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ENVIRONMENTAL RADIOACTIVITY INTERCOMPARISON 1993

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ABSTRACT

The results for the fourth in a series of environmental radioactivity intercomparison exercises is detailed. In general, the results are satisfactory and a comparable performance to the previous exercises has been achieved. In particular there has been an improvement in the results for some nuclides where cascade summing has hitherto been a problem.

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Approved on behalf of Director and Chief Executive, NPL,
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1 INTRODUCTION

This exercise was the fourth in a series of environmental radioactivity intercomparison exercises [1, 2, 3, 4, 5], conducted by the National Physical Laboratory's (NPL) Division of Radiation Science and Acoustics (DRSA). Previous exercises had shown the phenomenon of cascade summing and, to a lesser extent, nuclear decay data to be perennial problems. Participants had indicated their on-going interest and their willingness to participate in further exercises. The exercise was organised along similar lines to the previous ones, with two sets of samples; one containing α - and pure β -emitting radionuclides that required extensive chemical analysis and one containing β/γ -emitters.

2 ORGANISATION

2.1 Participants

The list of participants was chosen from the DRSA database; which includes approximately 230 contacts worldwide, although there is inevitably some duplication and redundancy. In the event there were 47 participants, all of whom took the β/γ -emitter containing samples and 24 of whom also took the α - and β -emitting samples. The participants were, as in previous years, from three groups; those with a primary interest in the nuclear industry, ie the industry itself and government departments; those with a secondary interest in the nuclear industry, eg local authorities and research organisations; third, those with no special interest in the nuclear industry, such as contract laboratories and finally, for the first time, laboratories not based in the UK. The participants are listed in section 12 in alphabetical order of the organisation concerned.

2.2 Samples

Two sets of samples were produced. Those containing α - and β -emitters were designated AB xxx/93 and those containing β/γ emitters BG xxx/93. The radionuclides included in the samples were chosen to include those that are emitted (in varying amounts) by the nuclear industry worldwide in the normal course of their operations. The limitations used when choosing the radionuclides for inclusion were (i) that half lives exceeded 100 days, (ii) that no particularly obscure species were chosen, and (iii) that they were all measurable by conventional and well known techniques. For those analysing the AB samples, the radionuclidic content was given to them. This was not done for the BG samples, although a candidate list of nuclides was provided. The levels of each of the nuclides are given in Table 1. It was possible to measure two of the nuclides (^{99}Tc and ^{239}Pu) directly by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), but no-one attempted this.

Table 1 Sample Data

Sample	Nuclide	Emission	Level/ (Bq/kg)
AB xxx/93	^3H	β	~ 500-600
"	$^{90}\text{Sr}/^{90}\text{Y}$	β	~ 50-60
"	^{99}Tc	β	~ 40-60
"	^{238}Pu	α	~ 4-6
"	^{239}Pu	α	~ 10-20
"	^{241}Am	α/γ	~ 10-20
BG xxx/93	^{60}Co	β/γ	~ 80-90
"	$^{106}\text{Ru}/^{106}\text{Rh}$	$\beta, \beta/\gamma$	~ 40-50
"	^{134}Cs	β/γ	~ 10-20
"	$^{137}\text{Cs}/^{137\text{m}}\text{Ba}$	$\beta, \beta/\gamma$	~ 20-30
"	$^{144}\text{Ce}/^{144}\text{Pr}$	$\beta, \beta/\gamma$	~ 30-40
"	^{154}Eu	β/γ	~ 60-70
"	^{155}Eu	β/γ	~ 90-95

3 CALIBRATION

3.1 Absolute (or primary) calibration

The source materials were all calibrated in a manner which ensured traceability to national primary standards. In most cases, this is achieved relatively simply [6] and it is a process worth examining.

An absolute or primary standard of radioactivity is one that has been measured directly without reference to any other standard of activity; the activity has been determined from count-rate data alone, without reference to any supplementary data determined from previous experiments.

Consider a radionuclide decaying, by beta particle emission, to a single excited state of the daughter nuclide, which in turn decays to its ground state by the emission of a gamma ray. The time separation between these two events needs to be sufficiently small such that they can be regarded as occurring simultaneously; for the purposes of the technique described below, simultaneous means within 100 ns or less.

The activity of this radionuclide can be measured absolutely in the following manner. A solution is prepared at an activity concentration of between 50 and 5000 kBq g⁻¹. A few drops are accurately dispensed onto a thin, electrically-conducting plastic film supported on a metallic, annular holder. The liquid is then allowed to evaporate to dryness, leaving an effectively weightless, active deposit. This source is then assayed in a 4 π -proportional counter-gamma-coincidence system.

The basic system comprises two detectors, a gas-flow, proportional counter to detect the beta particles and a NaI(Tl) crystal to detect gamma ray emissions. In the ideal case, neither detector responds to the radiation detected by the other detector. The outputs of the two detectors are compared in time using a coincidence unit (a simple AND gate) in order to register the number of occasions when both a beta particle and a gamma ray are detected simultaneously.

If the absolute activity of the sample is given by N_0 and the detection efficiencies of the beta and gamma detectors are given by ϵ_β and ϵ_γ respectively, the events recorded in the three

channels (beta, gamma and coincidence respectively) are given by:

$$\begin{aligned} N_{\beta} &= N_o \epsilon_{\beta} \\ N_{\gamma} &= N_o \epsilon_{\gamma} \\ N_c &= N_o \epsilon_{\beta} \epsilon_{\gamma} \end{aligned}$$

From these three equations, it can be seen that:

$$N_o = N_{\beta} \cdot N_{\gamma} / N_c = N_o \epsilon_{\beta} \cdot N_o \epsilon_{\gamma} / N_o \epsilon_{\beta} \epsilon_{\gamma}$$

and

$$N_c / N_{\gamma} = \epsilon_{\beta}$$

In practice there is always a finite probability of the beta detector registering a gamma event and the initial activity equation becomes modified to:

$$N_{\beta} \cdot N_{\gamma} / N_c = N_o [1 + k (1 - \epsilon_{\beta}) / \epsilon_{\beta}]$$

where k is a constant.

In practice the detection of every β event ($\epsilon_{\beta} = 1$) cannot be achieved and it becomes necessary to vary the beta counting efficiency. If the recorded values of $N_{\beta}N_{\gamma}/N_c$ are plotted against the corresponding values of $(1 - \epsilon_{\beta}) / \epsilon_{\beta}$ and the resulting curve is extrapolated to the point where ϵ_{β} is equal to 1, N_o is obtained. This method is known as the 4π (proportional counter)-gamma-coincidence counting technique.

This technique can be used for all radionuclides which decay by particle emission followed by simultaneous gamma ray emissions; such radionuclides include ^{241}Am , ^{60}Co , ^{134}Cs , ^{154}Eu and ^{155}Eu . For those parent-daughter systems where the parent decays only by particle emission but the daughter is relatively short-lived and decays by coincident particle-gamma ray emissions, the same technique may be applied; $^{106}\text{Ru}/^{106}\text{Rh}$ and $^{144}\text{Ce}/^{144}\text{Pr}$ fall into this category.

For those radionuclides (such as ^{90}Sr , ^{99}Tc , ^{238}Pu and ^{239}Pu) which decay effectively only by particle emission or where the gamma ray is significantly delayed, a tracer technique is used. A radionuclide which decays by both particle and coincident gamma ray emission is selected on the basis of the particulate energy spectrum being similar to that of the pure particle emitter. By combining known quantities (in terms of activity) of the tracer and then counting samples of the mixture by the coincidence counting technique, it is possible to determine the activity of the pure particle emitter (ppe). In essence, the observed particle counting rate may be written as:

$$N_{\beta\text{total}} = N_{\text{otracer}} \{1 - (1 - \epsilon_{\beta\text{tracer}})\} + N_{\text{oppe}} \{1 - (1 - \epsilon_{\beta\text{ppe}})\}$$

Since the pure particle emitter cannot contribute to either the gamma or coincidence counting rates, the ratio of N_c/N_{γ} is equal to $\epsilon_{\beta\text{tracer}}$. By careful matching of tracer and pure particle emitter spectral characteristics, it allows the assumption to be made that

$$(1 - \epsilon_{\beta\text{ppe}}) = a(1 - \epsilon_{\beta\text{tracer}})$$

where a is a constant.

As with the coincidence method, if $\epsilon_{\beta\text{tracer}}$ is varied, one may then plot values of $N_{\beta\text{total}}$ against values of $(1 - \epsilon_{\beta\text{tracer}})$. By extrapolating to the point where $(1 - \epsilon_{\beta\text{tracer}})$ is equal to zero, the value of N_{oppe} may be derived.

Tritium presents special difficulties. It is standardised in the gaseous phase using a series of

cylindrical proportional counters which have identical diameters but different lengths, which allows end effects to be eliminated.

3.2 Secondary Standardisation

The absolute counting techniques described above suffer from the disadvantages that they are labour intensive, require a high level of operator expertise and a limit is placed on the lowest level of activity that can be measured since the uncertainties become unacceptably high. All of this requires a more robust and routine method of calibration to be used. The approach taken is to use the absolutely calibrated material - in most cases a solution - to calibrate a secondary standard instrument. In the case of α/γ - or β/γ -emitters this is usually a well type ionisation chamber that responds only to the γ rays emitted. NPL's ion chambers have been operating for over 40 years with a variation in this time of less than $\pm 0.2\%$. Sodium iodide detectors or hyper pure Germanium γ -ray spectrometers (HPGe) can also be used but they suffer from a lack of long term stability. For pure α - or most pure β -emitters, there is no satisfactory secondary standard instrument.

4 SOURCE PREPARATION

4.1 Purity

The raw materials for the sources were procured from a number of suppliers and the originator, along with the laboratory responsible for calibration, are given in Table 2 below.

In each case, the starting material was a single nuclide standard. Each of the materials was checked by γ -spectrometry for impurities and this is detailed in Table 3. It was not felt that any of the impurities present seriously compromised the sources prepared.

Table 2 Radionuclide Provenance

Nuclide	Origin	Calibrating Laboratory	Traceability
^3H	AIL	AIL	US Primary Standard/NIST
$^{90}\text{Sr}/^{90}\text{Y}$	AIL	NPL	UK Primary Standard/NPL
^{99}Tc	CEA/France	NPL	UK Primary Standard/NPL
^{238}Pu	AIL	AIL	UK Primary Standard/NPL
^{239}Pu	AEA, Harwell	AEA/NPL	UK Primary Standard/NPL
^{241}Am	AEA, Harwell	NPL	UK Primary Standard/NPL
^{60}Co	AIL	NPL	UK Primary Standard/NPL
^{106}Ru	AIL	NPL	UK Primary Standard/NPL
^{134}Cs	AIL	NPL	UK Primary Standard/NPL
^{137}Cs	AIL	NPL	UK Primary Standard/NPL
$^{144}\text{Ce}/^{144}\text{Pr}$	AIL	NPL	UK Primary Standard/NPL
^{154}Eu	NPL	NPL	UK Primary Standard/NPL
^{155}Eu	UVVVR, Czech Republic	NPL	UK Primary Standard/NPL

Table 3 Radionuclide impurities

Nuclide	Impurity	Level/(% of main nuclide)
^{238}Pu	^{239}Pu	0.1
^{239}Pu	^{238}Pu	0.1
	^{240}Pu	0.53
	^{241}Pu	8.4
^{154}Eu	^{152}Eu	0.4
	^{155}Eu	1.1
^{155}Eu	^{154}Eu	1.0

4.2 Activity Confirmation

The activity of the starting materials was checked either by liquid scintillation counting (in the case of the pure α - and β -emitters) or by γ -spectrometry.

4.3 Preparation

Stock solutions were prepared by diluting the required amount of each radionuclide into either the α/β stock solution or the β/γ stock solution. These solutions were then used to prepare the participants samples by diluting ~ 2.5 g to ~ 500 g. All samples were made up to be ~ 0.1M with respect to nitric acid. In addition, carriers were added, where appropriate. The AB xxx/93 samples had strontium and yttrium added as carriers, there being no suitable carriers for technetium, plutonium or americium. The aqueous matrix of this sample acts as a carrier for ^3H . The BG xxx/93 samples had cobalt, ruthenium, rhodium, caesium, barium, cerium, praseodymium and europium carriers added. All carriers were present at a level of approximately 50 $\mu\text{g/g}$.

5 TREATMENT OF DATA

5.1 Numerical data

A data collection form was constructed for each set of samples. This was devised in the light of the data to be collected and in view of comments from participants in previous exercises. These forms were distributed with the samples. As is the usual practice for these intercomparisons, the style of reporting does not allow the reported data to be assigned to a particular participant.

The practice established in previous exercises was followed [1, 2, 3, 4, 5], but it is re-iterated here for the benefit of the participants. Two main parameters are calculated. First, the deviation is measured as a percentage:

$$\text{Deviation} = (\text{Analyst result} - \text{NPL value}) \cdot 100\% / \text{NPL value}$$

The value of the u-statistic [8] is also calculated;

$$\text{u-statistic} = |(\text{Analyst result} - \text{NPL value})| / [(\text{Analyst uncertainty})^2 + (\text{NPL uncertainty})^2]^{1/2}$$

This statistic is compared with values in the t-statistic tables [9] and assessed according to the criteria in Table 4. It should be noted that this comparison is somewhat subjective; there are no hard and fast rules.

Table 4 u-test criteria

Condition	Comment
$1.64 > u$	The reported result does not differ from the actual value
$1.95 > u > 1.64$	The reported result probably does not differ from the actual value, but more data is required to support this
$2.58 > u > 1.95$	It is not clear whether the results differ or not
$3.29 > u > 2.58$	The reported result is probably different from the actual value, but more data is required to support this
$u > 3.29$	The reported result differs from the actual value

5.2 Nuclear decay data

The participants approach to this is described later, but the data used by NPL for this work is given in Table 5. In each case P_α , P_β and P_γ values (the emission probabilities) are only given if they are greater than 0.01.

Table 5 - Nuclear decay data

Nuclide	Half life (days)	E_γ (keV)	P_γ (%)	Reference
^3H	4507.2 ± 3.7	-	-	[9]
^{60}Co	1925.5 ± 0.5	1173.2	99.857 ± 0.022	[10]
		1332.5	99.983 ± 0.006	[10]
^{90}Sr	10460 ± 60	-	-	[11]
^{99}Tc	$7.72 (\pm 0.04) \times 10^6$	-	-	[11]
^{106}Ru	368.2 ± 1.2	511.9	20.7 ± 0.6	[12]
		621.8	9.81 ± 0.57	[12]
		1050.4	1.50 ± 0.10	[12]
^{134}Cs	754.28 ± 0.22	475.4	1.49 ± 0.02	[10]
		563.2	8.36 ± 0.03	[10]
		569.3	15.39 ± 0.06	[10]
		604.7	97.63 ± 0.06	[10]
		795.9	85.4 ± 0.3	[10]
		801.9	8.69 ± 0.03	[10]
		1168.0	1.792 ± 0.07	[10]
		1365.2	3.016 ± 0.011	[10]

Table 5 - Nuclear decay data (cont)

Nuclide	Half life (days)	E_γ (keV)	$P_\gamma(\%)$	Reference
^{137}Cs	11020 ± 60	661.7	85.1 ± 0.2	[10]
^{144}Ce	284.9 ± 0.2	133.5	11.09 ± 0.2	[13]
		696.5	1.342 ± 0.0014	[13]
^{154}Eu	3136.8 ± 2.9	123.1	41.2 ± 0.5	[10]
		247.9	6.95 ± 0.09	[10]
		591.8	4.99 ± 0.06	[10]
		692.4	1.80 ± 0.03	[10]
		723.3	20.2 ± 0.2	[10]
		756.8	4.58 ± 0.06	[10]
		873.2	12.24 ± 0.15	[10]
		996.3	10.48 ± 0.13	[10]
		1004.7	18.2 ± 0.2	[10]
		1274.4	35.0 ± 0.4	[10]
		1596.5	1.81 ± 0.02	[10]
^{155}Eu	1770 ± 50	60.0	1.14 ± 0.12	[10]/[14]
		86.5	34.0 ± 2.4	[14]
		105.3	20.6 ± 1.4	[14]
^{238}Pu	32030 ± 110	-	-	[15]
^{239}Pu	$8.807(\pm 0.015) \times 10^6$	-	-	[12]
^{241}Am	$1.5785(\pm 0.0024) \times 10^5$	59.5	36.0 ± 0.4	[10]

6 RESULTS FOR α - AND β - EMITTERS

6.1 General Comments

In total, 24 samples were despatched. In all cases where data were returned, radiochemical analysis was performed to resolve the mixture of radionuclides. A variety of techniques were used, which are considered under each radionuclide as appropriate. Although the analysis schemes are more complex than those for the β/γ - emitters, the data processing is simpler. In general, the activity is calculated as follows;

$$\text{Activity} = (\text{net count rate})/\epsilon.Y.W.D$$

where ϵ is the detection efficiency, Y is the chemical yield, W is the sample weight and D is the radioactive decay factor.

The detection efficiency can be determined by calibrating the counter with a suitable

Čerenkov counting on the sample; this was possible since none of the other nuclides present emits particles with a high enough energy to give spurious Čerenkov counts. A number of participants used ^{85}Sr as a yield tracer for ^{90}Sr , the rest using stable strontium. Overall there is a slightly high bias on the results, but it is insignificant.

6.4 ^{99}Tc (Tables 8 and 12)

This nuclide had 8 sets of data returned. The lack of a stable isotope of technetium which complicates the analysis. It is possible to use another isotope of technetium, such as $^{95\text{m}}\text{Tc}$, but this may interfere with the low level determination of ^{99}Tc . All other isotopes of technetium are practically impossible to obtain in a pure state. The other alternative [18] is to use rhenium, the vertical homologue of technetium in the periodic table, as a yield tracer. Again, a variety of analytical techniques were used and, like ^{90}Sr , included solvent extraction, ion exchange and precipitation. The counting techniques used were either liquid scintillation or gas-filled β -detectors. The data have a slightly low, but insignificant bias.

6.5 ^{238}Pu and ^{239}Pu (Tables 7, 13 and 14)

For these nuclides, 15 sets of data returned. In most cases, ion exchange chromatography was used as a separation and purification technique as well as solvent extraction and precipitation in some cases, followed by electrodeposition onto a stainless steel disc to prepare a source for counting. Plutonium does not have a stable isotope and a yield tracer is required, the possible candidates being ^{236}Pu , ^{242}Pu or ^{244}Pu (really only suitable for mass spectrometry). There was an equal split between the use of ^{236}Pu and ^{242}Pu . The results have a low bias of $\sim 28\%$, but the precision is good. The source of this bias is not clear, but it appears to have arisen in the preparation of the final stock solution.

6.6 ^{241}Am (Tables 7 and 15)

Eighteen participants returned data for this nuclide. It differs from plutonium in that it is possible to measure it by γ -ray spectrometry as well as α -particle spectrometry. If γ -ray spectrometry is employed, there is little in the way of source preparation necessary, but if α -particle spectrometry is used it is necessary to separate it from the plutonium present, especially ^{238}Pu which has the same α -particle energy. The separation techniques used are similar to those for plutonium. The results are spread about the mean, but with a worse precision than those for plutonium, this being more so for data generated by γ -ray spectrometry. This is presumably due to the high degree of attenuation experienced by the 60 keV γ -ray of ^{241}Am .

7 RESULTS FOR β/γ -EMITTERS

7.1 General Comments

As usual, most of the data were reported for the β/γ -emitters, with a total of 41 sets of data being received. All participants used a semiconductor detector, with the increasing popularity of n-type detectors being noted. As one might expect, most of the data handling software is obtained from commercial organisations, although two laboratories used their own in-house software. Without exception, the standards used to calibrate the detectors were of the mixed radionuclide type with one laboratory using ^{152}Eu as an energy calibration source. This represents a considerable improvement on previous years when a number of unsuitable standards have been used.

As regards nuclear data, it was extremely encouraging that many of the participants made use of the recently published IAEA evaluated data file [10] (a copy of which is available free of charge from NPL - contact J L Makepeace). Other data sources used were all well

standard of the nuclide being measured; this term may be combined with the yield in α -spectrometry. The chemical yield is determined by adding a known amount of a chemical yield tracer and measuring its recovery. The yield tracer used should be easily measurable but should not interfere with the measurement of the radionuclide of interest. Reference [16] discusses the rationale underlying the use of chemical yield tracers when measuring α -emitters but the general principles apply to the use of stable or radioactive yield tracers. Tritium, however, is a special case when the sample matrix is aqueous. No carrier is added, since the bulk matrix acts as the yield tracer. Caution is required since the mass difference between normal hydrogen and tritium exacerbates the possibilities for isotope fractionation [17].

As far as nuclear data are concerned, the main area of interest is that of the half-lives, see Table 5.

^3H (Tritium, Tables 8 and 10)

In all cases, distillation was employed to purify the ^3H in the sample, followed by liquid scintillation counting. As noted above there may be problems with isotopic fractionation but only one participant corrected for this, see Table 6

Table 6 - Isotopic ratio (vapour:liquid) of tritiated water [17]

Temperature/°C	$[\text{}^3\text{H}]/[\text{}^1\text{H}]_{\text{vapour}} : [\text{}^3\text{H}]/[\text{}^1\text{H}]_{\text{liquid}}$
0	0.86
0.5	0.88
14	0.90
19.5	0.91
20	0.91
30	0.92
50	0.94
56.5	0.95
70	0.96
90	0.97

There were 15 sets of data returned. The distillation techniques used were either low temperature (ie, evaporation) or high temperature (boiling). There appears to be no difference between the two methods. Overall the results show a bias of $3.8 \pm 6.9\%$. The results are satisfactory and are similar to those of previous exercises.

$^{90}\text{Sr}/^{90}\text{Y}$ (Tables 8 and 11)

There were 12 sets of data returned for this nuclide. Several different techniques were used for analysis, including ion exchange, solvent extraction or precipitation. In most, but not all, cases, strontium was separated chemically and yttrium removed. The ^{90}Y daughter was then allowed to grow in over a measured time period at the end of which the ^{90}Y was again removed from the ^{90}Sr parent and counted. Differing counting techniques were used and these included liquid scintillation counting, Čerenkov counting or the use of a gas-filled β -counter. One participant counted the sample without separation of yttrium by using

documented and up to date. Several of the analysts used the nuclide libraries supplied with their software as their sole data source, although most of them had updated their libraries with the IAEA data.

^{60}Co (Tables 9 and 16)

This is an activation product, widely observed in radioactive waste. The results for this nuclide were comparable to the previous exercises. No significant bias is observed; this is expected since it is a calibration nuclide. Debertin and Schötzig [19] have measured a correction factor for cascade summing of ~2-5% for extended geometries, although it is not known how many participants applied such a correction.

7.3 $^{106}\text{Ru}/^{106}\text{Rh}$ (Tables 9 and 17)

This is a fission product nuclide, with low abundance γ -rays. In addition it may, under some circumstances, be volatile and has the potential to travel long distances from the actual source, as in the Chernobyl accident. The results have no significant bias with a moderate spread on the data. The variation here may be caused by two factors. Cascade summing may reduce the count rates by ~2-5% [19], similar to ^{60}Co . A further source of discrepancy may arise from basing results on the 511.9 keV γ line of ^{106}Rh . This may be enhanced due to the presence of the 511.0 keV annihilation radiation peak; however this should be apparent from the ratios between the 511.9 and 621.8 keV γ -rays.

7.4 ^{134}Cs (Tables 9 and 18)

This is an activation product. It occurs widely in nuclear waste and is distributed throughout various ecosystems. Measurements are complicated by cascade summing. The bias is, again, insignificant and several laboratories did make allowance for, or noted the effect of cascade summing. The results are biased low by about 10%. The correction factor quoted [19] is ~ 6%. If this were applied, most of the results would be statistically indistinguishable from the true value. It is encouraging that some of the participants have applied a correction for cascade summing.

7.5 ^{137}Cs (Tables 9 and 19)

A fission product which is also used as a calibration nuclide. There is no bias on the results for this nuclide, which is to be expected since it is used as a calibration nuclide and it only emits a single γ -ray. The results for this nuclide are indistinguishable from the true value.

7.6 ^{144}Ce (Tables 9 and 20)

This is another fission product. As has been noted in previous exercises it's main γ -ray occurs at 133.5 keV in a part of the spectrum where an accurate energy calibration is essential since the γ -ray efficiency, ϵ_γ is changing rapidly with energy. The results show a positive bias and the spread of results may reflect the difficulties with calibration although the spread is not as large as has been observed on previous occasions.

^{154}Eu (Tables 9 and 21)

This nuclide is formed by neutron activation of stable europium (formed in fission). ^{154}Eu emits several γ -rays, some of which are emitted in cascade. In addition, some of them are coincident with γ -lines from the decay of ^{22}Na , ^{57}Co , ^{95}Zr and ^{207}Bi . However, there are sufficient γ -rays with no interferences to enable permit identification to take place and it is a matter for concern that these nuclides were identified instead of ^{154}Eu in some cases. The results have an insignificant bias, but a large spread. Again, the correction factor [19] due to

cascade summing for the principal γ lines is of the order 4-8% and applying this factor will have the same effect as for ^{106}Ru and ^{134}Cs .

7.8 ^{155}Eu (Tables 9 and 22)

Another fission product, although of lower fission yield than the other fission products in the exercise. In many respects, the problems presented in the measurement of this nuclide are similar to those of ^{144}Ce in that the γ -rays emitted are of low energy and occur in the energy range where the rate of change of ϵ_γ with energy is greatest. As can be seen from the data, the results have a low bias, but a large spread, although this is probably a result of self-absorption, rather than cascade summing.

7.9 Discussion

Overall, the results were good for the β/γ -emitters. The difficulties caused by cascade summing are ongoing and, although correction factors are available [19,20] it should be noted that these are dependent on factors that are individual to the user.

Cascade summing occurs when a radionuclide emits two (or more) γ -rays within the resolving time of the detector [21]. If the second γ -ray is detected as well as the first γ -ray, then the detector will record a γ -ray with the energy of the sum of the two γ -rays, rather than two individual γ -rays at their own energies. This in turn means that the observed count rate for each γ -ray is reduced. It can be shown that the count rate is given by:

$$T_1 = O_1 / (1 - \epsilon_2)$$

and

$$T_2 = O_2 / (1 - \epsilon_1)$$

where T_1 and T_2 are the expected count rates for γ -rays 1 and 2, O_1 and O_2 are the observed count rates for γ -rays 1 and 2 and ϵ_1 and ϵ_2 are the *total* counting efficiencies of γ -rays 1 and 2

The counting efficiencies will depend on the nuclide, detector and source geometry; they are individual to each laboratory. A compromise is necessary in practice since a low activity sample needs to be placed as close to the detector as possible to maximise counting efficiency, which also maximises the probability of summing! The main obstacle to solving the difficulties of cascade summing is that of cost; it requires considerable investment in time and money to determine the relevant correction factors. This may not be justified when the levels of activity and the magnitude of other sources of uncertainty are considered; it is probably only worthwhile noting that the nuclide being measured exhibits the effect. However, when a result is at or near an action or intervention level an estimate of the summing effect may be needed to ensure that the best possible data are presented.

Some participants did not identify ^{154}Eu and ^{144}Ce , reporting ^{22}Na , ^{57}Co , ^{95}Zr and ^{207}Bi instead of, or in addition to, these nuclides. This is a matter for some concern, since it should be clear from the data provided by the analysis system that these results are in error.

8 CONCLUSIONS

The exercise produced data in line with previous exercises and the results for the various nuclides were generally satisfactory. However, and this has been noted in the past, there is some concern that the uncertainties quoted are unrealistically low in some cases. This can disadvantage the analyst in that this narrows the 'band of acceptability' for the data quoted;

it is assumed that this is due to consideration of sample counting data only.

Corrections for cascade summing are now being applied in some cases, and the ongoing improvement in the quality of nuclear decay data used is noted.

There appear to be some problems with the measurement of ^{155}Eu ; these are probably due to the low energy of the γ -rays that it emits and the low probability with which they are emitted.

The identification of spurious nuclides is also of concern and the reason for this is unknown; careful examination of the data generated should eliminate this.

NAMAS are continuing to accredit laboratories for the measurement of radioactivity and see such exercises as an important part of laboratory proficiency testing and quality assurance.

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11 TABLES

Table 7 u-test results for α -emitters

Nuclide	Limits	Number of results		
^{238}Pu	< 1.64	0		
	1.64-1.95	0		
	1.95-2.58	1		
	2.58-3.29	0		
	> 3.29	15		
^{239}Pu	< 1.64	0		
	1.64-1.95	0		
	1.95-2.58	0		
	2.58-3.29	0		
	> 3.29	16		
^{241}Am	< 1.64	Total	α	γ
	1.64-1.95	10	7	3
	1.95-2.58	1	0	1
	2.58-3.29	3	1	2
	> 3.29	1	0	1
		6	0	6

Table 8 u-test for β -emitters

Nuclide	Limits	Number of results		
^3H	< 1.64	11		
	1.64-1.95	1		
	1.95-2.58	2		
	2.58-3.29	1		
	> 3.29	0		
$^{90}\text{Sr}/^{90}\text{Y}$	< 1.64	6		
	1.64-1.95	0		
	1.95-2.58	2		
	2.58-3.29	0		
	> 3.29	4		
^{99}Tc	< 1.64	7		
	1.64-1.95	0		
	1.95-2.58	0		
	2.58-3.29	1		
	> 3.29	0		

Table 9 u-test for β/γ -emitters

Nuclide	Limits	Number of results
^{60}Co	< 1.64	27
	1.64-1.95	0
	1.95-2.58	4
	2.58-3.29	1
	> 3.29	8
$^{106}\text{Ru}/^{106}\text{Rh}$	< 1.64	27
	1.64-1.95	3
	1.95-2.58	3
	2.58-3.29	2
	> 3.29	2
^{134}Cs	< 1.64	21
	1.64-1.95	4
	1.95-2.58	6
	2.58-3.29	4
	> 3.29	6
$^{137}\text{Cs}/^{137\text{m}}\text{Ba}$	< 1.64	34
	1.64-1.95	0
	1.95-2.58	1
	2.58-3.29	1
	> 3.29	5
$^{144}\text{Ce}/^{144}\text{Pr}$	< 1.64	30
	1.64-1.95	1
	1.95-2.58	2
	2.58-3.29	0
	> 3.29	3
^{154}Eu	< 1.64	17
	1.64-1.95	2
	1.95-2.58	6
	2.58-3.29	4
	> 3.29	11
^{155}Eu	< 1.64	10
	1.64-1.95	2
	1.95-2.58	1
	2.58-3.29	1
	> 3.29	22

Table 10 Results for ^3H

Sample	NPL data		Analyst data		Deviation (%)	u-statistic
	^3H (Bq/kg)	$\pm$ (Bq/kg)	^3H (Bq/kg)	$\pm$ (Bq/kg)		
AB001/93	515.7	8.4	507	10	-1.7	0.67
AB002/93	516.2	8.4	492	9	-4.7	1.96
AB003/93	521.7	8.5	548	45	5.0	0.58
AB004/93	520.7	8.5	540	8	3.7	1.66
AB005/93 (a)	527.1	8.6	503	9	-4.5	1.92
AB005/93 (b)	527.1	8.6	514	9	-2.5	1.07
AB005/93 (c)	527.1	8.6	527	9	-0.1	0.03
AB005/93 (d)	527.1	8.6	521	9	-1.2	0.51
AB008/93	506.5	8.3	585	107	15.5	0.73
AB011/93	516.0	8.4	598	30	15.9	2.63
AB012/93	501.0	8.2	537	24	7.1	1.43
AB013/93	507.3	8.3	569	87	12.2	0.71
AB016/93	444.6	7.2	434	25	-2.4	0.41
AB018/93	498.5	8.1	554	50	11.2	1.10
AB021/93	502.5	8.2	518	24	3.0	0.58
				Mean	3.77	
				Std Dev	6.88	

Table 12 Results for ^{99}Tc

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{99}Tc (Bq/kg)	$\pm$ (Bq/kg)	^{99}Tc (Bq/kg)	$\pm$ (Bq/kg)		
AB004/93	52.2	1.7	54.0	2.0	3.5	0.70
AB005/93 (a)	52.8	1.7	51.9	2.1	-1.7	0.34
AB005/93 (b)	52.8	1.7	51.0	2.0	-3.4	0.69
AB005/93 (c)	52.8	1.7	56.6	2.3	7.2	1.33
AB005/93 (d)	52.8	1.7	50.7	2.0	-4.0	0.81
AB005/93 (e)	52.8	1.7	45.7	1.8	-13.5	2.88
AB005/93 (f)	52.8	1.7	50.0	2.0	-5.3	1.07
AB016/93	44.5	1.4	48.7	5.1	9.3	0.79
				Mean	-0.99	
				Std Dev	6.92	

Table 14 Results for ^{239}Pu

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{239}Pu (Bq/kg)	$\pm$ (Bq/kg)	^{239}Pu (Bq/kg)	$\pm$ (Bq/kg)		
AB003/93	26.7	0.5	19.4	1.0	-27.4	6.78
AB004/93	26.6	0.5	19.3	0.4	-27.5	12.1
AB005/93 (a)	27.0	0.5	19.9	0.2	-26.2	13.0
AB005/93 (b)	27.0	0.5	20.1	0.2	-25.3	12.6
AB005/93 (c)	27.0	0.5	20.1	0.2	-25.4	12.7
AB006/93	26.8	0.5	18.8	0.5	-30.0	11.0
AB007/93	26.2	0.5	16.1	0.9	-38.7	9.84
AB009/93	29.1	0.5	20.9	0.6	-28.1	10.1
AB010/93	27.3	0.5	14.9	0.4	-45.6	18.9
AB012/93	25.6	0.5	19.9	1.1	-22.3	4.78
AB014/93	26.2	0.5	18.0	0.5	-31.3	11.9
AB015/93 (a)	25.7	0.5	11.5	0.1	-55.4	29.5
AB015/93 (b)	25.7	0.5	15.8	0.2	-38.7	19.4
AB019/93	26.2	0.5	20.0	1.0	-23.6	5.56
AB023/93	25.5	0.5	18.2	1.6	-28.7	4.39
AB024/94	26.0	0.5	21.9	0.1	-15.8	14.7
				Average	-30.63	
				Std Dev	9.71	

Table 16 Results for ^{60}Co

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{60}Co (Bq/kg)	$\pm$ (Bq/kg)	^{60}Co (Bq/kg)	$\pm$ (Bq/kg)		
BG001/93	92.14	0.92	92.60	3.60	0.50	0.124
BG002/93	93.74	0.94	91.90	4.80	-1.96	0.376
BG003/93	93.28	0.93	94.30	1.20	1.09	0.671
BG005/93	89.30	0.89	88.40	2.70	-1.00	0.315
BG006/93	91.93	0.92	92.10	6.50	0.19	0.026
BG007/93	91.59	0.92	84.27	3.10	-7.99	2.265
BG008/93	97.13	0.97	104.00	0.52	7.08	6.238
BG009/93	79.85	0.80	85.40	1.44	6.95	3.371
BG010/93	185.43	1.85	185.70	3.60	0.15	0.067
BG011/93	91.47	0.92	100.00	3.02	9.33	2.704
BG012/93	93.60	0.94	98.13	0.23	4.84	4.695
BG013/93	89.45	0.90	94.50	2.40	5.65	1.972
BG015/93	87.49	0.88	86.00	2.00	-1.70	0.682
BG016/93	89.22	0.89	86.66	8.67	-2.87	0.294
BG017/93	99.68	1.00	94.60	4.73	-5.10	1.052
BG018/93	93.01	0.93	96.90	2.20	4.18	1.629
BG019/93	93.28	0.93	89.70	1.20	-3.84	2.353
BG020/93	90.81	0.91	49.50	1.24	-45.5	26.88
BG021/93	91.04	0.91	89.70	0.10	-1.47	1.462
BG022/93	88.47	0.89	86.90	1.80	-1.77	0.781
BG023/93	92.24	0.92	90.60	0.63	-1.78	1.468
BG024/93	91.34	0.91	90.40	1.00	-1.03	0.696
BG025/93	90.16	0.90	88.10	1.06	-2.28	1.479
BG026/93	92.74	0.93	92.00	1.80	-0.80	0.366
BG027/93	91.89	0.92	92.00	1.80	0.13	0.057
BG028/93	91.58	0.92	89.84	0.92	-1.90	1.339
BG029/93	91.61	0.92	92.18	2.35	0.62	0.226
BG030/93	94.55	0.95	92.20	1.80	-2.49	1.156

Table 17 Results for ^{106}Ru

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{106}Ru (Bq/kg)	$\pm$ (Bq/kg)	^{106}Ru (Bq/kg)	$\pm$ (Bq/kg)		
BG001/93	49.16	0.49	47.00	3.70	-4.40	0.579
BG002/93	50.02	0.50	49.80	4.10	-0.43	0.052
BG003/93	49.77	0.50	51.00	4.10	2.47	0.298
BG005/93	47.65	0.48	47.50	4.00	-0.31	0.036
BG006/93	49.05	0.49	49.00	4.80	-0.10	0.010
BG009/93	42.61	0.43	47.50	5.34	11.5	0.914
BG010/93	98.94	0.99	85.80	0.10	-13.3	13.21
BG011/93	48.80	0.49	39.70	3.90	-18.6	2.316
BG012/93	49.94	0.50	49.88	0.97	-0.13	0.059
BG013/93	47.73	0.48	57.00	9.80	19.43	0.945
BG015/93	46.68	0.47	38.00	8.00	-18.6	1.083
BG016/93	47.61	0.48	59.54	29.6	25.1	0.403
BG017/93	53.19	0.53	37.90	7.58	-28.7	2.012
BG018/93	49.63	0.50	44.90	3.30	-9.53	1.416
BG018/93	49.63	0.50	47.60	3.30	-4.08	0.607
BG019/93	49.77	0.50	48.00	7.00	-3.56	0.252
BG020/93	48.45	0.49	60.40	4.23	24.7	2.807
BG021/93	48.58	0.49	47.60	0.30	-2.01	1.709
BG022/93	47.20	0.48	45.70	1.70	-3.18	0.852
BG023/93	49.22	0.49	48.00	8.30	-2.47	0.146
BG024/93	48.74	0.49	49.50	1.40	1.56	0.514
BG025/93	48.10	0.48	45.20	4.81	-6.04	0.601
BG026/93	49.48	0.50	48.00	8.30	-9.06	0.812
BG027/93	49.03	0.49	45.00	6.70	-8.21	0.599
BG028/93	48.86	0.49	44.50	3.00	-8.93	1.435
BG029/92	48.88	0.49	44.70	2.27	-8.55	1.800
BG030/93	50.45	0.50	45.40	4.60	-10.0	1.091
BG031/93	47.37	0.47	35.40	4.36	-25.3	2.729
BG032/93	47.63	0.48	46.00	12.5	-3.43	0.131

Table 18 Results for ^{134}Cs

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{134}Cs (Bq/kg)	$\pm$ (Bq/kg)	^{134}Cs (Bq/kg)	$\pm$ (Bq/kg)		
BG001/93	13.97	0.14	13.50	0.70	-3.34	0.654
BG002/93	14.21	0.14	12.70	0.70	-10.6	2.113
BG003/93	14.14	0.14	13.60	0.70	-3.82	0.756
BG005/93	13.54	0.14	11.80	0.60	-12.8	2.823
BG006/93	13.94	0.14	12.20	1.10	-12.5	1.565
BG007/93	13.88	0.14	12.35	1.70	-11.0	0.899
BG009/93	12.10	0.12	10.20	0.74	-15.7	2.539
BG010/93	18.11	0.28	24.80	1.80	-11.8	1.816
BG011/93	13.87	0.14	13.40	0.61	-3.36	0.744
BG012/93	14.19	0.14	13.33	0.13	-6.06	4.465
BG013/93	13.56	0.14	14.60	0.90	7.67	1.143
BG015/93	13.26	0.13	13.00	1.00	-1.98	0.260
BG016/93	13.53	0.14	12.15	1.54	-10.2	0.888
BG017/93	15.11	0.15	12.00	1.20	-20.6	2.572
BG018/93	14.10	0.14	12.90	0.70	-8.51	1.679
BG018/93	14.10	0.14	13.50	0.70	-4.25	0.839
BG019/93	14.14	0.14	13.60	0.90	-3.82	0.592
BG020/93	13.77	0.14	13.60	0.55	-1.20	0.291
BG021/93	13.80	0.14	14.20	0.10	-2.90	2.345
BG022/92	13.41	0.13	12.10	0.30	-9.77	3.987
BG023/93	13.98	0.14	13.50	0.44	-3.45	1.045
BG024/93	13.85	0.14	13.60	0.30	-1.78	0.746
BG025/93	13.67	0.14	15.50	0.62	13.4	2.887
BG026/93	14.06	0.14	13.00	0.60	-7.53	1.717
BG027/93	13.93	0.14	13.00	0.60	-6.67	1.508
BG028/93	13.88	0.14	12.67	0.38	-8.73	2.996
BG029/93	13.89	0.14	14.47	0.38	4.20	1.442
BG030/93	14.33	0.14	11.90	0.50	-17.0	4.677
BG031/93	13.46	0.14	12.20	0.52	-9.34	2.340

Table 19 Results for ^{137}Cs

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{137}Cs (Bq/kg)	$\pm$ (Bq/kg)	^{137}Cs (Bq/kg)	$\pm$ (Bq/kg)		
BG001/93	22.55	0.23	21.70	1.00	-3.75	0.826
BG002/93	22.94	0.23	22.70	1.20	-1.04	0.195
BG003/93	22.83	0.23	22.00	0.60	-3.62	1.286
BG005/93	21.85	0.22	21.70	0.90	-0.69	0.163
BG006/93	22.50	0.23	23.00	1.70	2.24	0.294
BG007/93	22.41	0.22	23.03	2.10	2.76	0.292
BG008/93	23.77	0.24	28.00	0.50	17.8	7.596
BG009/93	19.54	0.20	17.50	0.65	-10.4	3.004
BG010/93	45.38	0.45	45.70	1.10	0.72	0.273
BG011/93	22.38	0.22	23.30	0.61	4.10	1.413
BG012/93	22.91	0.23	23.30	0.13	1.72	1.500
BG013/93	21.89	0.22	24.50	1.20	11.9	2.141
BG015/93	21.41	0.21	22.00	1.00	2.76	0.578
BG016/93	21.83	0.22	20.37	2.04	-6.70	0.714
BG017/93	24.39	0.24	23.10	1.85	-5.30	0.694
BG018/93	22.76	0.23	23.30	0.90	2.37	0.582
BG019/93	22.83	0.23	24.00	0.80	5.15	1.412
BG020/93	22.22	0.22	26.70	1.21	20.2	3.641
BG021/93	22.28	0.22	22.10	0.10	-0.80	0.727
BG022/93	21.65	0.22	21.70	0.50	0.24	0.096
BG023/93	22.57	0.23	20.70	0.34	-8.29	4.585
BG024/93	22.35	0.22	22.30	0.50	-0.23	0.095
BG025/93	22.06	0.22	21.60	0.65	-2.09	0.673
BG026/93	22.69	0.23	23.00	0.60	1.35	0.478
BG027/93	22.48	0.23	23.00	0.60	2.29	0.805
BG028/93	22.41	0.22	22.46	0.56	0.23	0.084
BG029/93	22.42	0.22	23.34	0.65	4.12	1.343
BG030/93	23.14	0.23	22.50	1.00	-2.75	0.620
BG031/93	21.72	0.22	21.50	0.85	-1.03	0.255

Table 20 Results for ^{144}Ce

Sample	NPL Data		Analyst		Deviation (%)	u-statistic
	^{144}Ce (Bq/kg)	$\pm$ (Bq/kg)	^{144}Ce (Bq/kg)	$\pm$ (Bq/kg)		
BG001/93	45.04	1.53	46.10	3.40	2.36	0.285
BG002/93	45.82	1.56	47.80	4.70	4.32	0.400
BG003/93	45.60	1.55	47.70	2.10	4.61	0.806
BG005/93	43.65	1.48	36.10	1.80	-17.3	3.236
BG006/93	44.94	1.53	45.00	3.80	0.14	0.016
BG007/93	44.77	1.52	41.73	9.30	-6.79	0.323
BG009/93	39.03	1.33	40.90	3.05	4.79	0.562
BG010/93	90.64	3.08	97.40	2.60	7.46	1.677
BG011/93	44.71	1.52	46.40	1.85	3.78	0.706
BG012/93	45.76	1.56	49.20	0.47	7.53	2.120
BG013/93	43.72	1.49	47.40	3.30	8.41	1.016
BG016/93	43.61	1.48	49.24	12.7	12.9	0.440
BG017/93	48.73	1.66	53.80	5.92	10.4	0.826
BG018/93	45.46	1.55	44.60	2.00	-1.90	0.342
BG019/93	45.60	1.55	45.60	3.00	0.01	0.002
BG020/93	44.39	1.51	45.50	1.14	2.51	0.589
BG021/93	44.50	1.51	60.00	0.20	34.8	10.15
BG022/93	43.24	1.47	42.30	1.20	-2.18	0.497
BG023/93	45.09	1.53	47.00	0.93	4.24	1.066
BG024/93	44.65	1.52	46.90	1.30	5.04	1.126
BG025/93	44.07	1.50	45.10	3.52	2.34	0.269
BG026/93	45.33	1.54	50.00	3.00	10.3	0.384
BG027/93	44.92	1.53	50.00	3.60	11.3	1.300
BG028/93	44.76	1.52	43.00	1.80	-3.94	0.749
BG029/93	44.78	1.52	44.42	1.19	-0.80	0.186
BG030/93	46.22	1.57	48.10	4.00	4.07	0.438
BG031/93	43.39	1.48	39.80	3.09	-8.28	1.050
BG032/93	43.64	1.48	49.00	6.90	12.3	0.760

Table 21 Results for ^{154}Eu

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{154}Eu (Bq/kg)	$\pm$ (Bq/kg)	^{154}Eu (Bq/kg)	$\pm$ (Bq/kg)		
BG001/93	28.39	0.28	27.50	1.00	-3.14	0.856
BG002/93	28.88	0.29	25.10	2.40	-13.1	1.565
BG003/93	28.74	0.29	27.70	1.60	-3.62	0.641
BG005/93	27.51	0.28	24.20	1.00	-12.0	3.196
BG006/93	28.33	0.28	27.70	3.30	-2.21	0.189
BG007/93	28.22	0.28	21.37	2.20	-24.3	3.089
BG009/93	24.60	0.25	21.30	1.66	-13.4	1.968
BG010/93	57.14	0.57	53.10	0.90	-7.06	3.785
BG011/93	28.18	0.28	25.40	0.49	-9.88	4.923
BG012/93	28.84	0.29	28.54	0.28	-1.05	0.750
BG013/93	27.56	0.28	28.00	1.10	1.59	0.387
BG015/93	26.96	0.27	28.00	2.00	3.87	0.517
BG016/93	27.49	0.28	26.19	3.56	-4.73	0.364
BG017/93	30.72	0.31	26.60	2.13	-13.4	1.914
BG018/93	28.66	0.29	26.40	2.30	-7.88	0.974
BG018/93	28.66	0.29	27.80	2.30	-3.00	0.370
BG019/93	28.74	0.29	30.90	1.90	7.51	1.124
BG020/93	27.98	0.28	26.00	2.60	-7.07	0.757
BG021/93	28.05	0.28	29.10	0.40	3.74	2.147
BG022/93	27.26	0.27	26.10	0.60	-4.25	1.758
BG023/93	28.42	0.28	25.20	0.69	-11.3	4.317
BG024/93	28.15	0.28	26.40	0.50	-6.20	3.041
BG025/93	27.78	0.28	27.40	1.43	-1.37	0.261
BG026/93	28.58	0.29	27.00	0.60	-5.51	2.370
BG027/93	28.31	0.28	27.00	0.60	-4.63	1.978
BG028/93	28.22	0.28	24.50	0.56	-13.2	5.928
BG029/93	28.23	0.28	22.68	0.71	-19.7	7.260
BG030/93	29.13	0.29	25.20	0.90	-13.5	4.158

Table 22 Results for ^{155}Eu

Sample	NPL	Data	Analyst	Data	Deviation (%)	u-statistic
	^{155}Eu	$\pm$	^{155}Eu	$\pm$		
BG001/93	49.63	0.50	43.80	1.70	-11.7	3.291
BG002/93	50.49	0.51	42.50	2.60	-15.8	3.016
BG003/93	50.24	0.50	43.40	1.40	-13.6	4.600
BG005/93	48.10	0.48	65.20	2.60	35.6	6.468
BG007/93	49.33	0.49	62.45	9.90	26.6	1.323
BG008/93	52.32	0.52	319.00	10.5	510	25.30
BG009/93	43.01	0.43	34.20	1.42	-20.4	5.937
BG010/93	99.88	1.00	84.10	2.10	-15.8	6.785
BG011/93	49.27	0.49	44.00	1.13	-10.7	4.272
BG012/93	50.42	0.50	45.20	0.25	-10.3	9.271
BG013/93	48.18	0.48	46.60	1.90	-3.28	0.806
BG015/93	47.12	0.47	59.00	3.00	25.2	3.911
BG016/93	48.06	0.48	42.34	4.23	-11.9	1.342
BG017/93	53.69	0.54	51.90	4.15	-3.34	0.428
BG018/93	50.10	0.50	46.70	2.30	-6.78	1.443
BG019/93	50.24	0.50	47.20	1.70	-6.05	1.716
BG020/93	48.91	0.49	38.40	0.96	-21.5	9.755
BG021/93	49.04	0.49	37.80	0.50	-22.9	16.04
BG022/93	47.65	0.48	41.30	1.70	-13.3	3.597
BG023/93	49.68	0.50	44.20	0.75	-11.0	6.094
BG024/93	49.20	0.49	40.30	0.70	-18.1	10.40
BG025/93	48.56	0.49	41.50	3.95	-14.5	1.774
BG026/93	49.95	0.50	45.00	1.20	-9.91	3.810
BG027/93	49.49	0.50	45.00	1.20	-9.08	3.460
BG028/93	49.33	0.49	44.40	1.10	-9.99	4.086
BG029/93	49.34	0.49	41.88	1.31	-15.1	5.331
BG030/93	50.93	0.51	39.60	2.30	-22.2	4.808
BG031/93	47.82	0.48	45.90	1.91	-4.01	0.973

Table 11 Results for ^{90}Sr

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	$^{90}\text{Sr}/^{90}\text{Y}$ (Bq/kg)	$\pm$ (Bq/kg)	$^{90}\text{Sr}/^{90}\text{Y}$ (Bq/kg)	$\pm$ (Bq/kg)		
AB001/93	18.9	0.1	22.0	2.5	16.1	1.22
AB003/93	19.2	0.1	30.4	4.6	58.9	2.45
AB005/93 (a)	19.4	0.1	18.0	0.4	-6.8	3.68
AB005/93 (b)	19.4	0.1	18.2	0.3	-6.2	3.41
AB005/93 (c)	19.4	0.1	18.2	0.3	-6.0	3.30
AB009/93	20.9	0.1	20.0	3.0	-4.2	0.29
AB010/93	19.6	0.1	17.5	0.4	-10.9	5.10
AB011/93	19.0	0.1	16.9	3.0	-10.8	0.68
AB012/93	18.4	0.1	25.3	4.4	37.5	1.57
AB016/93	16.3	0.1	15.8	3.0	-3.2	0.18
AB019/93	18.8	0.1	21.0	1.0	11.7	2.19
AB023/93	18.3	0.1	17.3	0.7	-5.6	1.47
				Mean	5.87	
				Std Dev	20.95	

Table 18 Results for ^{134}Cs (cont)

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{134}Cs (Bq/kg)	$\pm$ (Bq/kg)	^{134}Cs (Bq/kg)	$\pm$ (Bq/kg)		
BG032/93	13.53	0.14	25.00	3.00	84.7	3.818
BG032/93	13.53	0.14	15.00	0.80	10.8	1.808
BG033/93	14.02	0.14	11.90	0.42	-15.1	4.769
BG034/93	13.82	0.14	14.52	0.24	5.04	2.518
BG035/93	13.64	0.14	11.30	0.70	-17.1	3.276
BG036/93	13.62	0.14	12.33	0.17	-9.48	5.930
BG037/93	13.55	0.14	19.20	5.00	41.7	1.130
BG040/93	13.85	0.14	14.10	1.70	1.80	0.146
BG041/93	13.72	0.14	13.01	0.54	-5.16	1.269
BG042/93	13.73	0.14	12.80	1.00	-6.77	0.920
BG044/93	13.88	0.14	13.60	1.20	-2.05	0.235
BG045/93	13.74	0.14	14.00	1.00	1.89	0.258
				Average	-2.128	
				Std Dev	17.216	

Table 13 Results for ^{238}Pu

Sample	NPL Data	Analyst Data	Deviation (%)	u-statistic		
	²³⁸ Pu (Bq/kg)	± (Bq/kg)	²³⁸ Pu (Bq/kg)	± (Bq/kg)		
AB003/93	5.22	0.03	3.87	0.21	-25.9	6.37
AB004/93	5.21	0.03	3.80	0.11	-27.1	12.3
AB005/93 (a)	5.28	0.03	4.00	0.06	-24.2	18.7
AB005/93 (b)	5.28	0.03	4.02	0.06	-23.8	18.4
AB005/93 (c)	5.28	0.03	3.93	0.06	-25.5	19.7
AB006/93	5.24	0.03	3.87	0.13	-26.2	10.3
AB007/93	5.14	0.03	3.33	0.28	-35.2	6.41
AB009/93	5.69	0.04	4.90	0.40	-13.9	1.97
AB010/93	5.34	0.03	3.02	0.13	-43.5	17.3
AB012/93	5.02	0.03	4.00	0.10	-20.3	9.71
AB014/93	5.13	0.03	3.60	0.12	-29.8	12.3
AB015/93 (a)	5.04	0.03	2.35	0.03	-53.3	62.0
AB015/93 (b)	5.04	0.03	3.38	0.09	-32.9	17.4
AB019/93	5.13	0.03	4.00	0.20	-22.0	5.56
AB023/93	5.00	0.03	3.60	0.30	-28.0	4.63
AB024/93	5.14	0.03	4.32	0.04	-16.0	12.6
				Average	-27.98	
				Std Dev	9.81	

Table 19 Results for ^{137}Cs (cont)

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{137}Cs (Bq/kg)	$\pm$ (Bq/kg)	^{137}Cs (Bq/kg)	$\pm$ (Bq/kg)		
BG032/93	21.85	0.22	24.00	3.40	9.86	0.632
BG032/93	21.85	0.22	21.00	2.10	-3.87	0.401
BG033/93	22.63	0.23	22.20	0.79	-1.89	0.519
BG034/93	22.31	0.22	26.20	0.41	17.4	8.326
BG035/93	22.01	0.22	20.10	1.20	-8.69	1.568
BG036/93	22.00	0.22	23.23	0.27	5.64	3.564
BG037/93	21.87	0.22	28.00	5.00	28.0	1.225
BG040/93	22.36	0.22	27.10	2.90	21.2	1.630
BG041/93	22.14	0.22	23.40	0.93	5.68	1.316
BG042/93	22.16	0.22	21.10	2.00	-4.79	0.528
BG044/93	22.41	0.22	23.20	0.80	3.51	0.948
BG045/93	22.18	0.22	23.00	1.00	3.70	0.801
				Average	2.659	
				Std Dev	8.277	

Table 15 Results for ^{241}Am

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{241}Am (Bq/kg)	$\pm$ (Bq/kg)	^{241}Am (Bq/kg)	$\pm$ (Bq/kg)		
AB001/93 (γ)	13.1	0.05	12.7	0.8	-3.05	0.50
AB003/93	13.3	0.05	13.1	0.3	-1.69	0.66
AB004/93 (γ)	13.3	0.05	10.4	1.0	-21.7	2.90
AB005/93 (a)	13.4	0.05	13.7	0.2	1.83	1.46
AB005/93 (b)	13.4	0.05	13.4	0.2	-0.03	0.0
AB005/93 (c)	13.4	0.05	13.4	0.2	-0.03	0.0
AB006/93 (γ)	13.4	0.05	15.2	0.8	13.8	2.23
AB007/93	13.1	0.05	10.6	0.5	-18.8	4.98
AB009/93	14.5	0.05	13.7	1.0	-5.46	0.80
AB010/93	13.6	0.05	11.3	0.3	-17.3	7.56
AB012/93	12.8	0.05	11.2	0.7	-12.4	2.28
AB014/93	13.1	0.05	12.0	1.3	-8.14	0.85
AB014/93 (γ)	13.1	0.05	14.1	1.0	7.93	1.00
AB015/93 (a)	12.8	0.05	19.5	0.1	52.2	59.9
AB015/93 (b)	12.8	0.05	17.2	0.1	34.3	39.4
AB015/93 (c) (γ)	12.8	0.05	11.4	0.7	-10.9	1.99
AB016/93 (γ)	11.3	0.05	10.0	0.6	-11.8	2.16
AB017/93 (γ)	12.5	0.05	13.0	0.6	4.18	0.83
AB019/93	13.1	0.05	10.2	0.4	-21.9	7.19
AB023/93	12.7	0.05	12.4	1.1	-2.59	0.27
AB024/93	12.9	0.05	12.2	0.1	-5.15	6.26
				Average	-1.27 $\pm$ 17.75	

Table 20 Results for ^{144}Ce (cont)

Sample	NPL Data		Analyst		Deviation (%)	u-statistic
	^{144}Ce (Bq/kg)	$\pm$ (Bq/kg)	^{144}Ce (Bq/kg)	$\pm$ (Bq/kg)		
BG033/93	47.90	1.54	47.90	2.39	5.97	0.950
BG034/93	44.57	1.51	51.28	1.29	15.0	3.370
BG035/93	43.97	1.50	42.40	2.50	-3.57	0.540
BG036/93	43.93	1.49	47.94	0.93	9.14	2.282
BG040/93	44.66	1.52	50.20	13.0	12.4	0.423
BG041/93	44.23	0.50	45.92	1.77	3.82	0.727
BG042/93	44.27	1.51	46.10	4.60	4.13	0.378
BG044/93	44.77	1.52	48.40	2.60	8.11	1.204
				Average	4.673	
				Std Dev	8.186	

Table 16 Results for ^{60}Co (cont)

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{60}Co (Bq/kg)	$\pm$ (Bq/kg)	^{60}Co (Bq/kg)	$\pm$ (Bq/kg)		
BG031/93	88.78	0.89	84.40	1.85	-4.93	2.132
BG032/93	89.28	0.89	95.00	4.50	6.41	1.248
BG032/93	89.28	0.89	93.00	3.30	4.17	1.090
BG033/93	92.47	0.93	90.30	1.28	-2.35	1.373
BG034/93	91.19	0.91	103.43	0.49	13.4	11.83
BG035/93	89.96	0.90	82.80	1.50	-7.96	4.091
BG036/93	89.86	0.90	93.85	0.80	4.44	3.316
BG037/93	89.38	0.90	97.00	7.00	8.83	1.080
BG041/93	90.49	0.91	95.30	0.91	5.62	3.738
BG042/93	90.57	0.91	84.80	5.00	-6.37	1.135
BG044/93	91.59	0.92	91.30	2.50	-0.32	0.110
BG045/93	90.64	0.91	92.00	5.00	1.50	0.268
				Average	-0.522	
				Std Dev	8.602	

Table 21 Results for ^{154}Eu (cont)

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{154}Eu (Bq/kg)	$\pm$ (Bq/kg)	^{154}Eu (Bq/kg)	$\pm$ (Bq/kg)		
BG031/93	27.35	0.27	24.90	1.01	-8.97	2.345
BG032/93	27.51	0.28	25.00	1.90	-9.12	1.306
BG032/93	27.51	0.28	24.00	1.40	-12.8	2.458
BG033/93	28.49	0.29	25.60	0.71	-10.2	3.785
BG035/93	27.72	0.28	22.90	1.20	-17.4	3.912
BG036/93	27.69	0.28	25.67	0.38	-7.29	4.292
BG037/93	27.54	0.28	82.00	7.00	197.7	7.774
BG040/93	28.15	0.28	26.40	3.00	-6.23	0.582
BG041/93	27.88	0.28	25.22	0.75	-9.55	3.331
BG042/93	27.91	0.28	25.10	2.00	-10.1	1.390
BG044/93	28.22	0.28	25.90	0.80	-8.23	2.737
BG045/93	27.93	0.28	28.00	1.00	0.26	0.069
				Average	-2.262	
				Std Dev	32.649	

Table 17 Results for ^{106}Ru (cont)

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{106}Ru (Bq/kg)	$\pm$ (Bq/kg)	^{106}Ru (Bq/kg)	$\pm$ (Bq/kg)		
BG033/93	49.34	0.49	45.40	4.15	-7.98	0.942
BG034/93	48.65	0.49	57.05	3.53	17.3	2.356
BG035/93	48.00	0.48	44.30	2.70	-7.70	1.348
BG036/93	47.95	0.48	44.49	1.81	-7.21	1.846
BG037/93	47.69	0.48	84.00	7.00	76.1	5.175
BG041/93	48.28	0.48	45.01	2.70	-6.78	1.193
BG042/93	48.32	0.48	43.80	5.00	-9.36	0.901
BG044/93	48.87	0.49	46.60	7.50	-4.65	0.302
				Average	-1.416	
				Std Dev	17.052	

Table 22 Results for ^{155}Eu (cont)

Sample	NPL Data		Analyst Data		Deviation (%)	u-statistic
	^{155}Eu	$\pm$	^{155}Eu	$\pm$		
BG032/93	48.09	0.48	47.00	3.00	-2.26	0.357
BG033/93	49.81	0.50	42.90	0.89	-13.9	6.784
BG034/93	49.15	0.49	49.64	0.41	1.07	0.820
BG035/93	48.45	0.49	18.10	1.30	-62.6	21.88
BG036/93	48.40	0.48	32.96	0.48	-31.9	22.65
BG040/93	49.21	0.49	53.50	4.10	8.71	1.038
BG044/93	49.33	0.49	48.80	3.70	-1.08	0.143
BG045/93	48.82	0.49	42.00	3.00	-14.0	2.244
				Average	4.455	
				Std Dev	85.727	

Table 23 Cascade summing corrections for nuclides in aqueous solution in a 1 dm³ container placed on top of a Ge(Li) detector [19]

Nuclide	E _{γ} (keV)	Summing correction
²² Na	1274	+7.7%
⁶⁰ Co	1173	+3.3%
	1332	+3.4%
⁸⁸ Y	898	+2.9%
	1836	+3.5%
¹⁰⁶ Ru/ ¹⁰⁶ Rh	512	+2.5%
	622	+4.4%
¹²⁵ Sb	176	+0.6%
	428	+0.2%
	601	+0.1%
	636	+0.1%
¹³³ Ba	81	+5.4%
	276	+2.8%
	303	+1.5%
	356	+1.2%
	384	-1.1%
¹³⁴ Cs	605	+5.0%
	796	+4.9%
¹³⁹ Ce	166	+0.3%
¹⁵² Eu	122	+5.3%
	245	+7.2%
	344	+3.1%
	411	+7.1%
	444	+6.3%
	779	+4.2%
	964	+3.4%
	1086	-0.6%
	1112	+2.5%
	1408	+2.7%
¹⁵⁴ Eu	123	+4.8%
	248	+7.6%
	592	+7.4%
	723	+5.2%
	873	+6.1%
	996	+2.3%
	1005	+2.9%
	1275	+2.4%

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