on the assumption of non-parametric distribution of data to which distribution-free statistics are applicable. The "less than" values are segregated from the results and the remaining values are checked for the presence of outliers using a box and whisker plot test. Outliers were then removed from the data set and medians were calculated from the results passing the test. These values are considered to be the most reliable estimates of the true values. Confidence intervals were taken from a non-parametric sample population. They represent a two-sided interval at the 95% confidence level.

Following the IUPAC[8] and ISO[9] recommendations for assessment of laboratory performance, Z-score methodology has been used in the evaluation of results. The Z-score is calculated according to the formula:

$$Z = (x_i - x_a) / s_b,$$

where x_i is the robust mean of the values of massic activity in the sample reported by laboratory i, x_a is the assigned value (a mean value of accepted results), and s_b is the target standard deviation.

The selection of the right target value depends on the objectives of the exercise. For radionuclide analysis, laboratories are required to have a relative bias below 20% (s_b <10%). The uncertainty of the assigned value (s_{tu}) has been taken into account using the formula[10]

$$Z = (x_i - x_a) / \sqrt{s_b^2 + s_{tu}^2}$$

where Z, x_i, x_a, s_b and s_{tu} are as previously assigned.

The performance of laboratories in terms of accuracy has been expressed by Z-scores calculated for each radionuclide. The performance is considered to be acceptable if the difference between the robust mean of the laboratory and the assigned value (in s_b units) is less than or equal to two. A Z-score from 2 to 3 indicates that the results are of questionable quality. If $|Z|$>3, the measurement is rejected.

The presented distributions of Z-scores are symmetric (outliers were excluded from the evaluation) which indicates that the performances of the laboratories were satisfactory (usually with Z-score values below 2). A typical example of the Z-score analysis for ^{60}Co is shown in Fig. 1. The Z-score evaluation represents a simple method which gives participating laboratories a normalized performance score for bias.

Criteria for certification

The certification process was carried out following ISO Guide 35[11] using the most precise data from the intercomparison exercise and additional data from expert laboratories. For data sets comprising 5 or more accepted laboratory means, median values and confidence intervals were calculated as estimations of true massic activities. The median was considered as the certified value when:

1. At least 5 laboratory means were available, calculated from at least 3 different laboratories, and;
2. The relative uncertainty of the median did not exceed ± 5% for activities higher than 100 Bq kg^{-1}, ± 10% for activities from 1 to 100 Bq kg^{-1} and ± 20% for activities lower than 1 Bq kg^{-1}.

An activity value was classified as an information value when at least 5 laboratory means of the same order of magnitude calculated from the results of at least 2 different laboratories were available.

Results and discussion

Homogeneity tests

The homogeneity of the sample was checked by measuring the activity of ^{60}Co, ^{137}Cs, ^{155}Eu, ^{210}Pb, ^{226}Ra, $^{239+240}$Pu and ^{241}Am on 30 bottles taken at random. Gamma-spectrometric measurements were performed on 10 to 220 g samples, ^{238}Pu, $^{239+240}$Pu and ^{241}Am were determined by alpha spectrometry on 0.1 to 5 g samples. Homogeneity was determined using one-way analysis of variance. The coefficient of variation was below 10% for both gamma and alpha-spectrometrically determined radionuclides. The "between samples" variances showed no significant differences from the "within sample" variances for all radionuclides tested. Thus the material could be considered homogeneous for the tested radionuclides at the weights used.

Anthropogenic radionuclides with certified activities

Results of analysis of 10 anthropogenic radionuclides (^{60}Co, ^{90}Sr, ^{137}Cs, ^{155}Eu, ^{238}Pu, ^{239}Pu, ^{240}Pu, $^{239+240}$Pu, ^{241}Pu and ^{241}Am) in Fangataufa lagoon sediment as supplied by participants were evaluated. The evaluation of the full data set consisted of identifying and eliminating outlying values, then calculating the median and the confidence intervals (95% significance level). The obtained results are presented in Table 1 for radionuclides with certified values and in Table 2 for radionuclides with information values. A number of accepted laboratory means were used to calculate the certified massic activities and the corresponding confidence intervals.

As an example, Figs. 2 to 4 present the evaluation results with the corresponding standard deviations in order of ascending massic activity for ^{60}Co, $^{239+240}$Pu and ^{241}Am. Also shown are the distribution medians (full lines) and corresponding confidence intervals (dashed horizontal lines).

^{60}*Co:* Data representing 45 laboratory means were used in the certification process (Fig. 2). Non-destructive gamma-spectrometry was used in the analyses. The data show very good homogeneity, they fall less than two standard deviations from the distribution mean. Z-score values are below 2.2 showing reasonable performances by the laboratories (Fig. 1). The median, given as the certified value, is 2.50 Bq kg^{-1} dry weight [95% confidence interval is (2.40 – 2.60) Bq kg^{-1}].

^{155}*Eu:* 30 laboratory means were used in the certification process. Non-destructive gamma-spectrometry was used in the analyses. The data set is homogeneous, within two standard deviations of the distribution mean. Z-score values are below 2.0, showing good performances by the laboratories. The median, given as the certified value, is 6.97 Bq kg^{-1} dw [95% confidence interval is (6.70 – 7.34) Bq kg^{-1} dry weight].

Plutonium isotopes

The majority of participants used conventional methods of sample analysis based on sample treatment, ion-exchange separation followed by electrodeposition and alpha-spectrometry. Some laboratories combined ion-exchange separation with liquid-liquid extraction, or used only the liquid-liquid extraction. Resins (a single TRU column or double UTEVA + TRU columns) for separation and subsequent electrodeposition for alpha-spectrometry (^{238}Pu) and/or for direct ICPMS and AMS analysis (^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu) have also been used. The samples for mass spectrometry analyses were either leached from stainless steel discs after alpha-spectrometry measurements, or analysed directly in mass spectrometers. Generally, a reasonable agreement was obtained between alpha-spectrometry and mass spectrometry results, although these latter were mostly on the right side of the distribution mean.

^{238}*Pu:* 35 data sets obtained by alpha-spectrometry were evaluated. The data is homogeneous, within two standard deviations of the distribution mean. Z-score values are below 2.2 showing reasonable performances by the laboratories. The median, given as the certified value, is 39.0 Bq kg^{-1} dry weight [95% confidence interval is (38.3 – 39.1) Bq kg^{-1}].

$^{239+240}$Pu: 44 data sets obtained by alpha-spectrometry, ICPMS and AMS were included in the evaluation (Fig. 3). The data is homogeneous, within two standard deviations of the distribution mean. Z-score values are below 2.1 showing reasonable performances by the laboratories. The median, given as the certified value, is 107.3 Bq kg^{-1} dry weight [95% confidence interval is (104.2 – 110.0) Bq kg^{-1}].

^{241}Am: 57 results (38 obtained by gamma-spectrometry and 19 by alpha-spectrometry) were available for statistical treatment (Fig. 4). The data falls less than 2 standard deviations from the distribution mean. Generally, the results obtained by gamma-spectrometry were in agreement with alpha-spectrometry data. The combined Z-score values are below 2. The median, given as the certified value, is 7.10 Bq kg^{-1} dry weight [95% confidence interval is (6.96 – 7.40) Bq kg^{-1}].

Natural radionuclides with certified values

Results of analysis of 10 natural radionuclides in Fanfataufa lagoon sediment were evaluated and 6 were certified (^{40}K, ^{210}Pb (^{210}Po), ^{230}Th, ^{234}U and ^{238}U, Table 1) and for 4 radionuclides (^{226}Ra, ^{228}Ra, ^{232}Th and ^{235}U, Table 2) information values were estimated. As an example, Fig. 5 presents the evaluation results with the corresponding standard deviations in order of ascending massic activity for ^{238}U, as obtained by alpha-spectrometry, ICPMS and TIMS.

^{40}K: 25 laboratory means obtained by gamma-spectrometry were available for data evaluation. Some of the laboratories had problems with calibration and the correct estimation of the background under the ^{40}K photopeak. The data show reasonable homogeneity. Z-score values of accepted data are below 2.1, showing reasonable performances by the laboratories. The median, given as the certified value, is 6.80 Bq kg^{-1} dry weight [95% confidence interval is (6.50 – 7.10) Bq kg^{-1}].

^{210}Pb and ^{210}Po: 27 and 7 laboratory means were available for ^{210}Pb and ^{210}Po, respectively. Mostly gamma and alpha-spectrometry (^{210}Po ingrowth) were used for ^{210}Pb, and alpha-spectrometry for ^{210}Po. The Z-score values were below 1.7. Taking into account the time elapsed between collection and analysis of the sediment sample, it seems reasonable to assume that ^{210}Pb and ^{210}Po are in secular equilibrium. As the precision of alpha-spectrometry results was much better, the median was calculated from 13 laboratory means obtained by alpha-spectrometry only. The median given as the certified value is 22.3 Bq kg^{-1} dry weight [95% confidence interval is (22.2 – 22.7) Bq kg^{-1}].

^{230}Th: 10 reported laboratory means were accepted in the evaluation. The data set was homogeneous, and gamma-spectrometry gave results similar to alpha-spectrometry. The Z-score values are below 2.3. The median, given as the certified value, is 2.50 Bq kg^{-1} dry weight [95% confidence interval is (2.38 – 2.61) Bq kg^{-1}].

^{234}U: 18 laboratory means were accepted for the evaluation. Mostly total dissolution followed by alpha-spectrometry, with the exception of 3 results obtained by ICPMS, was used in the analysis. The Z-score values were below 1.8 showing very good performances by the laboratories. The median, given as the certified value, is 40.6 Bq kg^{-1} dry weight [95% confidence interval is (40.0 – 43.5) Bq kg^{-1}].

^{238}U: 11 laboratory means obtained by gamma-spectrometry, 15 obtained by alpha-spectrometry, 2 by ICPMS and 1 by TIMS were used in the evaluation. As the precision and accuracy of gamma-spectrometry results was lower, they were not included in the certification. The Z-score values were below 1.8. The median of the combined alpha and mass spectrometry data (Fig. 5) gives the certified value of 35.5 Bq kg^{-1} dry weight [95% confidence interval is (33.4 – 36.8) Bq kg^{-1}].

Anthropogenic radionuclides with information values

^{90}Sr: 10 laboratory means available for the evaluation show reasonable homogeneity, all fall less than two standard deviations from the distribution mean. Z-score values are below 1.7, showing good performances by the laboratories. The median, given as the information value, is 1.71 Bq kg^{-1} dry weight [95% confidence interval is (1.53 – 1.82) Bq kg^{-1}].

^{137}Cs: The evaluated data set based on 26 laboratory means is homogenous within two standard deviations of the distribution mean. Z-score values are below 2.2, showing reasonable performances by the laboratories. The median, given as the information value, is 0.30 Bq kg^{-1} dry weight [95% confidence interval is (0.24 – 0.50) Bq kg^{-1}].

^{239}Pu: 8 laboratory means (obtained by ICPMS and AMS) were available for data treatment, all results passed the outlier test. Z-score values are below 1.7. The median, given as the information value, is 98.5 Bq kg^{-1} dry weight [95% confidence interval is (84.6 – 104.7) Bq kg^{-1}].

^{240}Pu: 8 laboratory means were available for data treatment, all results passed the outlier test. Z-score values are below 1.7. The median, given as the information value, is 17.5 Bq kg^{-1} dry weight [95% confidence interval is (15.1 – 18.7) Bq kg^{-1}].

^{241}Pu: As ^{241}Pu is pure beta-emitter, usually liquid scintillation spectrometry (LSS) was used for its determination (either directly on prepared liquid samples, or on discs (or leached discs) after alpha-spectrometry analysis). Alpha-spectrometry of the daughter ^{241}Am, and ICPMS analyses were also used. Eight laboratory means were available for the evaluation, however, as some of the ICPMS results were affected by analytical problems (e.g. contamination, calibration, etc.), they were not included in the calculation of the median. The Z-score values are below 1.5. The median, given as the information value, is 55.0 Bq kg^{-1} dry weight [95% confidence interval is (41.0 – 69.0) Bq kg^{-1}].

Natural radionuclides with information values

^{232}Th: Alpha-spectrometry, gamma-spectrometry and mass spectrometry techniques were used in analyses. Only 5 laboratory means were accepted in the analysis, 3 produced by alpha-spectrometry and 2 measurements by ICPMS. The Z-score values are below 1.4, showing good performances by the laboratories. The median, given as the information value, is 0.022 Bq kg^{-1} dry weight {95% confidence interval is (0.008 – 0.024) Bq kg^{-1}}.

^{235}U: 17 gamma-spectrometry results, 11 alpha-spectrometry and 2 ICPMS and 1 TIMS results were used in the evaluation. The Z-score values are below 2. As the precision and accuracy of gamma-spectrometry measurements was lower, the median was calculated using the alpha and mass spectrometry results only. The median, given as the information value, is 1.70 Bq kg^{-1} dry weight [95% confidence interval is (1.60 – 1.96) Bq kg^{-1}].

^{226}Ra: 21 laboratory means obtained by gamma-spectrometry were available for evaluation. The Z-score values are below 2. The median, given as the information value, is 2.41 Bq kg^{-1} dry weight [95% confidence interval is (2.00 – 2.80) Bq kg^{-1}].

^{228}Ra: 5 laboratory means obtained by gamma-spectrometry were available for the evaluation. The median, given as the information value, is 1.60 Bq kg^{-1} dry weight [95% confidence interval is (0.80 – 2.00) Bq kg^{-1}].

Less frequently reported radionuclides

^{228}Th: The average massic activity from four laboratory means obtained by alpha and gamma-spectrometry is (1.6 ± 1.4) Bq kg^{-1} dry weight.

^{236}U: One result obtained by AMS gave a massic activity of (8.4 ± 1.7) 10^{-6} Bq kg^{-1} dry weight.

^{237}Np: Three laboratory means obtained by alpha-spectrometry were available for the evaluation. The average massic activity is (0.010±0.003) Bq kg^{-1} dry weight.

^{242}Pu: One result obtained by ICPMS and one by AMS gave massic activities of (1.93 ± 0.46) Bq kg^{-1} dry weight and (0.66 ± 0.03) Bq kg^{-1} dry weight, respectively.

Isotopic ratios

^{137}Cs/^{90}Sr: The average ^{137}Cs/^{90}Sr activity ratio is 0.21±0.08, much lower than the global fallout ratio (1.5), due to the enhanced concentration of ^{90}Sr in the sediment of Fangataufa lagoon.

^{137}Cs/^{239,240}Pu: The average ^{137}Cs/^{239,240}Pu activity ratio is 0.003±0.001, lower by 3 orders of magnitude than the global fallout ratio due to the enhanced concentration of plutonium in the sediment.

^{238}Pu/$^{239+240}$Pu: The average ^{238}Pu/$^{239+240}$Pu activity ratio is 0.362 ± 0.003, much higher than the global fallout value 0.025, documenting that plutonium found in the lagoon is of nuclear weapons origin.

^{241}Pu/^{239,240}Pu: The average ^{241}Pu/^{239,240}Pu activity ratio is 0.52 ± 0.06, much smaller than the global fallout ratio (15), due to the enhanced concentration of ^{239}Pu in the sediment.

^{241}Am/$^{239+240}$Pu: The average ^{241}Am/$^{239+240}$Pu activity ratio is 0.066 ± 0.003, much smaller than the global fallout ratio (0.40), again due to the enhanced concentration of ^{239}Pu in the sediment.

^{240}Pu/^{239}Pu: The average ^{240}Pu/^{239}Pu activity ratio from 8 measurements made by ICPMS and AMS and 1 by high-resolution alpha-spectrometry is 0.179 ± 0.002, much lower than the global fallout ratio (0.66). The average ^{240}Pu/^{239}Pu mass ratio is 0.049 ± 0.001 (the global fallout ratio is

0.180), documenting again that the origin of plutonium in Fangataufa lagoon is from close-in fallout from nuclear weapons tests.

Conclusions

The accurate and precise determinations of radionuclide concentrations in marine samples have been important implications in assessing the fate and transport of radionuclide in the marine environment and in studies of the oceanographic processes. To address the problem of data quality, and to assist IAEA's Member States in verifying the performance of their laboratories, the IAEA-MEL has been developing RMs, which represent important benchmarks in quality management of laboratories.

In this paper a Fangataufa lagoon sediment sample (IAEA-384) has been described and data on anthropogenic and natural radionuclides have been evaluated. The medians of massic activities with 95 % confidence intervals have been chosen as the most reliable estimates of the true values. The Fangataufa lagoon sediment (IAEA-384) has been certified as a RM for 11 radionuclides [^{40}K, ^{60}Co, ^{155}Eu, ^{210}Pb (^{210}Po), ^{230}Th, ^{234}U, ^{238}U, ^{238}Pu, $^{239+240}$Pu and ^{241}Am], and information values have been estimated for 9 radionuclides (^{90}Sr , ^{137}Cs, ^{226}Ra, ^{228}Ra, ^{232}Th, ^{235}U, ^{239}Pu, ^{240}Pu, ^{241}Pu). This RM may be used for quality management of radioanalytical laboratories, for identifying suspect methodologies and laboratory training needs, for up-grading the quality of a laboratory'ss performance, and generally assessing the accurately and validity of using different analytical methods. The material is available from the IAEA in 100-g units.

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Table 1. Certified massic activities in IAEA-384 (Fangataufa lagoon sediment)

Radionuclide	Mean ± std. dev. Bq/kg	Median Bq/kg	95% confidence interval (Bq/kg)	Number of lab. means*	Relative uncertainty (%)
^{40}K	6.84 ± 0.45	6.80	6.50 – 7.10	25	8.8
^{60}Co	2.53 ± 0.37	2.50	2.40 – 2.60	45	8.0
^{155}Eu	6.97 ± 0.69	6.97	6.70 – 7.34	30	9.2
^{210}Pb (^{210}Po)	22.2 ± 2.1	22.3	22.2 – 22.7	13	2.2
^{230}Th	2.50 ± 0.21	2.50	2.38 – 2.61	10	9.2
^{234}U	39.2 ± 3.8	40.6	40.0 – 43.5	18	8.6
^{238}U	34.7 ± 3.2	35.5	33.4 – 36.8	18	9.7
^{238}Pu	38.9 ± 2.5	39.0	38.3 – 39.1	35	2.1
$^{239+240}Pu$	107.3 ±6.5	107.3	104.2 – 110.0	44	5.4
$^{241}Am^{\#}$	7.15 ± 0.81	7.10	6.96 – 7.40	57	6.2

Reference date for decay correction: 1 August 1996

* Number of accepted laboratory means which were used to calculate the certified activities and the corresponding confidence intervals.

The value should be corrected for in-growth from ^{241}Pu.

Table 2. Information massic activities in IAEA-384 (Fangataufa lagoon sediment)

Radionuclide	Mean ± std. dev. Bq/kg	Median Bq/kg	95% confidence interval (Bq/kg)	Number of lab. means*	Relative uncertainty (%)
^{90}Sr	1.73 ± 0.36	1.71	1.53 – 1.82	10	17
^{137}Cs	0.36 ± 0.18	0.30	0.24 – 0.50	26	87
^{226}Ra	2.43 ± 0.33	2.41	2.00 – 2.80	21	33
^{228}Ra	1.78 ± 0.42	1.60	0.80 – 2.00	5	75
^{232}Th	0.018 ± 0.008	0.022	0.008 – 0.024	5	73
^{235}U	1.74 ± 0.28	1.70	1.60 – 1.96	14	21
^{239}Pu	97.0 ± 7.0	98.5	84.6 – 104.7	8	20
^{240}Pu	17.2 ± 1.5	17.5	15.1 – 18.7	8	21
^{241}Pu	55.7 ± 5.3	55.0	41.0 – 69.0	8	51

Reference date for decay correction: 1 August 1996.

* Number of accepted laboratory means which were used to calculate the information activities and the corresponding confidence intervals.

Figure captions:

Fig. 1. Z-score values for ^{60}Co in IAEA-384.

Fig. 2 . Data evaluation for ^{60}Co in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

Fig. 3. Data evaluation for $^{239+240}Pu$ in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

Fig. 4. Data evaluation for ^{241}Am in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

Fig. 5. Data evaluation for ^{238}U in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

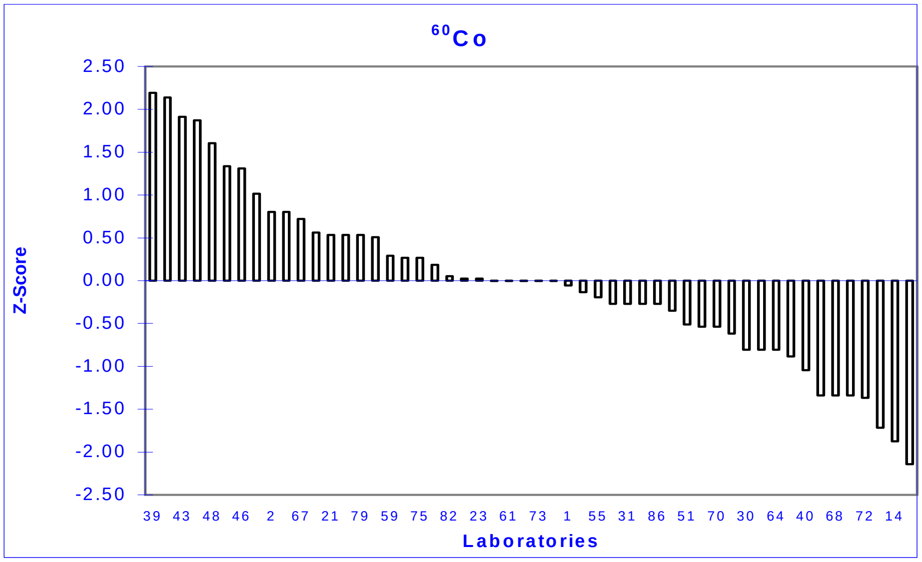

Fig. 1. Z-score values for ^{60}Co in IAEA-384.

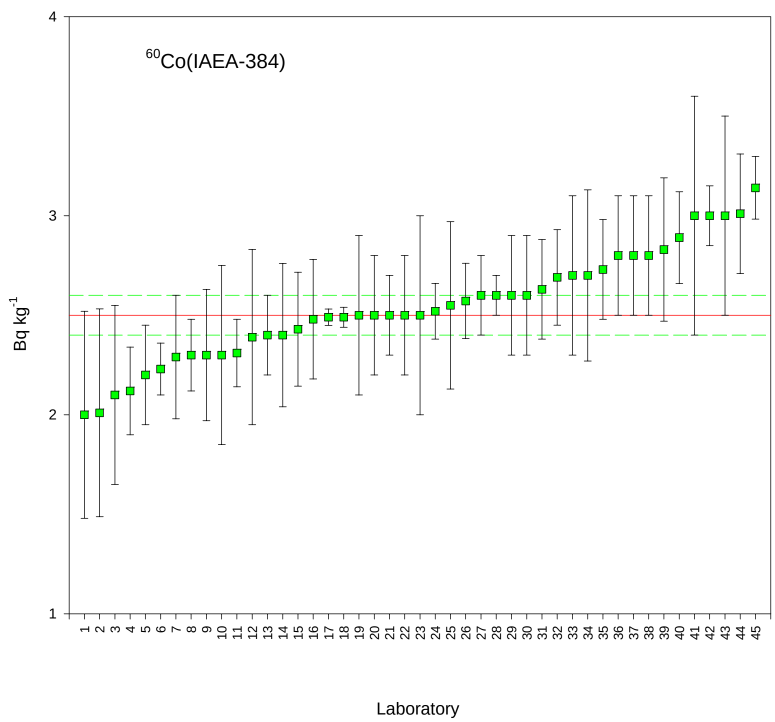

Fig. 2 . Data evaluation for ^{60}Co in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

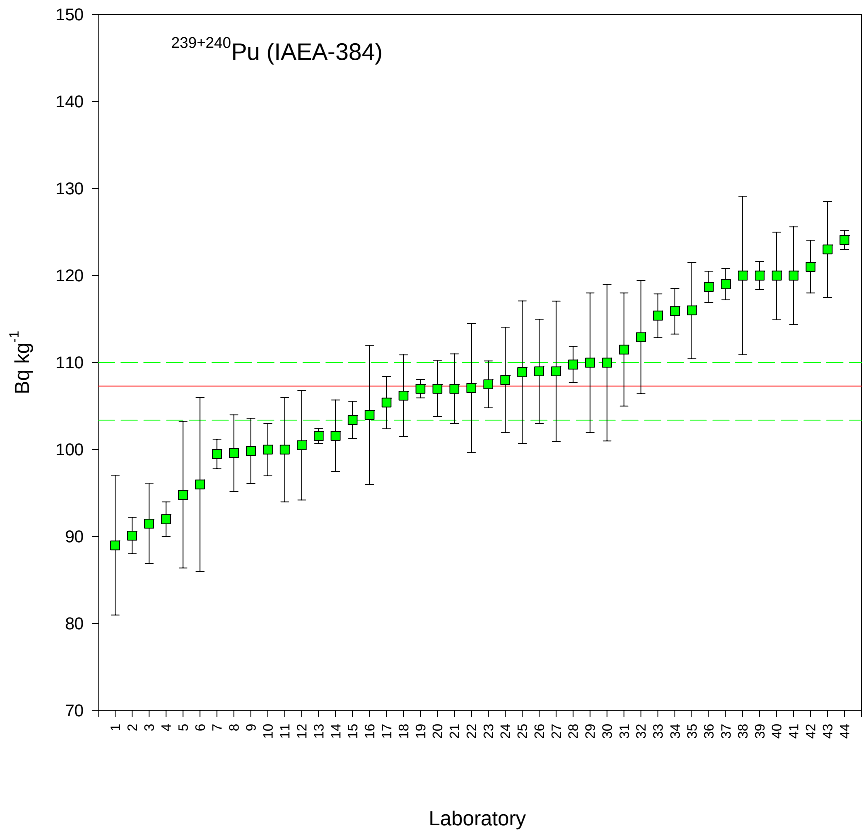

Fig. 3. Data evaluation for $^{239+240}$Pu in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

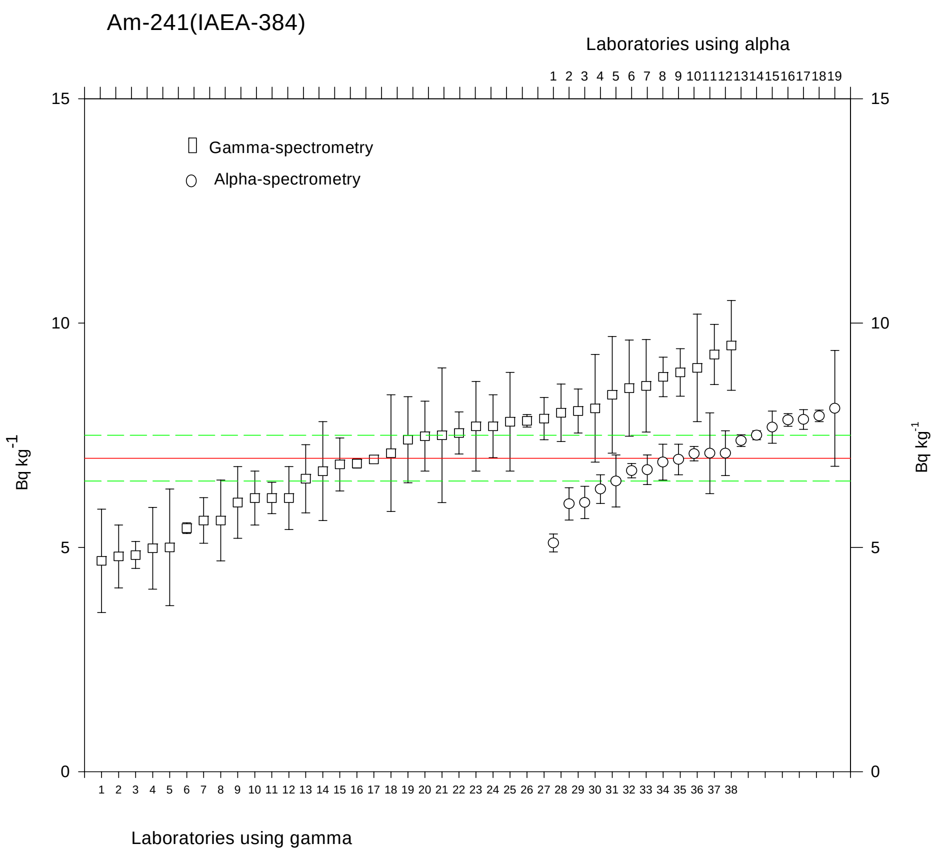

Fig. 4. Data evaluation for ^{241}Am in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

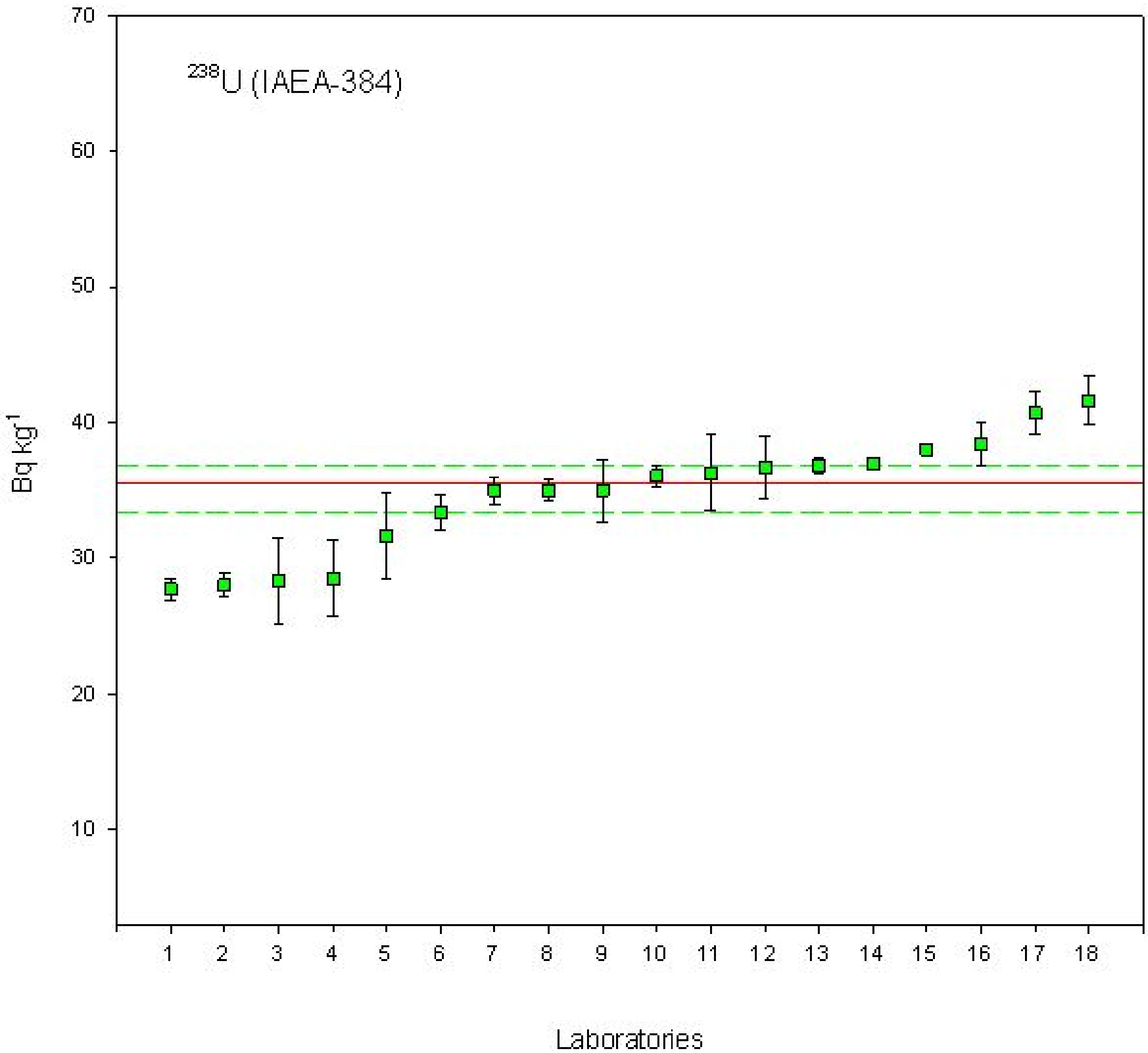

Fig. 5. Data evaluation for ^{238}U in IAEA-384 (only alpha-spectrometry and mass spectrometry results). The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

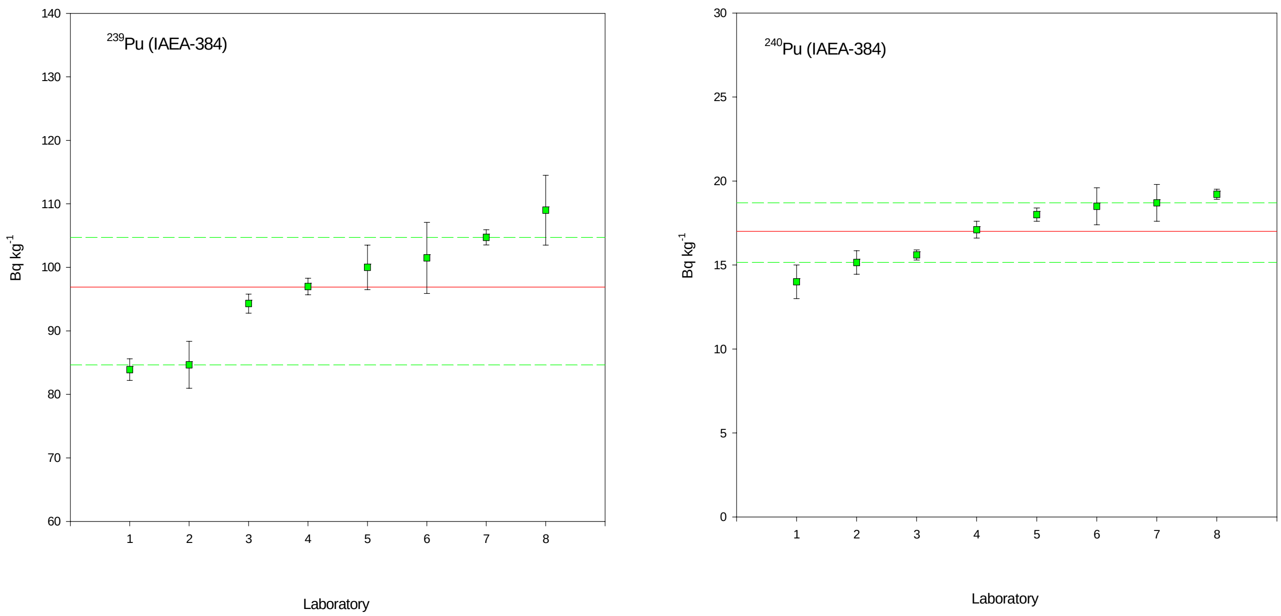

Fig. 6. Data evaluation for ^{239}Pu and ^{240}Pu in IAEA-384. The median (full line) and corresponding 95 % confidence intervals (dashed horizontal lines) are also shown.

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