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## **Environmental Radioactivity Intercomparison 1992**

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### **ABSTRACT**

An account is given of the third in the series of environmental radioactivity measurement intercomparison exercises carried out by NPL. The results for this exercise are reported. In general, the results are satisfactory and the level of performance is comparable to previous years.

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## 1 INTRODUCTION

Following on from two previous environmental radioactivity measurement intercomparison exercises [1,2,3,4], NPL conducted a third intercomparison of this sort in 1992. The results from the first two exercises were generally satisfactory, although the problem of cascade summing appears to be perennial. At the follow up meetings to these events [3], the participants had made it clear that further exercises would be of interest to them. This time, particular interest was shown in  $\alpha$ -emitting radionuclides, an area of great environmental concern. Therefore the exercise was organised on the same grounds as the previous one, ie two sets of samples were prepared, one containing  $\alpha$ -emitters and one containing  $\beta$ - and  $\beta/\gamma$ -emitters.

## 2 ORGANISATION

### 2.1 Participants

The potential participants were drawn from our Divisional database, with approximately 130 contacts. Of these, 39 took the sample containing  $\beta$ - and  $\beta/\gamma$ -emitters and 17 took the sample containing the  $\alpha$ -emitters. As before, the participants fell into three groups: (i) the nuclear industry and the statutory monitoring organisations, (ii) organisations with a secondary interest in the nuclear industry (such as water companies) and (iii) contract laboratories. The split was fairly equal for the  $\beta/\gamma$  analyses, whereas the  $\alpha$ -emitters and  $\beta$ -emitters tended to be measured by laboratories from group (i). The actual participants are given in the Appendix.

### 2.2 Samples

As noted above, two sets of samples were prepared; one containing  $\alpha$ -emitters and one containing  $\beta$ - and  $\beta/\gamma$ -emitters. All of the chosen nuclides are released (in varying amounts) to the environment from the various nuclear sites in the UK. The individual nuclides and their approximate concentrations are given in Table 1: the participants were told the acidity of the solutions and which  $\alpha$ - and  $\beta$ - emitting nuclides were included in the samples and given a list of potential  $\beta/\gamma$  emitters.

Table 1      Samples

| Sample         | Nuclide           | Emission        | Level (Bq/kg) |
|----------------|-------------------|-----------------|---------------|
| $\alpha$       | $^{239}\text{Pu}$ | $\alpha$        | $\sim 15$     |
| $\alpha$       | $^{241}\text{Am}$ | $\alpha/\gamma$ | $\sim 9$      |
| $\beta/\gamma$ | $^3\text{H}$      | $\beta$         | $\sim 600$    |
| $\beta/\gamma$ | $^{90}\text{Sr}$  | $\beta$         | $\sim 40$     |
| $\beta/\gamma$ | $^{99}\text{Tc}$  | $\beta$         | $\sim 1$      |
| $\beta/\gamma$ | $^{60}\text{Co}$  | $\beta/\gamma$  | $\sim 40$     |
| $\beta/\gamma$ | $^{95}\text{Zr}$  | $\beta/\gamma$  | $\sim 250$    |
| $\beta/\gamma$ | $^{95}\text{Nb}$  | $\beta/\gamma$  | Ingrowing     |
| $\beta/\gamma$ | $^{134}\text{Cs}$ | $\beta/\gamma$  | $\sim 300$    |
| $\beta/\gamma$ | $^{137}\text{Cs}$ | $\beta/\gamma$  | $\sim 100$    |
| $\beta/\gamma$ | $^{144}\text{Ce}$ | $\beta/\gamma$  | $\sim 60$     |
| $\beta/\gamma$ | $^{154}\text{Eu}$ | $\beta/\gamma$  | $\sim 120$    |

### 3      SAMPLE PREPARATION AND DISTRIBUTION

The samples were produced from a number of single nuclide standards, the majority of which were produced at NPL. The exceptions were  $^3\text{H}$ , which was obtained from Amersham International plc, and  $^{239}\text{Pu}$  that was supplied by AEA Technology's Actinide Chemistry and Analysis Department. The gamma emitting nuclides were all measured in high pressure ionisation chambers that are calibrated against primary national radioactivity standards. The  $^{90}\text{Sr}/^{90}\text{Y}$  and  $^{99}\text{Tc}$  had been previously standardised at NPL and the  $^3\text{H}$  and  $^{239}\text{Pu}$  activities were derived from the manufacturers certificates, the activities being checked by liquid scintillation counting. The purities of the solutions were also checked by gamma spectrometry, this being especially important for  $^{154}\text{Eu}$  which had been produced by neutron irradiation of  $^{153}\text{Eu}$  separated in the NPL isotope separator to reduce the  $^{151}\text{Eu}$  content to a minimum; this material had then been activated at the Imperial College reactor at Ascot; inevitably, it contained some  $^{152}\text{Eu}$  from the residual  $^{151}\text{Eu}$  and also from the  $^{153}\text{Eu}(n,2n)^{152}\text{Eu}$  reaction [5]. The  $^{154}\text{Eu}$  contained  $\sim 1.1\%$   $^{155}\text{Eu}$  and  $\sim 0.4\%$   $^{152}\text{Eu}$ . The  $^{239}\text{Pu}$  solution also contained other isotopes of plutonium and the activity ratios are given in Table 2.

Table 2  $^{239}\text{Pu}$  impurities

| Nuclide           | Activity/ $^{239}\text{Pu}$ activity |
|-------------------|--------------------------------------|
| $^{238}\text{Pu}$ | $1 \times 10^{-3}$                   |
| $^{239}\text{Pu}$ | 1                                    |
| $^{240}\text{Pu}$ | $5.3 \times 10^{-3}$                 |
| $^{241}\text{Pu}$ | $8.4 \times 10^{-2}$                 |

One of the participants was, indeed, able to detect  $^{241}\text{Pu}$  in their sample. The other materials were free from measurable contamination.

Accurately weighed aliquots of the single radionuclide solutions were dispensed into two double sealed polythene bottles and made up to (i) ~ a 100 g solution of  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  and (ii) a ~ 100 g solution of  $^3\text{H}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}/^{90}\text{Y}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{99}\text{Tc}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{144}\text{Ce}$  and  $^{154}\text{Eu}$ . These solutions were 0.1 M with respect to hydrochloric and 0.01 M with respect to hydrofluoric acid. Participants samples were prepared from these solutions by diluting ~ 0.5 g to 500 g with an inactive solution of 0.1M HCl/0.01 M HF.

The samples were prepared and distributed on demand to the participants, along with the relevant data collection paperwork. As in previous exercises, the results reported here do not allow the reported data to be identified with individual participants.

#### 4 TREATMENT OF DATA

In this report, uncertainties are quoted at the 1- $\sigma$  (68%) confidence level. As before [1,2], the data received were compared with the NPL data by means of u-test [6] where

$$u = \frac{[(\text{Analyst result} - \text{NPL result})]}{[(\text{Uncertainty on analyst result})^2 + (\text{Uncertainty on NPL result})^2]^{1/2}}$$

and is compared against values in the t statistic tables with infinite degrees of freedom. The percentage deviation which is defined as;

$$\left( \frac{\text{Analyst result} - \text{NPL result}}{\text{NPL result}} \right) \times 100\%$$

A t-test [7] was also applied where necessary. The value of t is calculated in the same way as u, but is compared with values in the t statistic tables with the relevant numbers of degrees of freedom, where appropriate. In the cases where the reported

data were grossly at variance with the NPL data, the information was checked with the analyst (except in the case of  $^{95}\text{Nb}$ , see Section 7.3).

For the u-test, the results are assessed in Table 3:

Table 3 u-test limits

| Condition         | Comment                                                                                                                  |
|-------------------|--------------------------------------------------------------------------------------------------------------------------|
| $1.64 > u$        | The reported value does not differ from the actual value.                                                                |
| $1.64 < u < 1.95$ | The reported value probably does not differ from the actual value, but more data is required to support this conclusion. |
| $1.95 < u < 2.58$ | It is not clear whether the reported values differ from the actual value or not. More evidence is required.              |
| $2.58 < u < 3.29$ | The reported value probably differs from the actual value, but more data is required to support this conclusion.         |
| $3.29 < u$        | The repeated value differs from the actual value.                                                                        |

## 5 RESULTS FOR $\alpha$ -EMITTERS

In total, 18 samples were sent out and 12 sets of data were received. In most cases, a straightforward sample analysis was followed, the sample being a fairly simple one. Few problems seem to have been encountered by the analysts and most results were acceptable, see Tables 7 and 8.

Three methods of chemical yield determination are generally available. One is to assume a chemical yield. This is problematical since the chemical yield is definitely method dependent, certainly sample dependent and probably operator dependent. The reader is referred to a paper on plutonium analysis [8] where the chemical yield of  $^{239}\text{Pu}$  is reported as 94% and that of  $^{241}\text{Pu}$  as 97% for the same technique. The second approach is to use a stable element as a tracer for the radionuclide in question. This can lead to problems where the tracer and the radionuclide are different elements since they will, at some point, speciate and thus, ideally, the tracer and radionuclide should be isotopes of the same element. Since neither of the elements concerned in this analysis exists in a non-radioactive form, this approach has severe limitations. However, neither of the above techniques should be written off, especially when a screening measurement is required. The third method, that employed by most of the participants, is to use a radioactive tracer; this gives the most reliable results. For a fuller discussion, the reader is referred to Harvey and Lovett's review of this subject [9]. The tracer ideally needs to be the same element, in the same chemical form and preferably measured by the same counting technique as the analyte.

For plutonium, the preferred tracers are  $^{242}\text{Pu}$  (difficult to obtain in a sufficiently pure state),  $^{244}\text{Pu}$  (virtually impossible to obtain in a pure state) or  $^{236}\text{Pu}$  (cyclotron produced and hence easily purified, since the target is a different element). The difficulty with  $^{236}\text{Pu}$  is its short half-life. This is not necessarily a problem, but if uranium analysis is also being performed, the daughter  $^{232}\text{U}$  needs to be removed, prior to use. The preferred tracer is therefore  $^{242}\text{Pu}$ .

For  $^{241}\text{Am}$ , the only tracer that fits the above criteria is  $^{243}\text{Am}$ . None of the other nuclides of americium are suitable, due to their short half lives, or difficult means of production.

In general, nuclear decay data cause no problems for the nuclides used here when making measurements by alpha counting, since the decay of  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  are by 100% alpha emission with some  $\gamma$  and x-ray emission.

### 5.1 $^{239}\text{Pu}$

In all cases, ion exchange chromatography was used to isolate the plutonium, sometimes following a preconcentration step, such as evaporation or precipitation. The separation of plutonium from americium in most cases depended on the fact that americium remains in the 3+ oxidation state under most normal conditions, whereas plutonium, similar to uranium and neptunium, has the 3+, 4+, 5+ and 6+ oxidation states available. In the higher oxidation states plutonium forms stable anionic complexes with both nitrate and chloride ions in strong acid solutions. These can then be absorbed by suitable anion exchange resins; americium is usually only absorbed by cationic resins, unless thiocyanate is used [11]. The procedure is one of oxidation, adsorption and then desorption in weak acid. This process may be repeated to obtain sufficient source purity. Electroplating is usually employed to prepare a source for counting; there are a number of variations. One participant used the simpler technique, of evaporating the separated plutonium onto a planchet. This technique, whilst effective for simple screening measurements, does not usually give sources suitable for  $\alpha$ -spectrometry. Even so, the participant was at a loss to explain the variation in results for this particular sample.

Only two tracers were employed;  $^{242}\text{Pu}$  (9 analysts) and  $^{236}\text{Pu}$  (3 analysts). The participant who produced an evaporated source used no tracer but counted against a  $^{239}\text{Pu}$  standard. There appears to be no identifiable bias between techniques. Overall, there appears to be a high bias, but there does not appear to be a simple explanation for this on the evidence presented.

### 5.2 $^{241}\text{Am}$

There were two methods employed to measure this nuclide. Two analysts made a direct measurement, by assaying the 59.5 keV photon emission by gamma spectrometry. The rest used chemical separation, followed by alpha spectrometry, to assay the americium. In two cases, where the latter technique was used, the

residue from the plutonium analysis was reduced in volume and then plated, for the rest, a more rigorous approach was developed, involving additional cation and anion exchange steps. It is interesting to note that there appears to be a significant difference between the means of the alpha spectrometry and gamma spectrometry results, as shown in Table 4.

Table 4 Comparison of  $\alpha$ - and  $\gamma$  spectrometry for  $^{241}\text{Am}$  data

| Technique              | $\bar{x}$ | $\delta x$ | Degrees of freedom |
|------------------------|-----------|------------|--------------------|
| Overall                | 1.00515   | 0.09651    | 10                 |
| $\alpha$ -spectrometry | 0.97109   | 0.05944    | 9                  |
| $\gamma$ -spectrometry | 1.17544   | 0.11222    | 1                  |

Where  $\bar{x}$  = mean (analyst result/NPL result) and  $\delta x$  is the uncertainty of this ratio.

A t - test between the means of the results from the two different methods gives  $t = 4.259$  between  $\alpha$  and  $\gamma$  spectrometry with 10 degrees of freedom. This is significant at the 0.1% confidence limit, so it is concluded that there is a significant difference between the two techniques. Overall, the results show no bias.

## 6 RESULTS FOR $\beta$ -EMITTERS

Fourteen analysts returned results for some or all of the pure  $\beta$ -emitters in the sample. As with the  $\alpha$ -emitters, the separation chemistry required was not too onerous and, in general, the results were fairly encouraging, see Tables 9 and 10.

### 6.1 $^3\text{H}$

Eleven results were returned for this nuclide, with a variety of techniques being used. All involved distillation of some sort with six analysts boiling the solution and the other five using a low temperature evaporation. In each case, liquid scintillation counting was employed to assay the activity of the distillate and no carry over of other nuclides was noted. One analyst assumed a 95% factor for recovery, presumably based on previous experience of isotopic separation. There appears to be no difference between the two techniques. When the obvious outliers are excluded from the results, there appears to be a general negative bias.

### 6.2 $^{90}\text{Sr}/^{90}\text{Y}$

There were eight results returned for this nuclide with a large spread of results. The analytical techniques varied somewhat and this may explain the differences in the results. The methods used fall into two groups; (i) separating strontium from the

sample and its yttrium daughter followed by in-growth and measurement of  $^{90}\text{Y}$  and (ii) separation of  $^{90}\text{Y}$  by solvent extraction followed by purification of yttrium and then counting. The two analysts using the solvent extraction technique reported high results and it is assumed that this may be due to the extraction of  $^{154}\text{Eu}$  as well as  $^{90}\text{Y}$ . This would not normally be a problem - the fission yield of  $^{90}\text{Sr}/^{90}\text{Y}$  is many times greater than that for  $^{154}\text{Eu}$  and hence any contamination by  $^{154}\text{Eu}$  would not be noticeable. The solvent extraction technique is considerably shorter than the 'in-growth' technique and may be used as a rapid method for screening or emergency measurements. Using such a technique cannot be criticised on the basis of this exercise since, as noted above, the sample contains much more  $^{154}\text{Eu}$  than one would normally find; also, a high result for a rapid method is no bad thing since it may prompt a further investigation of the sample in question. The low results seem to have been caused by mathematical errors on the part of the analyst and one is still under investigation. The remaining results are spread about the mean.

### 6.3

Unfortunately, no results were reported for this nuclide.

## 7 RESULTS FOR $\beta/\gamma$ -EMITTERS

As usual, most of the data were reported for the  $\beta/\gamma$ -emitters, with a total of 32 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  $^{152}\text{Eu}$  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 [12] (a copy of which is available free of charge from NPL - contact Dr D Smith). Other data sources used were all well 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.

### 7.1

This is an activation product, widely observed in radioactive waste. The results for this nuclide were comparable to the previous exercise. No bias is observed; this is expected since it is a calibration nuclide. Debertin and Schötzig [13] have measured a correction factor for cascade summing of  $\sim 2\%$  for extended geometries, although it is not known how many participants applied such a correction.

## 7.2

The results for this nuclide showed a wide spread, with a high bias. This reflects the interference observed with  $^{154}\text{Eu}$ ; the  $\gamma$ -ray emission probabilities of these two nuclides are shown in Table 5.

Table 5 Selected  $\gamma$ -ray emission probabilities for  $^{95}\text{Zr}$  and  $^{154}\text{Eu}$

| Nuclide           | keV   | $P_\gamma$          | Reference | Summing<br>Correction [13]<br>(Approx %) |
|-------------------|-------|---------------------|-----------|------------------------------------------|
| $^{95}\text{Zr}$  | 724.2 | $0.441 \pm 0.009$   | [14]      | 0                                        |
| $^{154}\text{Eu}$ | 723.3 | $0.202 \pm 0.002$   | [12]      | ~ 6                                      |
| $^{95}\text{Zr}$  | 756.7 | $0.545 \pm 0.002$   | [14]      | 0                                        |
| $^{154}\text{Eu}$ | 756.8 | $0.0458 \pm 0.0006$ | [12]      | -                                        |

It is possible to resolve the peaks at 724.2 and 723.2 keV with modern detectors, although not the peaks at 756.7 and 756.8 keV. It should be apparent, however, from the peak ratios whether there may be another nuclide present. According to Debertin and Schötzig [13], there are no summing problems with this nuclide. The quality of the results suggests that the mutual interference problem is not a problem for many analysts.

## 7.3

This is the daughter of  $^{95}\text{Zr}$ . At the time of the intercomparison exercise, this was 'growing-in' and was not in radioactive equilibrium with the  $^{95}\text{Zr}$  parent. It was included in the exercise since it was hoped that it would pose an additional facet to the measurement of  $^{95}\text{Zr}$ . It was clear from the measurements made that the ratio of  $^{95}\text{Nb}$  to  $^{95}\text{Zr}$  was not at the theoretical maximum of  $2.2041 \pm 0.0034$  and that a non-equilibrium situation was at hand. Unfortunately, the data request was for the activity at 1 January 1992, rather than the measurement date. Now, there is a choice as to how the data is decayed back; (i) using the  $^{95}\text{Nb}$  half life ( $34.975 \pm 0.007$  d [12]), (ii) using the  $^{95}\text{Zr}$  half life ( $64.04 \pm 0.04$  d [14]), or (iii) assuming equilibrium, by calculating the separation date from the activity ratio and then calculate the ingrowth on 1 January 1992, using Bateman's solution [15]. Without further information from the analysts, it was not possible to know how the decay back to 1 January 1992 was calculated; it is possible to make a guess from the reported data, but it is rather unfair to do so and NPL acknowledges that it too has succumbed to the failing, previously criticised in others, in not having provided a data reporting protocol which fully recognises all of the intricacies of the analytical and reporting requirements. For this reason, the  $^{95}\text{Nb}$  results are not included.

$^{134}\text{Cs}$ 

This is another activation product. It was included, as in previous years, due to its ubiquitous presence in nuclear waste and its rapid uptake into various ecosystems. In addition, it exhibits cascade summing. As in previous years, the results are biased low by about 8%. The correction factor quoted in Debertin and Schötzig [13] is ~ 6%, which would bring most of the results to the point where they would be statistically indistinguishable from the true value. It is curious that, having identified the effects of cascade summing in both of the previous exercises, participants have, apparently, failed on this occasion to have applied any correction.

 $^{137}\text{Cs}$ 

This is a common fission product which is also used as a calibration nuclide. No bias is observable for the results for this nuclide and most of the results are indistinguishable from the true value.

 $^{144}\text{Ce}$ 

This is another fission product. It has a  $\gamma$ -ray at 133.5 keV which occurs in a part of the spectrum where an accurate energy calibration is essential for deriving the correct efficiency. This is not always easy and the results show a wide spread. However, most of the results are statistically acceptable and there appear to be no real problems with the measurement of this radionuclide.

 $^{154}\text{Eu}$ 

$^{154}\text{Eu}$  is an activation product of stable europium. Its decay produces several  $\gamma$ -lines, most of which are coincident with  $\gamma$ -lines from the decay of  $^{57}\text{Co}$ ,  $^{95}\text{Zr}$  and  $^{22}\text{Na}$ . There are enough 'in the clear', however, to permit identification. The results show a low bias of ~ 6%. The correction factor [13] due to cascade summing for the principal  $\gamma$  lines is of the order 4-8% and thus similar comments to those for  $^{134}\text{Cs}$  apply. The difficulties incurred with this nuclide were that (i) it does exhibit cascade summing, (ii) it may interfere with the analysis of  $^{90}\text{Sr}$  via  $^{90}\text{Y}$  ingrowth, and (iii) it may cause difficulties for  $^{95}\text{Zr}$  analysis by  $\gamma$ -spectrometry.

## Discussion

Overall, the results were good for the  $\beta/\gamma$ -emitters. The problem of cascade summing is a perennial one and, although mention has been made of some correction factors [13], it should be noted that these are matrix, geometry and detector dependent. It is thus difficult to correct for unless one is able to invest considerable time and money into determining the relevant correction factors. In the majority of measurements, where activity levels are sufficiently low so that screening criteria may be used, it is probably only worthwhile noting that the nuclide being measured exhibits the effect. The exception to this, of course, is when the result is at or near

an action or intervention limit when it is clear that an estimate of the summing effect is needed to enable more accurate values to be quoted.

As far as the interfering nuclides are concerned, there appeared to be few problems, although there was a batch of results that was high for  $^{95}\text{Zr}$ . Some participants also failed to identify  $^{154}\text{Eu}$  and  $^{144}\text{Ce}$ , reporting  $^{22}\text{Na}$ ,  $^{57}\text{Co}$ , and  $^{207}\text{Bi}$  instead. It should be clear from the peak/activity reports that these results are erroneous and it is worrying, therefore, that these discrepancies were not identified.

## 8 CONCLUSIONS

This exercise produced no real surprises. The results for the various nuclides were generally satisfactory, although it was disappointing that no-one made use of ICP-MS to measure  $^{99}\text{Tc}$  or  $^{239}\text{Pu}$ . It seems that a natural break has been reached in these exercises in that the level of competence, which is at a high level anyway, is not increasing noticeably. Also, the level of participation is dropping as time passes which may imply that the measurement of radionuclides in simple solutions is of less interest than other matrices to the environmental laboratory.

As in previous exercises, there is some concern that the uncertainties quoted are unrealistically low in some cases, presumably being due to the consideration of sample counting data only. This narrows the 'band of acceptability' for the data quoted and can only disadvantage the analyst.

Corrections for cascade summing are still not being applied, but there has been a profound improvement in the quality of nuclear decay data used.

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.

## 9 FUTURE EXERCISES

As noted above, there may be an interest in other matrices and nuclides. However, it is important for the user community to inform the NPL of what is required - we cannot always guess correctly. Possibilities include the following:

- 1) Long lived nuclides ( $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{231}\text{Pu}$ ,  $^{233}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$ ,  $^{238}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{239}\text{Pu}$ ) in aqueous solutions at levels up to 2,000 Bq/kg with measurement by radiometric and non-radiometric techniques.
- 2)  $\alpha$ ,  $\beta$  or  $\beta/\gamma$ -emitters chemically bound to kaolin.
- 3)  $\alpha$ ,  $\beta$  or  $\beta/\gamma$ -emitters surface bound (not chemically bound) to milk powder.

- 4) Parent/daughter nuclides.
- 5)  $\gamma$ -nuclides that are mutually interfering.

These possibilities will be addressed at the follow-up meeting, in an international workshop later this year and by other means.

## 10 ACKNOWLEDGEMENTS

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Table 6 u-test results

| Nuclide           | Limits    | Number of Results |
|-------------------|-----------|-------------------|
| <sup>239</sup> Pu | < 1.64    | 4                 |
|                   | 1.64-1.95 | 0                 |
|                   | 1.95-2.58 | 2                 |
|                   | 2.58-3.29 | 2                 |
|                   | > 3.29    | 5                 |
| <sup>241</sup> Am | < 1.64    | 6                 |
|                   | 1.64-1.95 | 1                 |
|                   | 1.95-2.58 | 3                 |
|                   | 2.58-3.29 | 1                 |
|                   | > 3.29    | 2                 |
| <sup>3</sup> H    | < 1.64    | 5                 |
|                   | 1.64-1.95 | 0                 |
|                   | 1.95-2.58 | 1                 |
|                   | 2.58-3.29 | 1                 |
|                   | > 3.29    | 6                 |
| <sup>90</sup> Sr  | < 1.64    | 5                 |
|                   | 1.64-1.95 | 0                 |
|                   | 1.95-2.58 | 1                 |
|                   | 2.58-3.29 | 0                 |
|                   | > 3.29    | 4                 |
| <sup>60</sup> Co  | < 1.64    | 26                |
|                   | 1.64-1.95 | 1                 |
|                   | 1.95-2.58 | 2                 |
|                   | 2.58-3.29 | 2                 |
|                   | > 3.29    | 3                 |
| <sup>95</sup> Zr  | < 1.64    | 14                |
|                   | 1.64-1.95 | 3                 |
|                   | 1.95-2.58 | 2                 |
|                   | 2.58-3.29 | 3                 |
|                   | > 3.29    | 13                |
| <sup>134</sup> Cs | < 1.64    | 12                |
|                   | 1.64-1.95 | 0                 |
|                   | 1.95-2.58 | 3                 |
|                   | 2.58-3.29 | 2                 |
|                   | > 3.29    | 17                |

Table 6 u-test results (con't)

| Nuclide           | Limits    | Number of Results |
|-------------------|-----------|-------------------|
| <sup>137</sup> Cs | < 1.64    | 23                |
|                   | 1.64-1.95 | 2                 |
|                   | 1.95-2.58 | 1                 |
|                   | 2.58-3.29 | 4                 |
|                   | > 3.29    | 4                 |
| <sup>144</sup> Ce | < 1.64    | 27                |
|                   | 1.64-1.95 | 2                 |
|                   | 1.95-2.58 | 2                 |
|                   | 2.58-3.29 | 2                 |
|                   | > 3.29    | 1                 |
| <sup>154</sup> Eu | < 1.64    | 10                |
|                   | 1.64-1.95 | 2                 |
|                   | 1.95-2.58 | 5                 |
|                   | 2.58-3.29 | 3                 |
|                   | > 3.29    | 12                |

Table 7  $^{239}\text{Pu}$  results

| Sample | NPL Data                            |                                         | Analyst Data                        |                                         | Deviation % | u-test |
|--------|-------------------------------------|-----------------------------------------|-------------------------------------|-----------------------------------------|-------------|--------|
|        | $^{239}\text{Pu}/\text{Bq kg}^{-1}$ | $\pm ^{239}\text{Pu}/\text{Bq kg}^{-1}$ | $^{239}\text{Pu}/\text{Bq kg}^{-1}$ | $\pm ^{239}\text{Pu}/\text{Bq kg}^{-1}$ |             |        |
| A 0001 | 16.0                                | 0.2                                     | 13.6                                | 0.6                                     | - 14.6      | 3.44   |
| A 0003 | 16.6                                | 0.3                                     | 17.9                                | 0.3                                     | 7.8         | 3.20   |
| A 0004 | 15.9                                | 0.2                                     | 12.3                                | 1.1                                     | - 22.3      | 3.08   |
| A 0005 | 16.2                                | 0.3                                     | 17.7                                | 1.0                                     | 8.9         | 1.36   |
| A 0006 | 16.4                                | 0.3                                     | 15.3                                | 0.4                                     | - 6.8       | 2.32   |
| A 0007 | 15.9                                | 0.2                                     | 17.6                                | 0.2                                     | 10.4        | 4.91   |
| A 0008 | 16.1                                | 0.2                                     | 17.6                                | 1.3                                     | 9.1         | 1.09   |
| A 0009 | 16.2                                | 0.2                                     | 17.9                                | 0.7                                     | 10.4        | 2.26   |
| A 0010 | 16.3                                | 0.3                                     | 5.0                                 | 0.4                                     | - 69.3      | 23.89  |
| A 0012 | 16.2                                | 0.2                                     | 19.0                                | 0.0                                     | 17.3        | 11.27  |
| A 0013 | 16.4                                | 0.3                                     | 18.6                                | 0.0                                     | 13.6        | 8.85   |
| A 0016 | 19.3                                | 0.3                                     | 18.4                                | 0.8                                     | - 5.1       | 1.17   |
| A 0017 | 19.1                                | 0.3                                     | 20.7                                | 1.2                                     | 8.6         | 1.32   |

Table 8  $^{241}\text{Am}$  results

| Sample | NPL Data                            |                                         | Analyst Data                        |                                         | Deviation % | u-test |
|--------|-------------------------------------|-----------------------------------------|-------------------------------------|-----------------------------------------|-------------|--------|
|        | $^{241}\text{Am}/\text{Bq kg}^{-1}$ | $\pm ^{241}\text{Am}/\text{Bq kg}^{-1}$ | $^{241}\text{Am}/\text{Bq kg}^{-1}$ | $\pm ^{241}\text{Am}/\text{Bq kg}^{-1}$ |             |        |
| A 0001 | 8.6                                 | 0.1                                     | 8.1                                 | 0.7                                     | - 6.5       | 0.80   |
| A 0003 | 9.0                                 | 0.1                                     | 9.1                                 | 0.2                                     | 1.8         | 0.69   |
| A 0004 | 8.6                                 | 0.1                                     | 8.9                                 | 0.2                                     | 4.1         | 2.18   |
| A 0005 | 8.8                                 | 0.1                                     | 7.9                                 | 0.2                                     | - 9.8       | 3.69   |
| A 0006 | 8.8                                 | 0.1                                     | 8.0                                 | 0.3                                     | - 9.7       | 3.03   |
| A 0007 | 8.6                                 | 0.1                                     | 8.5                                 | 0.2                                     | - 1.5       | 0.81   |
| A 0008 | 8.7                                 | 0.1                                     | 7.8                                 | 0.8                                     | - 11.1      | 1.23   |
| A 0009 | 8.8                                 | 0.1                                     | 9.8                                 | 0.9                                     | 11.8        | 1.14   |
| A 0010 | 8.8                                 | 0.1                                     | 10.5                                | 0.8                                     | 19.9        | 2.32   |
| A 0012 | 8.8                                 | 0.1                                     | 8.9                                 | 0.0                                     | 1.7         | 2.45   |
| A 0013 | 8.8                                 | 0.1                                     | 8.6                                 | 0.0                                     | - 2.8       | 4.07   |
| A 0016 | 10.4                                | 0.1                                     | 9.4                                 | 0.9                                     | - 10.5      | 1.28   |
| A 0017 | 10.3                                | 0.1                                     | 9.3                                 | 0.6                                     | - 9.8       | 1.66   |

Table 9  $^3\text{H}$  results

| Sample  | NPL Data                       |                                    | Analyst Data                   |                                    | Deviation % | u-test |
|---------|--------------------------------|------------------------------------|--------------------------------|------------------------------------|-------------|--------|
|         | $^3\text{H}/\text{Bq kg}^{-1}$ | $\pm ^3\text{H}/\text{Bq kg}^{-1}$ | $^3\text{H}/\text{Bq kg}^{-1}$ | $\pm ^3\text{H}/\text{Bq kg}^{-1}$ |             |        |
| BG 0006 | 606.1                          | 4.1                                | 548.3                          | 43.5                               | - 9.5       | 1.32   |
| BG 0008 | 608.0                          | 4.1                                | 1203.0                         | 118.0                              | 97.9        | 5.04   |
| BG 0009 | 604.9                          | 4.1                                | 579.9                          | 9.2                                | - 4.1       | 2.47   |
| BG 0011 | 616.3                          | 4.2                                | 562.8                          | 128.8                              | - 8.7       | 0.41   |
| BG 0016 | 604.3                          | 4.1                                | 587.3                          | 3.8                                | - 2.8       | 3.03   |
| BG 0021 | 599.6                          | 4.1                                | 480.0                          | 20.0                               | - 19.9      | 5.86   |
| BG 0023 | 590.9                          | 4.0                                | 571.9                          | 32.0                               | - 3.2       | 0.59   |
| BG 0024 | 598.9                          | 4.1                                | 1700.0                         | 0.0                                | 183.8       | 270.35 |
| BG 0025 | 668.4                          | 4.5                                | 1900.0                         | 0.0                                | 184.3       | 270.99 |
| BG 0031 | 583.5                          | 4.0                                | 555.2                          | 60.9                               | - 4.8       | 0.46   |
| BG 0032 | 602.9                          | 4.1                                | 579.0                          | 14.0                               | - 4.0       | 1.64   |
| BG 0037 | 567.1                          | 3.9                                | 617.7                          | 10.5                               | 8.9         | 4.53   |
| BG 0038 | 757.8                          | 5.2                                | 505.0                          | 5.0                                | - 33.4      | 35.20  |

Table 10  $^{90}\text{Sr}$  results

| Sample  | NPL Data                           |                                        | Analyst Data                       |                                        | Deviation % | u-test |
|---------|------------------------------------|----------------------------------------|------------------------------------|----------------------------------------|-------------|--------|
|         | $^{90}\text{Sr}/\text{Bq kg}^{-1}$ | $\pm ^{90}\text{Sr}/\text{Bq kg}^{-1}$ | $^{90}\text{Sr}/\text{Bq kg}^{-1}$ | $\pm ^{90}\text{Sr}/\text{Bq kg}^{-1}$ |             |        |
| BG 0001 | 44.4                               | 0.2                                    | 249.0                              | 30.0                                   | 460.4       | 6.82   |
| BG 0009 | 40.8                               | 0.2                                    | 39.9                               | 1.0                                    | - 2.2       | 0.89   |
| BG 0016 | 40.7                               | 0.2                                    | 47.3                               | 2.7                                    | 16.1        | 2.41   |
| BG 0020 | 40.6                               | 0.2                                    | 40.8                               | 0.4                                    | 0.3         | 0.28   |
| BG 0023 | 39.8                               | 0.2                                    | 36.3                               | 3.2                                    | - 8.8       | 1.10   |
| BG 0024 | 40.4                               | 0.2                                    | 40.6                               | 0.0                                    | 0.6         | 1.44   |
| BG 0025 | 45.0                               | 0.2                                    | 46.0                               | 0.0                                    | 2.1         | 5.11   |
| BG 0031 | 39.3                               | 0.2                                    | 35.9                               | 0.7                                    | - 8.7       | 4.67   |
| BG 0037 | 38.2                               | 0.2                                    | 23.5                               | 0.7                                    | - 38.4      | 19.62  |
| BG 0038 | 51.1                               | 0.2                                    | 50.4                               | 4.4                                    | - 1.3       | 0.15   |

Table 1  $^{60}\text{Co}$  results

| Sample  | NPL Data                           |                                        | Analyst Data                       |                                        | Deviation % | u-test |
|---------|------------------------------------|----------------------------------------|------------------------------------|----------------------------------------|-------------|--------|
|         | $^{60}\text{Co}/\text{Bq kg}^{-1}$ | $\pm ^{60}\text{Co}/\text{Bq kg}^{-1}$ | $^{60}\text{Co}/\text{Bq kg}^{-1}$ | $\pm ^{60}\text{Co}/\text{Bq kg}^{-1}$ |             |        |
| BG 0001 | 42.6                               | 0.2                                    | 37.9                               | 1.0                                    | - 11.0      | 4.84   |
| BG 0002 | 38.2                               | 0.2                                    | 39.7                               | 2.1                                    | 3.9         | 0.71   |
| BG 0004 | 39.7                               | 0.2                                    | 40.8                               | 0.9                                    | 2.8         | 1.21   |
| BG 0005 | 39.9                               | 0.2                                    | 40.0                               | 0.6                                    | 0.2         | 0.10   |
| BG 0006 | 39.1                               | 0.2                                    | 39.5                               | 1.1                                    | 0.8         | 0.30   |
| BG 0007 | 38.7                               | 0.2                                    | 39.0                               | 3.1                                    | 0.7         | 0.09   |
| BG 0008 | 39.3                               | 0.2                                    | 39.2                               | 0.5                                    | - 0.1       | 0.11   |
| BG 0009 | 39.1                               | 0.2                                    | 39.1                               | 0.3                                    | 0.1         | 0.15   |
| BG 0010 | 39.4                               | 0.2                                    | 40.2                               | 2.5                                    | 2.0         | 0.31   |
| BG 0011 | 39.8                               | 0.2                                    | 44.3                               | 3.5                                    | 11.3        | 1.29   |
| BG 0013 | 39.3                               | 0.2                                    | 39.8                               | 3.9                                    | 1.3         | 0.12   |
| BG 0014 | 39.2                               | 0.2                                    | 37.1                               | 0.9                                    | - 5.3       | 2.25   |
| BG 0015 | 36.7                               | 0.2                                    | 38.0                               | 0.4                                    | 3.3         | 2.86   |
| BG 0016 | 39.0                               | 0.2                                    | 38.8                               | 0.4                                    | - 0.5       | 0.48   |
| BG 0017 | 39.5                               | 0.2                                    | 39.7                               | 1.0                                    | 0.4         | 0.17   |
| BG 0019 | 39.3                               | 0.2                                    | 38.4                               | 1.0                                    | - 2.3       | 0.91   |
| BG 0020 | 38.9                               | 0.2                                    | 41.0                               | 2.0                                    | 5.3         | 1.03   |
| BG 0021 | 38.7                               | 0.2                                    | 41.4                               | 1.0                                    | 6.9         | 2.65   |
| BG 0022 | 38.0                               | 0.2                                    | 39.4                               | 1.4                                    | 3.6         | 0.98   |
| BG 0023 | 38.2                               | 0.2                                    | 38.0                               | 2.5                                    | - 0.4       | 0.06   |
| BG 0024 | 38.7                               | 0.2                                    | 38.8                               | 0.5                                    | 0.3         | 0.26   |
| BG 0025 | 43.2                               | 0.2                                    | 42.2                               | 1.5                                    | - 2.2       | 0.63   |
| BG 0026 | 38.0                               | 0.2                                    | 37.0                               | 0.5                                    | - 2.7       | 2.08   |
| BG 0027 | 38.9                               | 0.2                                    | 38.0                               | 1.0                                    | - 2.2       | 0.86   |
| BG 0028 | 38.3                               | 0.2                                    | 41.0                               | 0.8                                    | 7.0         | 3.29   |
| BG 0031 | 37.7                               | 0.2                                    | 40.2                               | 0.5                                    | 6.7         | 4.78   |
| BG 0032 | 38.9                               | 0.2                                    | 37.6                               | 0.7                                    | - 3.4       | 1.84   |
| BG 0033 | 46.5                               | 0.2                                    | 46.4                               | 0.9                                    | - 0.3       | 0.43   |
| BG 0034 | 44.2                               | 0.2                                    | 44.2                               | 1.5                                    | - 0.1       | 0.03   |
| BG 0035 | 46.9                               | 0.2                                    | 50.0                               | 5.0                                    | 6.5         | 0.61   |
| BG 0036 | 33.9                               | 0.2                                    | 33.9                               | 0.4                                    | - 0.1       | 0.05   |
| BG 0037 | 48.9                               | 0.2                                    | 50.0                               | 2.0                                    | 2.2         | 0.53   |
| BG 0038 | 37.1                               | 0.2                                    | 34.6                               | 1.6                                    | - 6.9       | 1.57   |
| BG 0040 | 39.5                               | 0.2                                    | 40.7                               | 1.4                                    | 3.0         | 0.85   |

Table 12  $^{95}\text{Zr}$  results

| Sample  | NPL Data                           |                                        | Analyst Data                       |                                        | Deviation % | u-test |
|---------|------------------------------------|----------------------------------------|------------------------------------|----------------------------------------|-------------|--------|
|         | $^{95}\text{Zr}/\text{Bq kg}^{-1}$ | $\pm ^{95}\text{Zr}/\text{Bq kg}^{-1}$ | $^{95}\text{Zr}/\text{Bq kg}^{-1}$ | $\pm ^{95}\text{Zr}/\text{Bq kg}^{-1}$ |             |        |
| BG 0001 | 259.1                              | 3.0                                    | 345.6                              | 8.1                                    | 33.4        | 10.03  |
| BG 0002 | 232.4                              | 2.7                                    | 312.7                              | 17.2                                   | 34.5        | 4.61   |
| BG 0004 | 241.5                              | 2.8                                    | 264.1                              | 6.7                                    | 9.3         | 3.11   |
| BG 0005 | 243.0                              | 2.8                                    | 262.0                              | 5.0                                    | 7.8         | 3.30   |
| BG 0006 | 238.1                              | 2.8                                    | 248.9                              | 16.9                                   | 4.5         | 0.63   |
| BG 0007 | 235.7                              | 2.7                                    | 278.0                              | 22.0                                   | 17.9        | 1.91   |
| BG 0008 | 238.9                              | 2.8                                    | 269.7                              | 5.9                                    | 12.9        | 4.72   |
| BG 0009 | 237.7                              | 2.8                                    | 239.5                              | 1.6                                    | 0.8         | 0.58   |
| BG 0010 | 239.9                              | 2.8                                    | 251.7                              | 16.2                                   | 4.9         | 0.72   |
| BG 0011 | 242.1                              | 2.8                                    | 254.0                              | 30.4                                   | 4.9         | 0.39   |
| BG 0013 | 239.2                              | 2.8                                    | 239.6                              | 15.7                                   | 0.2         | 0.02   |
| BG 0014 | 238.3                              | 2.8                                    | 222.5                              | 9.7                                    | - 6.6       | 1.57   |
| BG 0015 | 223.6                              | 2.6                                    | 227.1                              | 4.1                                    | 1.6         | 0.72   |
| BG 0016 | 237.4                              | 2.8                                    | 269.9                              | 3.6                                    | 13.7        | 7.17   |
| BG 0017 | 240.5                              | 2.8                                    | 303.1                              | 15.9                                   | 26.0        | 3.87   |
| BG 0019 | 239.1                              | 2.8                                    | 293.0                              | 6.7                                    | 22.5        | 7.40   |
| BG 0020 | 237.0                              | 2.7                                    | 238.0                              | 14.0                                   | 0.4         | 0.07   |
| BG 0021 | 235.6                              | 2.7                                    | 242.0                              | 7.9                                    | 2.7         | 0.77   |
| BG 0022 | 231.4                              | 2.7                                    | 228.4                              | 6.6                                    | - 1.3       | 0.41   |
| BG 0023 | 232.2                              | 2.7                                    | 242.4                              | 19.2                                   | 4.4         | 0.53   |
| BG 0024 | 235.3                              | 2.7                                    | 435.4                              | 6.0                                    | 85.0        | 30.35  |
| BG 0025 | 262.6                              | 3.0                                    | 490.2                              | 6.0                                    | 86.7        | 33.82  |
| BG 0026 | 231.5                              | 2.7                                    | 285.0                              | 6.3                                    | 23.1        | 7.78   |
| BG 0027 | 236.4                              | 2.7                                    | 249.0                              | 6.0                                    | 5.3         | 1.91   |
| BG 0028 | 233.1                              | 2.7                                    | 286.0                              | 10.0                                   | 22.7        | 5.10   |
| BG 0031 | 229.3                              | 2.7                                    | 160.1                              | 3.8                                    | - 30.2      | 14.91  |
| BG 0032 | 236.9                              | 2.7                                    | 274.0                              | 5.0                                    | 15.7        | 6.50   |
| BG 0033 | 282.9                              | 3.3                                    | 308.6                              | 10.7                                   | 9.1         | 2.30   |
| BG 0034 | 269.2                              | 3.1                                    | 260.3                              | 14.3                                   | - 3.3       | 0.61   |
| BG 0035 | 285.6                              | 3.3                                    | 295.0                              | 30.0                                   | 3.3         | 0.31   |
| BG 0036 | 206.4                              | 2.4                                    | 202.3                              | 5.5                                    | - 2.0       | 0.69   |
| BG 0038 | 297.8                              | 3.5                                    | 256.0                              | 17.0                                   | - 14.0      | 2.41   |
| BG 0039 | 226.1                              | 2.6                                    | 290.0                              | 22.0                                   | 28.3        | 2.89   |
| BG 0040 | 240.6                              | 2.8                                    | 290.5                              | 29.1                                   | 20.7        | 1.71   |

Table 13  $^{134}\text{Cs}$  results

| Sample  | NPL Data                            |                                         | Analyst Data                        |                                         | Deviation % | u-test |
|---------|-------------------------------------|-----------------------------------------|-------------------------------------|-----------------------------------------|-------------|--------|
|         | $^{134}\text{Cs}/\text{Bq kg}^{-1}$ | $\pm ^{134}\text{Cs}/\text{Bq kg}^{-1}$ | $^{134}\text{Cs}/\text{Bq kg}^{-1}$ | $\pm ^{134}\text{Cs}/\text{Bq kg}^{-1}$ |             |        |
| BG 0001 | 298.3                               | 2.2                                     | 249.8                               | 5.1                                     | - 16.3      | 8.73   |
| BG 0002 | 267.6                               | 2.0                                     | 253.5                               | 13.2                                    | - 5.3       | 1.06   |
| BG 0004 | 278.1                               | 2.1                                     | 259.7                               | 1.7                                     | - 6.6       | 6.89   |
| BG 0005 | 279.8                               | 2.1                                     | 257.0                               | 3.0                                     | - 8.2       | 6.27   |
| BG 0006 | 274.2                               | 2.0                                     | 258.1                               | 3.9                                     | - 5.9       | 3.69   |
| BG 0007 | 271.4                               | 2.0                                     | 243.0                               | 18.8                                    | - 10.5      | 1.50   |
| BG 0008 | 275.1                               | 2.0                                     | 235.2                               | 2.2                                     | - 14.5      | 13.30  |
| BG 0009 | 273.6                               | 2.0                                     | 253.1                               | 0.5                                     | - 7.5       | 9.84   |
| BG 0010 | 276.2                               | 2.0                                     | 240.4                               | 4.8                                     | - 13.0      | 6.87   |
| BG 0011 | 278.8                               | 2.1                                     | 264.0                               | 13.7                                    | - 5.3       | 1.07   |
| BG 0013 | 275.4                               | 2.0                                     | 258.8                               | 6.4                                     | - 6.0       | 2.48   |
| BG 0014 | 274.4                               | 2.0                                     | 253.3                               | 2.6                                     | - 7.7       | 6.36   |
| BG 0015 | 257.5                               | 1.9                                     | 255.4                               | 0.9                                     | - 0.8       | 0.98   |
| BG 0016 | 273.4                               | 2.0                                     | 240.2                               | 1.3                                     | - 12.2      | 13.91  |
| BG 0017 | 277.0                               | 2.0                                     | 250.9                               | 2.9                                     | - 9.4       | 7.34   |
| BG 0019 | 275.4                               | 2.0                                     | 268.0                               | 5.0                                     | - 2.7       | 1.35   |
| BG 0020 | 272.8                               | 2.0                                     | 241.0                               | 13.0                                    | - 11.7      | 2.42   |
| BG 0021 | 271.3                               | 2.0                                     | 254.7                               | 12.8                                    | - 6.1       | 1.28   |
| BG 0022 | 266.4                               | 2.0                                     | 232.3                               | 2.0                                     | - 12.8      | 12.14  |
| BG 0023 | 267.3                               | 2.0                                     | 247.7                               | 13.6                                    | - 7.4       | 1.43   |
| BG 0024 | 271.0                               | 2.0                                     | 256.6                               | 1.5                                     | - 5.3       | 5.74   |
| BG 0025 | 302.4                               | 2.2                                     | 290.6                               | 3.0                                     | - 3.9       | 3.15   |
| BG 0026 | 266.6                               | 2.0                                     | 232.0                               | 5.4                                     | - 13.0      | 5.99   |
| BG 0027 | 272.2                               | 2.0                                     | 258.0                               | 13.0                                    | - 5.2       | 1.08   |
| BG 0028 | 268.4                               | 2.0                                     | 258.0                               | 7.0                                     | - 3.9       | 1.43   |
| BG 0031 | 264.0                               | 2.0                                     | 251.9                               | 0.8                                     | - 4.6       | 5.73   |
| BG 0032 | 272.8                               | 2.0                                     | 250.0                               | 9.0                                     | - 8.3       | 2.47   |
| BG 0033 | 325.7                               | 2.4                                     | 318.3                               | 8.7                                     | - 2.3       | 0.82   |
| BG 0034 | 310.0                               | 2.3                                     | 284.3                               | 8.8                                     | - 8.3       | 2.82   |
| BG 0035 | 328.8                               | 2.4                                     | 306.0                               | 20.0                                    | - 6.9       | 1.13   |
| BG 0036 | 237.7                               | 1.8                                     | 207.1                               | 1.4                                     | - 12.9      | 13.41  |
| BG 0038 | 242.8                               | 2.5                                     | 317.0                               | 4.0                                     | - 7.5       | 5.45   |
| BG 0039 | 260.3                               | 1.9                                     | 214.0                               | 3.5                                     | - 17.8      | 11.49  |
| BG 0040 | 277.0                               | 2.1                                     | 273.8                               | 17.0                                    | - 1.2       | 0.19   |

Table 14  $^{137}\text{Cs}$  results

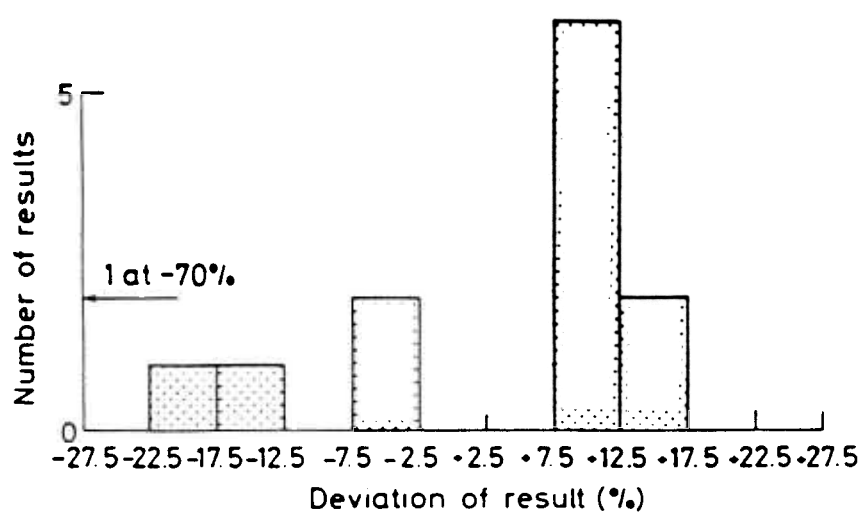
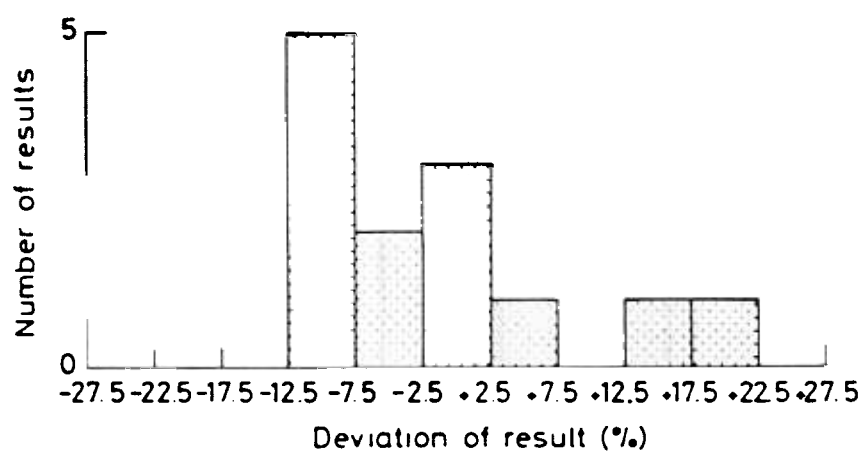
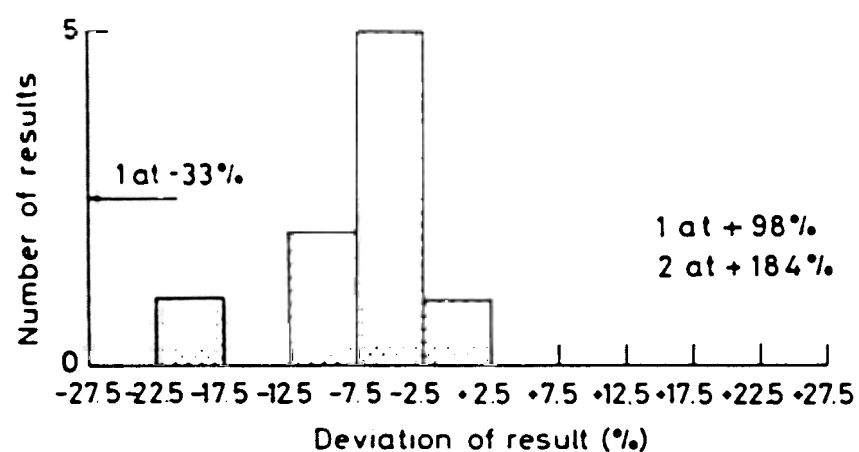
| Sample  | NPL Data                            |                                         | Analyst Data                        |                                         | Deviation % | u-test |
|---------|-------------------------------------|-----------------------------------------|-------------------------------------|-----------------------------------------|-------------|--------|
|         | $^{137}\text{Cs}/\text{Bq kg}^{-1}$ | $\pm ^{137}\text{Cs}/\text{Bq kg}^{-1}$ | $^{137}\text{Cs}/\text{Bq kg}^{-1}$ | $\pm ^{137}\text{Cs}/\text{Bq kg}^{-1}$ |             |        |
| BG 0001 | 100.9                               | 0.8                                     | 92.9                                | 2.0                                     | - 7.9       | 3.74   |
| BG 0002 | 90.5                                | 0.7                                     | 94.4                                | 4.8                                     | 4.3         | 0.81   |
| BG 0004 | 94.0                                | 0.7                                     | 96.7                                | 1.2                                     | 2.8         | 1.91   |
| BG 0005 | 94.6                                | 0.7                                     | 96.0                                | 2.0                                     | 1.5         | 0.65   |
| BG 0006 | 92.7                                | 0.7                                     | 94.2                                | 1.6                                     | 1.5         | 0.83   |
| BG 0007 | 91.8                                | 0.7                                     | 91.0                                | 7.1                                     | - 0.8       | 0.11   |
| BG 0008 | 93.0                                | 0.7                                     | 90.8                                | 1.9                                     | - 2.4       | 1.09   |
| BG 0009 | 92.5                                | 0.7                                     | 92.9                                | 0.4                                     | 0.4         | 0.46   |
| BG 0010 | 93.4                                | 0.7                                     | 98.2                                | 2.9                                     | 5.1         | 1.61   |
| BG 0011 | 94.3                                | 0.7                                     | 98.0                                | 6.3                                     | 3.9         | 0.59   |
| BG 0013 | 93.1                                | 0.7                                     | 95.2                                | 3.5                                     | 2.2         | 0.58   |
| BG 0014 | 92.8                                | 0.7                                     | 87.1                                | 1.8                                     | - 6.2       | 3.03   |
| BG 0015 | 87.1                                | 0.7                                     | 90.6                                | 0.9                                     | 4.0         | 3.06   |
| BG 0016 | 92.4                                | 0.7                                     | 91.6                                | 0.6                                     | - 0.9       | 0.91   |
| BG 0017 | 93.7                                | 0.7                                     | 94.9                                | 1.5                                     | 1.3         | 0.75   |
| BG 0019 | 93.1                                | 0.7                                     | 96.7                                | 2.0                                     | 3.9         | 1.68   |
| BG 0020 | 92.3                                | 0.7                                     | 90.0                                | 5.0                                     | - 2.4       | 0.45   |
| BG 0021 | 91.7                                | 0.7                                     | 96.1                                | 1.7                                     | 4.8         | 2.39   |
| BG 0022 | 90.1                                | 0.7                                     | 88.9                                | 2.0                                     | - 1.3       | 0.56   |
| BG 0023 | 90.4                                | 0.7                                     | 90.6                                | 6.0                                     | 0.2         | 0.03   |
| BG 0024 | 91.6                                | 0.7                                     | 97.0                                | 1.5                                     | 5.9         | 3.26   |
| BG 0025 | 102.2                               | 0.8                                     | 109.8                               | 1.5                                     | 7.4         | 4.48   |
| BG 0026 | 90.1                                | 0.7                                     | 90.0                                | 2.0                                     | - 0.1       | 0.06   |
| BG 0027 | 92.1                                | 0.7                                     | 92.0                                | 2.0                                     | - 0.1       | 0.04   |
| BG 0028 | 90.8                                | 0.7                                     | 91.1                                | 1.8                                     | 0.4         | 0.18   |
| BG 0031 | 89.3                                | 0.7                                     | 86.8                                | 0.5                                     | - 2.8       | 2.95   |
| BG 0032 | 92.2                                | 0.7                                     | 87.4                                | 0.9                                     | - 5.2       | 4.26   |
| BG 0033 | 110.1                               | 0.8                                     | 106.7                               | 2.1                                     | - 3.1       | 1.52   |
| BG 0034 | 104.8                               | 0.8                                     | 102.8                               | 3.4                                     | - 1.9       | 0.58   |
| BG 0035 | 111.2                               | 0.8                                     | 115.0                               | 10.0                                    | 3.4         | 0.38   |
| BG 0036 | 80.4                                | 0.6                                     | 80.0                                | 1.2                                     | - 0.5       | 0.27   |
| BG 0038 | 115.9                               | 0.9                                     | 119.0                               | 2.0                                     | 2.7         | 1.41   |
| BG 0039 | 88.0                                | 0.7                                     | 80.1                                | 2.1                                     | - 9.0       | 3.66   |
| BG 0040 | 93.7                                | 0.7                                     | 96.0                                | 2.8                                     | 2.5         | 0.79   |

Table 15  $^{144}\text{Ce}$  results

| Sample  | NPL Data                            |                                         | Analyst Data                        |                                         | Deviation % | u-test |
|---------|-------------------------------------|-----------------------------------------|-------------------------------------|-----------------------------------------|-------------|--------|
|         | $^{144}\text{Ce}/\text{Bq kg}^{-1}$ | $\pm ^{144}\text{Ce}/\text{Bq kg}^{-1}$ | $^{144}\text{Ce}/\text{Bq kg}^{-1}$ | $\pm ^{144}\text{Ce}/\text{Bq kg}^{-1}$ |             |        |
| BG 0001 | 64.3                                | 0.6                                     | 69.3                                | 3.9                                     | 7.8         | 1.27   |
| BG 0002 | 57.7                                | 0.5                                     | 62.2                                | 6.2                                     | 7.8         | 0.72   |
| BG 0004 | 60.0                                | 0.5                                     | 69.5                                | 3.9                                     | 15.9        | 2.43   |
| BG 0005 | 60.3                                | 0.5                                     | 69.0                                | 5.0                                     | 14.4        | 1.72   |
| BG 0006 | 59.1                                | 0.5                                     | 65.2                                | 4.1                                     | 10.4        | 1.48   |
| BG 0007 | 58.5                                | 0.5                                     | 55.0                                | 5.9                                     | - 6.0       | 0.60   |
| BG 0008 | 59.3                                | 0.5                                     | 58.8                                | 1.6                                     | - 0.8       | 0.30   |
| BG 0009 | 59.0                                | 0.5                                     | 61.7                                | 1.6                                     | 4.6         | 1.61   |
| BG 0010 | 59.6                                | 0.5                                     | 63.3                                | 9.0                                     | 6.3         | 0.42   |
| BG 0011 | 60.1                                | 0.5                                     | 64.9                                | 3.9                                     | 8.0         | 1.22   |
| BG 0013 | 59.4                                | 0.5                                     | 65.2                                | 5.6                                     | 9.8         | 1.04   |
| BG 0014 | 59.2                                | 0.5                                     | 55.0                                | 6.3                                     | - 7.0       | 0.66   |
| BG 0015 | 55.5                                | 0.5                                     | 57.8                                | 1.9                                     | 4.2         | 1.16   |
| BG 0016 | 58.9                                | 0.5                                     | 60.2                                | 2.1                                     | 2.1         | 0.58   |
| BG 0017 | 59.7                                | 0.5                                     | 60.2                                | 8.0                                     | 0.8         | 0.06   |
| BG 0019 | 59.4                                | 0.5                                     | 69.9                                | 4.0                                     | 17.8        | 2.62   |
| BG 0020 | 58.8                                | 0.5                                     | 61.0                                | 5.0                                     | 3.7         | 0.43   |
| BG 0021 | 58.5                                | 0.5                                     | 67.0                                | 4.5                                     | 14.6        | 1.88   |
| BG 0022 | 57.4                                | 0.5                                     | 64.3                                | 8.5                                     | 12.0        | 0.81   |
| BG 0023 | 57.6                                | 0.5                                     | 63.0                                | 6.7                                     | 9.3         | 0.80   |
| BG 0024 | 58.4                                | 0.5                                     | 69.8                                | 3.0                                     | 19.5        | 3.74   |
| BG 0025 | 65.2                                | 0.6                                     | 76.2                                | 4.5                                     | 16.9        | 2.43   |
| BG 0026 | 57.5                                | 0.5                                     | 60.8                                | 4.0                                     | 5.8         | 0.83   |
| BG 0027 | 58.7                                | 0.5                                     | 67.0                                | 8.0                                     | 14.2        | 1.04   |
| BG 0028 | 57.9                                | 0.5                                     | 62.5                                | 5.0                                     | 8.0         | 0.92   |
| BG 0031 | 56.9                                | 0.5                                     | 69.7                                | 4.7                                     | 22.5        | 2.71   |
| BG 0032 | 58.8                                | 0.5                                     | 61.7                                | 3.6                                     | 4.9         | 0.80   |
| BG 0033 | 70.2                                | 0.6                                     | 70.9                                | 3.0                                     | 1.0         | 0.22   |
| BG 0034 | 66.8                                | 0.6                                     | 71.6                                | 6.1                                     | 7.1         | 0.78   |
| BG 0035 | 70.9                                | 0.6                                     | 80.0                                | 10.0                                    | 12.9        | 0.91   |
| BG 0036 | 51.2                                | 0.5                                     | 52.6                                | 1.6                                     | 2.7         | 0.81   |
| BG 0038 | 73.9                                | 0.7                                     | 68.0                                | 12.0                                    | - 8.0       | 0.49   |
| BG 0039 | 56.1                                | 0.5                                     | 52.3                                | 5.5                                     | - 6.8       | 0.69   |
| BG 0040 | 59.7                                | 0.5                                     | 58.8                                | 8.2                                     | - 1.6       | 0.11   |

Table 16  $^{154}\text{Eu}$  results

| Sample  | NPL Data                            |                                         | Analyst Data                        |                                         | Deviation % | u-test |
|---------|-------------------------------------|-----------------------------------------|-------------------------------------|-----------------------------------------|-------------|--------|
|         | $^{154}\text{Eu}/\text{Bq kg}^{-1}$ | $\pm ^{154}\text{Eu}/\text{Bq kg}^{-1}$ | $^{154}\text{Eu}/\text{Bq kg}^{-1}$ | $\pm ^{154}\text{Eu}/\text{Bq kg}^{-1}$ |             |        |
| BG 0002 | 117.7                               | 1.0                                     | 111.5                               | 9.0                                     | - 5.3       | 0.69   |
| BG 0004 | 122.3                               | 1.0                                     | 110.0                               | 1.2                                     | - 10.1      | 7.92   |
| BG 0005 | 123.1                               | 1.0                                     | 116.0                               | 4.0                                     | - 5.8       | 1.72   |
| BG 0006 | 120.6                               | 1.0                                     | 112.1                               | 2.6                                     | - 7.1       | 3.12   |
| BG 0007 | 119.4                               | 1.0                                     | 113.0                               | 8.9                                     | - 5.3       | 0.71   |
| BG 0008 | 121.0                               | 1.0                                     | 107.0                               | 1.2                                     | - 11.6      | 9.04   |
| BG 0009 | 120.4                               | 1.0                                     | 113.5                               | 0.7                                     | - 5.7       | 5.62   |
| BG 0010 | 121.5                               | 1.0                                     | 111.3                               | 3.1                                     | - 8.4       | 3.14   |
| BG 0011 | 122.6                               | 1.0                                     | 117.0                               | 10.6                                    | - 4.6       | 0.53   |
| BG 0013 | 121.2                               | 1.0                                     | 117.0                               | 5.7                                     | - 3.4       | 0.72   |
| BG 0014 | 120.7                               | 1.0                                     | 116.8                               | 3.0                                     | - 3.3       | 1.24   |
| BG 0015 | 113.3                               | 0.9                                     | 106.2                               | 0.7                                     | - 6.2       | 6.25   |
| BG 0016 | 120.3                               | 1.0                                     | 111.5                               | 1.2                                     | - 7.3       | 5.79   |
| BG 0017 | 121.8                               | 1.0                                     | 113.5                               | 1.6                                     | - 6.8       | 4.43   |
| BG 0019 | 121.1                               | 1.0                                     | 120.0                               | 3.9                                     | - 0.9       | 0.28   |
| BG 0020 | 120.0                               | 1.0                                     | 115.0                               | 7.0                                     | - 4.2       | 0.71   |
| BG 0021 | 119.3                               | 1.0                                     | 111.0                               | 4.0                                     | - 7.0       | 2.02   |
| BG 0022 | 117.2                               | 0.9                                     | 98.6                                | 1.8                                     | - 15.9      | 9.13   |
| BG 0023 | 117.6                               | 1.0                                     | 105.1                               | 6.9                                     | - 10.6      | 1.79   |
| BG 0024 | 119.2                               | 1.0                                     | 105.8                               | 3.0                                     | - 11.2      | 4.25   |
| BG 0025 | 133.0                               | 1.1                                     | 131.4                               | 3.0                                     | - 1.2       | 0.51   |
| BG 0027 | 119.8                               | 1.0                                     | 106.0                               | 3.0                                     | - 11.5      | 4.37   |
| BG 0028 | 118.1                               | 1.0                                     | 150.0                               | 13.0                                    | 27.0        | 2.45   |
| BG 0031 | 116.1                               | 0.9                                     | 119.3                               | 1.3                                     | 2.7         | 1.98   |
| BG 0032 | 120.0                               | 1.0                                     | 112.0                               | 3.0                                     | - 6.7       | 2.53   |
| BG 0033 | 143.3                               | 1.2                                     | 135.0                               | 3.8                                     | - 5.8       | 2.08   |
| BG 0034 | 136.4                               | 1.1                                     | 124.5                               | 4.3                                     | - 8.7       | 2.67   |
| BG 0035 | 144.6                               | 1.2                                     | 133.0                               | 10.0                                    | - 8.1       | 1.16   |
| BG 0036 | 104.5                               | 0.8                                     | 86.9                                | 0.8                                     | - 16.9      | 15.31  |
| BG 0038 | 150.8                               | 1.2                                     | 138.0                               | 3.0                                     | - 8.5       | 3.95   |
| BG 0039 | 114.5                               | 0.9                                     | 94.4                                | 1.8                                     | - 17.6      | 10.06  |
| BG 0040 | 121.9                               | 1.0                                     | 119.1                               | 6.5                                     | - 2.2       | 0.41   |

Figure 1. Distribution of  $^{239}\text{Pu}$  resultsFigure 2. Distribution of  $^{241}\text{Am}$  resultsFigure 3. Distribution of  $^3\text{H}$  results

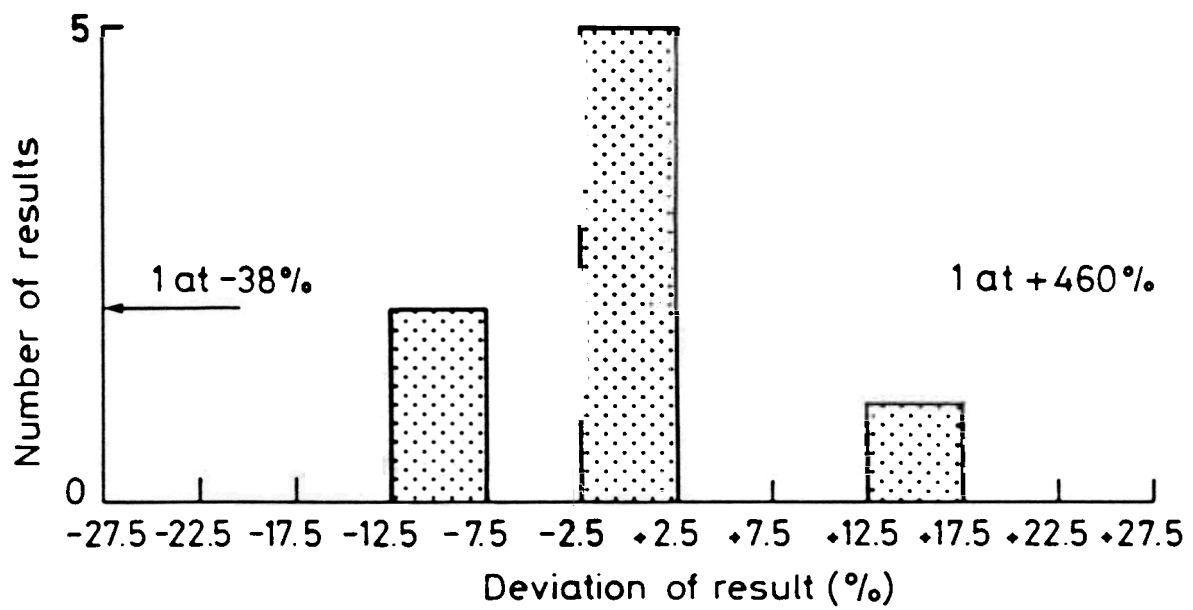


Figure 4. Distribution of  $^{90}\text{Sr}$  results

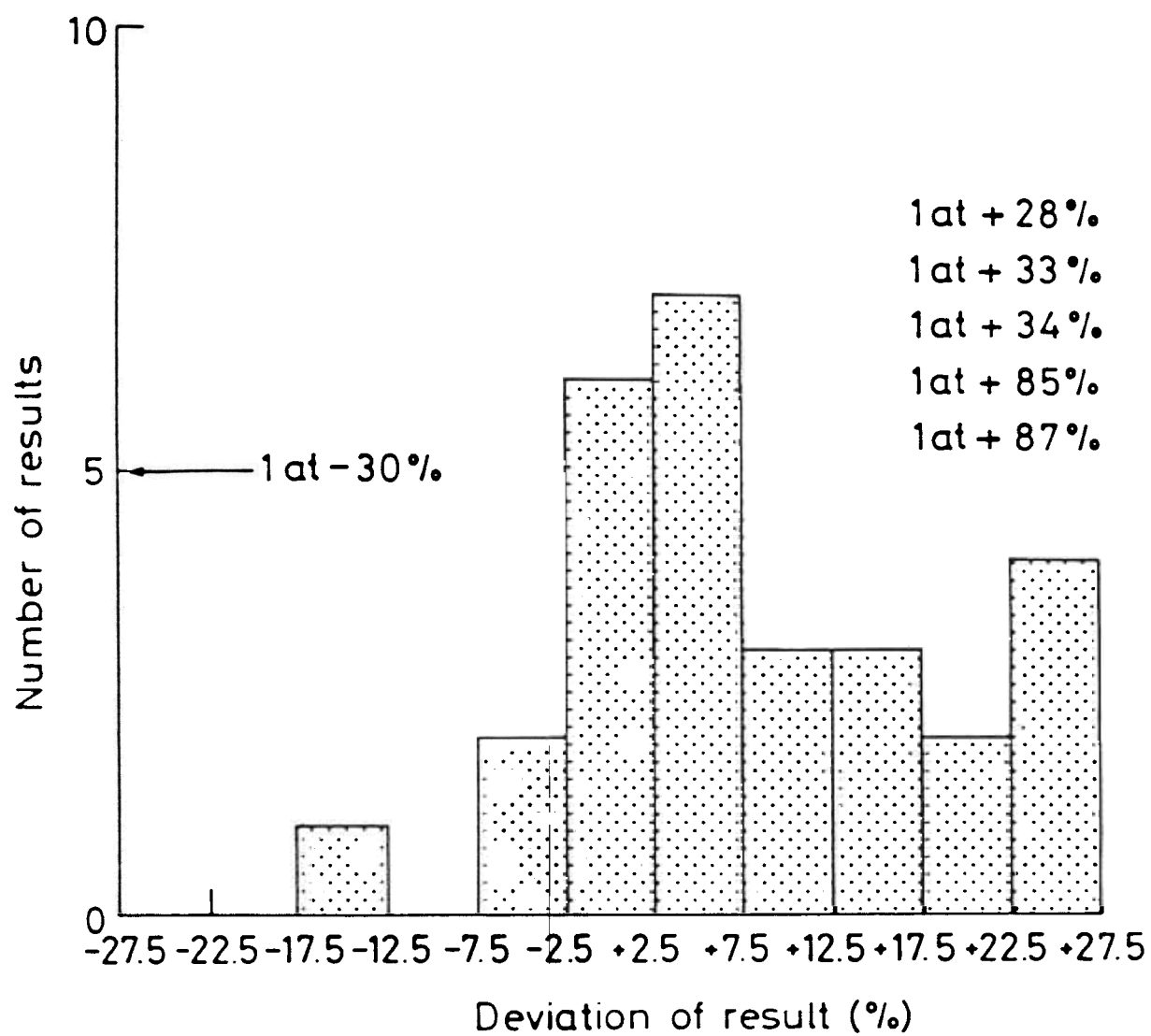


Figure 6. Distribution of  $^{95}\text{Zr}$  results

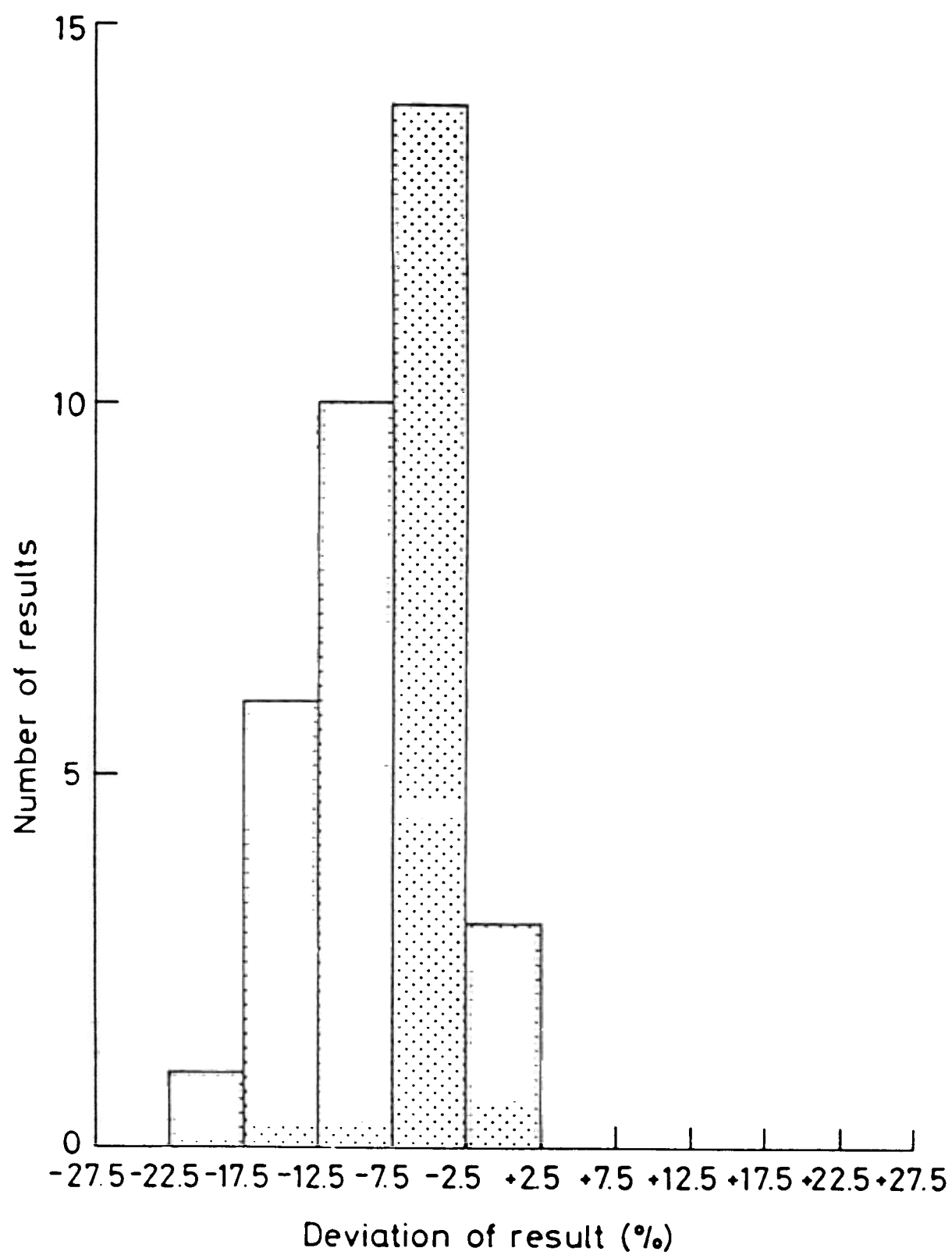


Figure 7. Distribution of  $^{134}\text{Cs}$  results

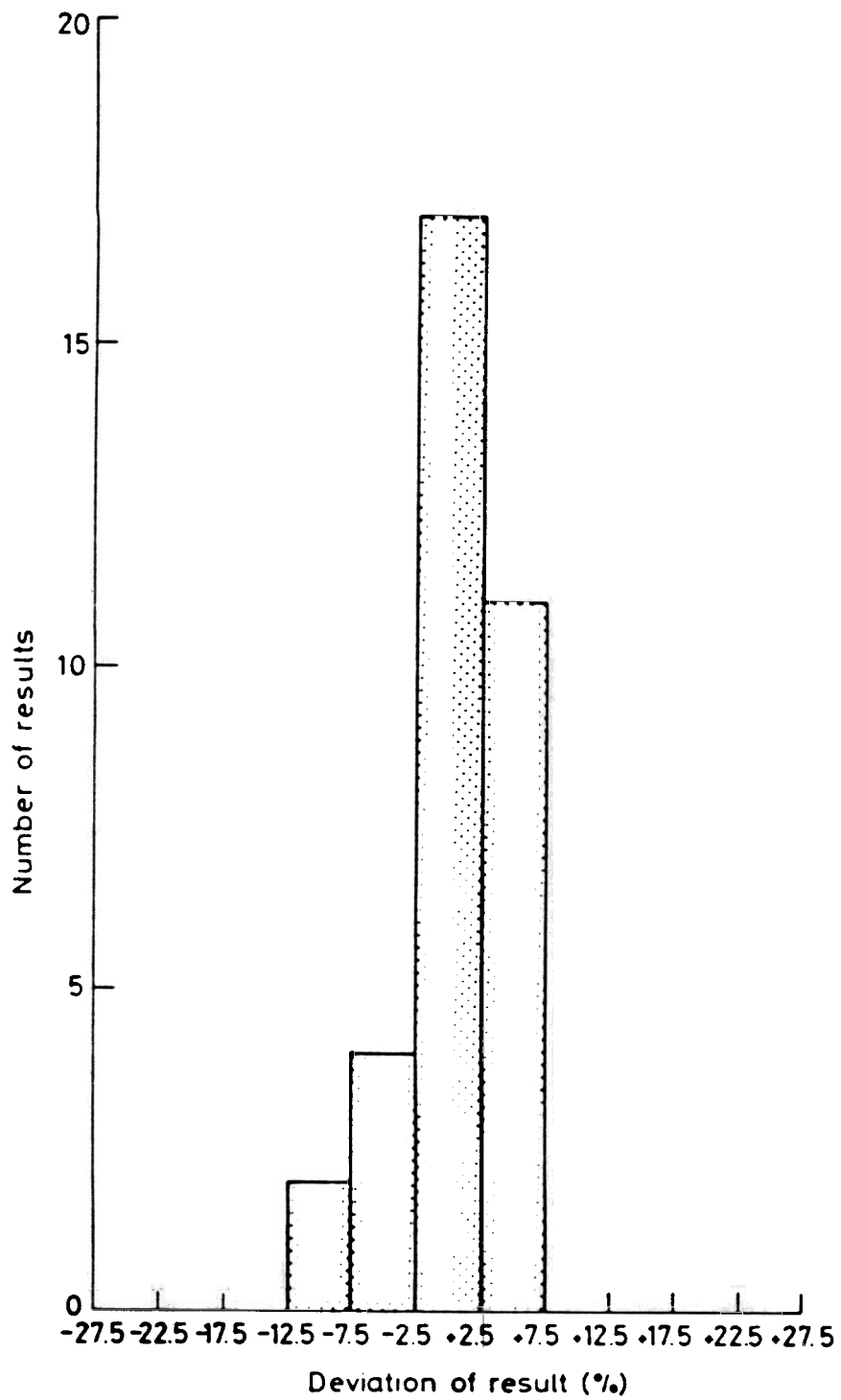


Figure 8 Distribution of  $^{137}\text{Cs}$  results



Figure 10. Distribution of  $^{154}\text{Eu}$  results

## APPENDIX

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