

NPL Nuclear Industry Proficiency Test Exercise 2011

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ABSTRACT

A third Nuclear Industry Proficiency Test Exercise (formerly known as 'drum comparisons') has been run by NPL. One sample was prepared, consisting of a nominal 200 L drum loaded with steel and Nylon sheets and 'bubble wrap'. The steel sheets were spiked with known weights of standard solutions of ^{241}Am , ^{60}Co and ^{137}Cs . The activity concentrations (averaged across the drum contents) were approximately 5.7 Bq g^{-1} , 2.3 Bq g^{-1} and 2.1 Bq g^{-1} respectively.

The participants were required to report the activity concentrations of the individual radionuclides. Information disclosed to the participants initially included the radionuclides present, a range for the total activity concentration, details of the empty drum (e.g. its mass and dimensions) and the percentage by mass of each material type present. The drum was measured by 16 UK and 4 overseas participants. After the initial results deadline, the 'internal structure' of the drum, more details of the materials present (e.g. which types of plastic) and the locations of the activity were disclosed. Participants were then invited to submit additional data. The exercise was followed by a workshop for discussion of the results.

Approximately 63% of the initial results submitted were 'in agreement' with the NPL value. Following disclosure of the additional information on the drum's contents, this figure increased to 73%, albeit from a subset of the participants who had submitted data initially (the figure for the initial results submitted by the subset was 71%). Different approaches to efficiency modelling (even using the same software) yielded varying results, some in agreement with NPL and others not. This emphasises the importance of accurate input data for efficiency modelling. As in previous exercises in this series, participants using Segmented Gamma Scanners submitted some discrepant data, possibly due to the small size of the active areas within the drum.

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CONTENTS

1	INTRODUCTION.....	1
2	BACKGROUND	1
	2.1 PREVIOUS EXERCISES	1
	2.2 PLAN FOR 2011 EXERCISE	2
3	PRIMARY AND SECONDARY STANDARDISATION	3
4	PREPARATION OF STANDARD DRUM	3
5	CIRCULATION OF DRUM AND REPORTING OF DATA.....	4
6	RESULTS AND DATA ANALYSIS	4
7	DISCUSSION	7
	7.1 AMERICIUM-241	7
	7.2 COBALT-60	8
	7.3 CAESIUM-137	8
8	CONCLUSIONS	8
9	ACKNOWLEDGEMENTS	9
10	REFERENCES.....	9
11	TABLES.....	10
12	FIGURES.....	18
13	APPENDIX A – INITIAL MAILSHOT	51
14	APPENDIX B – REPORTING FORM.....	54
15	APPENDIX C – LIST OF PARTICIPANTS	58
16	APPENDIX D – MINUTES OF POST-EXERCISE WORKSHOP, 26 OCTOBER 2011	61
17	APPENDIX E – PARTICIPANTS’ PRESENTATIONS FROM WORKSHOP	63

1 INTRODUCTION

There is an ongoing need for the accurate measurement of radioactivity in large volumes of potentially active materials (e.g. building materials, laboratory wastes, etc.) being produced in the context of nuclear site decommissioning. Such measurements are essential for correct waste categorisation, which is in turn necessary for public safety, minimisation of disposal costs and minimisation of volumes of Low Level Waste in accordance with UK LLW policy. To support this need, the National Physical Laboratory (NPL) has already run two ‘drum comparison’ exercises [Dean, 2007, 2010] to enable laboratories involved in the clearance and sentencing of bulk γ -emitting waste to test their existing measurement procedures. These exercises have proved very useful to the participating laboratories, both for validating the modelling techniques used for calculating detection efficiencies and for enabling participating laboratories to demonstrate their measurement capabilities to third parties.

These exercises were based on ‘standard drums’ prepared at NPL, containing waste simulants (e.g. ion exchange resins and vermiculite) loaded with traceable quantities of radioactivity. The participants suggested that future exercises should include samples of ‘real’ waste, to provide a more realistic test of measurement capability. A third exercise was therefore run in 2011, this time based on a standard drum containing materials that might be encountered in actual waste. This report covers:

- The background to the exercise;
- Briefly, the methods used to standardize the radioactive starting solutions;
- The preparation and certification of the ‘standard drum’;
- The conduct of the exercise;
- The results obtained and how they were analysed;
- Discussion and conclusions.

2 BACKGROUND

2.1 PREVIOUS EXERCISES

Prior to the current (2011) exercise, NPL had run Nuclear Industry Proficiency Test Exercises (known as ‘drum comparisons’) in 2007 and 2009 [Dean, 2007, 2010]. These exercises were run in response to needs for standards and reference materials for nuclear decommissioning and site clearance identified at an NPL workshop in 2005. The overall plan for each exercise was:

- Production and certification of a 200 L drum (or drums) containing a low-density matrix spiked with γ -emitters;
- Circulation to participating laboratories;
- Reporting of results;
- Data analysis at NPL;
- A follow-up workshop to discuss the results.

For each exercise, one or two ‘standard drums’ were prepared by filling (for each drum) approximately 240 plastic bottles (volume 500ml) with ion-exchange resin or vermiculite, spiking one or more of the bottles with a standard solution of the radionuclides of interest (^{241}Am , ^{60}Co and ^{137}Cs) and then loading the bottles into a 200 L steel drum. The approximate activity concentrations in the drums (i.e. total activity / total mass of drum contents) were:

- 2007: 0.3 Bq g⁻¹
- 2009 (Drum A): 0.3 Bq g⁻¹
- 2009 (Drum B): 9 Bq g⁻¹

These concentrations were chosen so that the drums' contents would be similar in activity concentration to 'RSA Exempt' waste and LLW respectively. In the 2007 exercise, the activity was distributed in a heterogeneous manner throughout the drum whereas in 2009 the activity in both drums was concentrated in a 'hot spot' (i.e. the activity distribution was heterogeneous). The participants were told the approximate activities present in the drums, and also information such as details of the matrix in the bottles.

Feedback from the 2009 exercise indicated that the exercises had been useful to the participants and that a further exercise should be run, this time using a drum with contents more like those of a 'real' sample. Suggestions included:

- A drum containing 'artefacts' (e.g. metal bars or tools);
- A 'standard canister' of activity for the participants to insert into their own 'regular' sample container;
- A drum containing uranium.

It would also be more 'realistic' if little or no information was provided on the contents of the drum. Typically, the only information provided with a real sample is the approximate percentage by mass of the matrices present.

A subsequent survey of users indicated that a 200L drum containing soft waste and/or metal and/or concrete at 1-10 Bq g⁻¹ would be preferred for a third exercise.

2.2 PLAN FOR 2011 EXERCISE

Based on the above feedback, it was decided to prepare a standard 200 L drum containing a mixture of plastic and thin metal sheets. This would keep the overall contents mass low and make it relatively easy to manufacture a 'drum insert' which would fit flush to the drum walls and be dimensionally stable in transit (thus ensuring that the drum's 'internal structure' and activity distribution would not change). The nuclides would again be ²⁴¹Am, ⁶⁰Co and ¹³⁷Cs and the overall activity concentration would be 1-10 Bq g⁻¹. However, to make the sample more realistic, the activity would be added as 'surface contamination' of the metal rather than as a spiked sample of resin or similar.

The overall plan would then follow the stages in 2.1 above but the only information supplied to the participants prior to measurement would be a generic description of the matrices present (e.g. 'metal', 'soft waste') and the percentage of each matrix present by mass, along with details of the empty drum (e.g. wall thickness).

As in the 2009 exercise, there would be two deadlines for the submission of results. The first deadline would be for results where the participants had only the information given in the previous paragraph. After this deadline, NPL would disclose details of the 'drum insert' and the location of the activity and invite additional results before the second deadline. This would enable participants to submit results for two different measurement scenarios. The two datasets would be treated separately and no changes to results submitted before the first deadline would be accepted after that deadline.

3 PRIMARY AND SECONDARY STANDARDISATION

NPL, as the UK's National Measurement Institute, carries out metrology-related research and provides measurement standards, reference materials, calibration services and support to users (e.g. via guidance documents, training, comparison exercises and workshops) across some 20 generic areas of science and engineering. At the international level, NPL forms part of the international measurement system, being one of many National Measurement Institutes (NMIs) worldwide who work together under the auspices of organisations such as the BIPM (Bureau International des Poids et Mesures) to share resources and to harmonise standards.

The Radioactivity Group holds NPL's expertise in radioactivity measurement, and amongst its many activities develops and maintains primary and secondary measurement systems. Some of these systems were used in the preparation and certification of the standard drum for this exercise. This ensured that the source would be directly traceable to national standards of radioactivity.

The activity concentrations of the solutions used in the preparation of the standard drum were measured using a re-entrant secondary standard ionisation chamber. The radionuclidic purity of these solutions was checked using high-resolution γ -spectrometry. Both instruments have been calibrated using solutions determined by absolute counting techniques using instruments maintained at NPL.

4 PREPARATION OF STANDARD DRUM

The following materials were procured:

- Standardised radionuclide solutions of ^{241}Am , ^{60}Co and ^{137}Cs (activity concentrations in the range 100 - 300 kBq g⁻¹);
- 3 x filter papers (80 mm x 80 mm);
- 1 x 200 L steel drum;
- 1 x roll of 'AirCap¹' plastic bubble film
- Approximately 12 kg of Nylon 6-6
- Approximately 5 kg of 0.9 mm BS 1449 stainless steel sheet.

The 'drum insert' was prepared at NPL and is illustrated in Figure 1. It consisted of a u-shaped frame on Nylon 6-6 with a sheet of stainless steel inserted in grooves on either side of the 'u'. This formed a box-like structure with the top of the 'box' open.

The activity was applied in the form of three 'spiked' filter papers taped to the outward-facing walls of the steel plates, as illustrated. These filter papers had been previously spiked (in a uniform manner) with weighed aliquots of the standard solutions of the radionuclides. The activities of these solutions has been previously determined by assay in glass ampoules in a re-entrant secondary standard ionisation chamber. Each ampoule had been measured using a γ -spectrometer to check for the presence of any γ -emitting impurities. The drum was certificated for the activity concentrations of each radionuclide present (see Table 1) and was designated source X11131.

¹ Sealed Air Limited, Kettering NN16 8UN, UK

5 CIRCULATION OF DRUM AND REPORTING OF DATA

A mailshot was prepared (see Appendix A) and circulated to around 440 contacts in the nuclear industry. The mailshot gave full details of the exercise. Key points made in the mailshot were:

- There would be one drum type available, with an activity concentration in the range 1 – 10 Bq g⁻¹
- The radionuclides present would be ²⁴¹Am, ⁶⁰Co and ¹³⁷Cs;
- The drums would contain a mixture of metal, hard plastic and soft-waste material;
- More details of the drum's contents would not be disclosed until after a 'first deadline' for submission of results. There would then be a 'second deadline', for submission of additional (but not replacement) results based on the additional information.

The drum was dispatched from NPL to one of the UK participants; thereafter, it was moved directly from one UK participant to the next according to a pre-determined timetable with NPL organising the courier service each time. Each participant was given three working days (including the delivery and pick-up days) in which to carry out their measurements. In one case, two participants at the same laboratory measured the drum within the three-day period. After most of the UK participants had measured the drum, it was returned to NPL and then dispatch to the overseas participants (and also two further UK participants).

A Reporting Form (see Appendix B) was sent to the participants for reporting their data. Also, the participants were provided with the following details:

- A supplier's 'Certificate of Packaging Performance' and other details of the 'empty' drum;
- The mass of the drum and its contents. These values were nominally:
 - Drum empty: 18.3 kg
 - Drum full: 35.4 kg
- The matrices present and the nominal percentage of each by mass. The values were:
 - 'Metal': 24.6%
 - 'Hard plastic': 68.6%
 - 'Soft-waste simulant': 6.8%

A total of 20 participants received the drum. This included four overseas participants (two in Belgium and one each in France and Italy). The participants are listed in Appendix C.

6 RESULTS AND DATA ANALYSIS

To preserve anonymity, each participant was assigned a code number, and their results were coded accordingly. Some of the laboratories had also participated in previous exercises and their codes were changed for the current exercise.

On receipt of the results, NPL carried out data analyses using the method described below [Harms, 2009(a)].

Firstly, the deviation from the assigned (NPL) value of each laboratory value was calculated, given by:

$$D = 100 \frac{L - N}{N} = 100 \left(\frac{L}{N} - 1 \right) \quad [1]$$

where:

D = deviation from the NPL value (%)

L = the participant's value (Bq g⁻¹)

N = the NPL value (Bq g⁻¹)

The deviations are given in Tables 2 - 7 and are plotted in Figures 2, 6, 10, 14, 18 and 22. They are also plotted 'by participant' in Figures 26 – 41. The figure titles indicate whether the results were submitted before or after disclosure of full details of the drum's contents. The error bars in the graphs represent the standard uncertainty ($k=1$) of the deviation:

$$u_D = 100 \frac{L}{N} \sqrt{\left(\frac{u_L}{L}\right)^2 + \left(\frac{u_N}{N}\right)^2} \quad [2]$$

where:

u_D = standard uncertainty of the deviation (%)

u_L = standard uncertainty of the participant's value (Bq g⁻¹)

u_N = standard uncertainty of the assigned value (Bq g⁻¹)

The results were evaluated using three tests:

$$\zeta = \frac{L - N}{\sqrt{u_L^2 + u_N^2}} \quad [3]$$

$$R_L = \frac{u_L}{L} \quad [4]$$

$$z = \frac{L - N}{R_{\text{med}} N} \quad [5]$$

where:

ζ = zeta score

R_L = relative uncertainty of the participant's value

z = z-score

R_{med} = median of the values of R_L

The zeta and z-scores were used to determine whether the difference between the participant's value and the NPL value was significantly different from zero. An IQR (Inter-Quartile Range) outlier test was used to determine whether a particular R_L value was significantly larger than the other values in a data set; the IQR was used to calculate a limiting value of R_L , ' R_{lim} ', for each data set. The zeta scores, z-scores and outlier results are also given in Tables 2 - 7. The zeta scores are plotted in Figures 3, 7, 11, 15, 19 and 23. The R_L values are plotted in Figures 4, 8, 12, 16, 20 and 24.

Results for which the absolute values of the zeta score and the z-score were both ≤ 2.576 (corresponding to a significance levels of $\alpha = 0.01$) and for which the relative uncertainty R_L was not significantly larger than the other values in the data set were regarded as being 'in agreement' with NPL. These are marked in dark blue in the deviation plots. If either (i) the relative uncertainty R_L was significantly larger than the other values in the data set, (ii) the result passes the zeta test but not the z-

test (i.e. large deviation from the NPL value combined with a large uncertainty), or (iii) the result passes the z-test but not the zeta test (small deviation from the NPL value combined with a small uncertainty), the participant's value is classified as 'questionable' (these are given in yellow in the deviation plots). If the absolute values of both the zeta score and the z-score are > 2.576 , then the participant's value is classified as 'discrepant' from the NPL value (red points), regardless of the value of the relative uncertainty R_L . The findings for all the data are also given in Tables 2 - 7.

Now, the zeta score and the z-score are related by:

$$\zeta = \frac{\sigma_p}{\sqrt{u_L^2 + u_N^2}} z \quad [6]$$

(where $\sigma_p = R_{\text{med}}N$, the standard uncertainty for proficiency assessment (Bq g^{-1})).

This can be rewritten as:

$$\frac{z^2}{\zeta^2} - \frac{u_N^2}{\sigma_p^2} = \frac{u_L^2}{\sigma_p^2} \quad [7]$$

The relative uncertainty of the laboratory R_L and the z-score are related by:

$$\frac{u_L}{R_L} = z\sigma_p + N \quad [8]$$

This can be rewritten as:

$$R_L^2 \left\{ z + \frac{N}{\sigma_p} \right\}^2 = \frac{u_L^2}{\sigma_p^2} \quad [9]$$

Consequently, comparison data can be neatly summarised by plotting the square of the ratio of u_L to σ_p against the z-score, and superimposing one parabola to represent a zeta score of 2.576, a second parabola representing the outlier limit R_{lim} of the relative laboratory uncertainty R_L , and vertical lines representing a z-score of 2.576. These are known as 'Kiri' plots [Harms, 2009(b)], and are given in Figures 5, 9, 13, 17, 21 and 25. Data points that are inside the zeta parabola (i.e., for which $\text{zeta} \leq 2.576$), within the z-score lines and outside the R_{lim} parabola (i.e., for which $R_L \leq R_{\text{lim}}$) are 'in agreement' with NPL. Data points which fail either the z-test, the zeta test or the relative uncertainty outlier test (but not both the z-test and zeta test), are classified as 'questionable' and are either:

- (i) inside the zeta parabola with a z-score < -2.576 or > 2.576 , or
- (ii) outside the zeta parabola with a z-score > -2.576 or < 2.576 ; or
- (iii) inside zeta parabola with a z-score > -2.576 or < 2.576 , but inside the R_{lim} parabola.

All other data points are classified as 'discrepant'.

7 DISCUSSION

n.b. As stated in Appendix A, NPL held a follow-up workshop for the PTE. This discussion takes into account comments and suggestions made at the workshop. The minutes of the workshop are given in Appendix D and the presentations given at the workshop are in Appendix E.

A total of 19 laboratories received the drum, and at one laboratory the drum was measured by two participants. Two laboratories did not submit results, therefore 18 participants in all submitted data. The data submitted by Participants 1 – 16 are given in Tables 2 – 7. Participant 17 did not report measurement uncertainties and Participant 18 did not submit results within the required timescales. The results from Participants 17 and 18 are given in Table 8.

Five participants (Participants 9, 11, 13, 14 and 16) reported additional data after disclosure of details of the drum's contents. (n.b. For convenience, the terms 'pre-disclosure' and 'post-disclosure' are used in the discussion and conclusions to differentiate between results submitted before and after disclosure of the details of the drum's contents.)

Two participants each reported results for more than one detector and one participant reported more than one result for one detector. All participants used γ -spectrometry. Six used collimated hyperpure Ge crystals, three used Segmented Gamma Scanners and a further two used three-crystal Ge detector systems. Source-to-detector distances varied from 10 cm to 185 cm. Count times varied widely, from 600 s to approximately 63000 s.

A variety of detector efficiency models and calibration standards was used. Six participants used ISOCS, three used ISOTOPIC, two used NDA 2000, one used SNAP, one used Winner/Track software and one participant used a 'Particle Swarm Imaging' model (PSIM). Seven participants derived calibrations using standard drums prepared 'in house' or using point sources from various suppliers (e.g. NIST and IPL).

7.1 AMERICIUM-241

Measurement of the ^{241}Am component is difficult because the attenuation within the drum wall is much greater than for the other radionuclides present. Small differences between the actual composition of the drums and their composition assumed in efficiency models will also be much more important.

'Pre-disclosure', 19 results were submitted for this radionuclide, including two results from Participant 6 (using two different detectors), three results from Participant 8 (different count-times) and three results from Participant 9 (different detectors). Three results were 'discrepant' (3, 13 and 14), three results were 'questionable' (4, 6B and 16) and the remainder were 'in agreement' with NPL. Participant 3 calibrated their system with 'standard drums' containing sources which simulate a homogeneous activity distribution, but corrections for heterogeneous matrix or source distribution could not be made at the time of measurement. Also, their quoted uncertainty was for counting statistics only. The classification of Participant 4's result may have been due to the low uncertainty quoted. Participant 6 (result 6B) used a Segmented Gamma Scanner (SGS) and obtained a low result, so possibly the emissions from the ^{241}Am filter was 'split' between two adjacent segments, with one segment containing a level of activity below the limit of detection.

Fewer results were submitted 'post-disclosure', indeed so few that it was not possible to carry out an outlier test. However, the results for Participants 13 and 14 were clearly much closer to the NPL value, and in agreement with that value. Also, Participant 4 was now in agreement. Curiously, the results for Participant 9 were lower than pre-disclosure, this participant having changed their calibration to a ISOCS 'complex box' model; however, they were still in agreement with NPL.

7.2 COBALT-60

Pre-disclosure, 25 results were submitted for ^{60}Co . Again, this included multiple data submissions. Participant 6 again submitted data for two different detectors. Participant 7 submitted a value for each of the two main γ -emissions from this radionuclide; Participant 8 did likewise, but for three different count-times (therefore submitting six results in all), and again Participant 9 submitted one result for each of their three detectors. Overall, four results were discrepant (2, 3, 4 and 10), eight were questionable (7A, 7B, 8A, 8B, 8C, 8D, 8E and 8F) and the remainder agreed with NPL.

Participant 2 submitted a low result using an SGS system, and again possibly this was due to the ^{60}Co emissions being split between adjacent segments of view, although Participant 10 also used an SGS and achieved a high result. It is curious that the results from Participants 7 and 8 were all questionable and were all obtained using an ISOCS ‘complex cylinder’ template. They both assumed the drum contained carbon steel, high-density polyethylene and cellulose. However, their data for the other two nuclides present were in agreement with NPL.

With the exception of Participant 11, the same group of participants submitted data post-disclosure as for americium-241, and again an outlier test was not possible due to the small data set. The data were very similar to the pre-disclosure values.

7.3 CAESIUM-137

Pre-disclosure, 21 results were submitted for this radionuclide. Participants 6, 8 and 9 once again submitted multiple results, as described in 7.1 above. The overall level of agreement with NPL for the participants as a whole was very good, with only 2 discrepant (2 and 6A) and 3 questionable (3, 4 and 6B) results being submitted.

The same group of participants as for cobalt-60 submitted data after disclosure of the drum’s internal contents. Again, the data were very similar to the pre-disclosure values. Again, Participant 4’s result was now in agreement with NPL.

8 CONCLUSIONS

Overall, 63% of the data submitted prior to disclosure of the drum’s internal structure were in agreement with the NPL assigned values, whereas 73 % of the data submitted after disclosure were in agreement with NPL. This is logical given that more information on the drum was available. It should be noted that the post-disclosure data came from a subset of the participants who had submitted data pre-disclosure; 71 % of the subset’s pre-disclosure data was in agreement with NPL, so there was still a slight improvement in the overall quality of the data even if one considers the data from just the subset.

Some participants who had used Segmented Gamma Scanners obtained discrepant data. These deviations may have been due to the small active areas within the drum and the possibility of one segment containing γ -emissions below the limit of detection. It is interesting to compare the (pre-disclosure) results from using SGSs with results from ‘open geometry’ measurements (see Table 9). Although there appears to be no significant difference between the SGS and open geometry means, the SGS mean is lower for all three radionuclides. Although these data sets are small and there are many other factors affecting the measurements, similar effects were observed in previous exercises [Dean, 2007, 2010]. This should be investigated further.

A key finding of the exercise (as in previous exercises) was that the participants who had used ISOCS had obtained differing results (noticeably for ^{241}Am and ^{60}Co), some in agreement with NPL and

others not. It may be significant that these participants had made various assumptions about the drums and their contents. These results once again underline the importance of accurate input data in obtaining accurate assay results with modelling software.

Also of interest is possible differences between results using different analysis software. For example, the means of the ISOCS results (pre-disclosure) appear higher than the means from using ISOTOPIC software (see Table 10). However, the data sets are again small and there are insufficient data from previous exercises to confirm this finding.

The specifications of the ‘standard drum’ (i.e. matrix, radionuclides present, etc.) had provided a useful test for the participants, but the next exercise should include something still closer to a ‘real’ sample, ideally containing some concrete, so often a component of field samples. This should not pose manual handling problems provided the sample mass is no more than around 100 kg. The activity concentration range should be kept at 1-10 Bq g⁻¹, although each participant should be allocated a sample count time of 2 days (excluding receipt and dispatch days).

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11 TABLES

Table 1 - Principal radionuclides in X11131

Radionuclide	Decay mode	Activity concentration (averaged across contents of drum) @ 1200 UTC* 01/03/11 (Bq g ⁻¹)	Standard uncertainty (<i>k</i> =1) (Bq g ⁻¹)	Standard uncertainty (<i>k</i> =1) (%)
²⁴¹ Am	α/γ	5.70	0.03	0.56
⁶⁰ Co	β^-/γ	2.293	0.011	0.47
¹³⁷ Cs	β^- , β^-/γ	2.053	0.016	0.81

* Universal Time, Co-ordinated. This replaced Greenwich Mean Time in 1972.

Table 2 - Reported results for ^{241}Am , pre-disclosureNPL activity concentration = $(5.70 \pm 0.03) \text{ Bq g}^{-1}$ ($k=1$)

Reference time = 1200 UTC 1 March 2011

Participant code	Reported ^{241}Am activity concentration (Bq g^{-1})	Reported uncertainty ($k=1$) (Bq g^{-1} , unless indicated)	Deviation from NPL value (%)	Z score	Zeta score	Result of outlier test	Conclusion
1	5.95	30%	4.39	0.28	0.14	Not outlier	In agreement
3	10.4	0.6	82.46	5.20	7.82	Not outlier	Discrepant
4	4.61	0.21	-19.12	-1.21	-5.14	Not outlier	Questionable
5	6.74	1.24	18.25	1.15	0.84	Not outlier	In agreement
6A	5.47	0.59	-4.04	-0.25	-0.39	Not outlier	In agreement
6B	3.58	0.2	-37.19	-2.35	-10.48	Not outlier	Questionable
7	6.29	3.16	10.35	0.65	0.19	Not outlier	In agreement
8A	7.02	1.57	23.16	1.46	0.84	Not outlier	In agreement
8B	6.38	1.43	11.93	0.75	0.48	Not outlier	In agreement
8C	6.58	1.38	15.44	0.97	0.64	Not outlier	In agreement
9A	5.99	1.200	5.09	0.32	0.24	Not outlier	In agreement
9B	7.045	1.413	23.60	1.49	0.95	Not outlier	In agreement
9C	6.42	1.29	12.63	0.80	0.56	Not outlier	In agreement
11	4.86	0.77	-14.74	-0.93	-1.09	Not outlier	In agreement
12	4.37	2.19	-23.33	-1.47	-0.61	Not outlier	In agreement
13	2.18	0.29	-61.75	-3.90	-12.07	Not outlier	Discrepant
14	3.35	0.24	-41.23	-2.60	-9.72	Not outlier	Discrepant
15	6.27	2.09	10.00	0.63	0.27	Not outlier	In agreement
16	8.79	1.21	54.21	3.42	2.55	Not outlier	Questionable

Table 3 - Reported results for ^{241}Am , post-disclosureNPL activity concentration = $(5.70 \pm 0.03) \text{ Bq g}^{-1}$ ($k=1$)

Reference time = 1200 UTC 1 March 2011

Participant code	Reported ^{241}Am activity concentration (Bq g^{-1})	Reported uncertainty ($k=1$) (Bq g^{-1} , unless indicated)	Deviation from NPL value (%)	Z score	Zeta score	Result of outlier test	Conclusion
4	4.69	0.51	-17.72	-1.42	-1.98	Not outlier	In agreement
9A	4.65	0.93	-18.42	-1.47	-1.13	Not outlier	In agreement
9B	4.58	0.92	-19.65	-1.57	-1.22	Not outlier	In agreement
9C	4.28	0.86	-24.91	-1.99	-1.65	Not outlier	In agreement
11	6.89	0.51	20.88	1.67	2.33	Not outlier	In agreement
13	5.24	0.74	-8.07	-0.65	-0.62	Not outlier	In agreement
14	6.16	0.540	8.07	0.65	0.85	Not outlier	In agreement
16	10.1	1.96	77.19	6.18	2.24	Not outlier	Questionable

Table 4 - Reported results for ^{60}Co , pre-disclosureNPL activity concentration = $(2.293 \pm 0.011) \text{ Bq g}^{-1}$ ($k=1$)

Reference time = 1200 UTC 1 March 2011

Participant code	Reported ^{60}Co activity concentration (Bq g^{-1})	Reported uncertainty ($k=1$) (Bq g^{-1} , unless indicated)	Deviation from NPL value (%)	Z score	Zeta score	Result of outlier test	Conclusion
1	2.6	20%	13.39	2.21	0.59	Not outlier	In agreement
2	1.26	0.02	-45.05	-7.44	-45.26	Not outlier	Discrepant
3	2.78	0.15	21.24	3.51	3.24	Not outlier	Discrepant
4	2.92	0.13	27.34	4.52	4.81	Not outlier	Discrepant
5	2.33	0.340	1.61	0.27	0.11	Not outlier	In agreement
6A	2.06	0.1	-10.16	-1.68	-2.32	Not outlier	In agreement
6B	2.33	0.01	1.61	0.27	2.49	Not outlier	In agreement
7A	2.9	0.9	26.47	4.37	0.67	Not outlier	Questionable
7B	3.0	0.9	30.83	5.09	0.79	Not outlier	Questionable
8A	2.95	0.38	28.65	4.73	1.73	Not outlier	Questionable
8B	2.97	0.37	29.52	4.88	1.83	Not outlier	Questionable
8C	2.88	0.34	25.60	4.23	1.73	Not outlier	Questionable
8D	2.90	0.37	26.47	4.37	1.64	Not outlier	Questionable
8E	3.01	0.37	31.27	5.16	1.94	Not outlier	Questionable
8F	2.85	0.34	24.29	4.01	1.64	Not outlier	Questionable
9A	2.280	0.322	-0.57	-0.09	-0.04	Not outlier	In agreement
9B	2.271	0.321	-0.96	-0.16	-0.07	Not outlier	In agreement
9C	2.21	0.32	-3.62	-0.60	-0.26	Not outlier	In agreement
10	3.44	0.13	50.02	8.26	8.79	Not outlier	Discrepant
11	2.44	0.11	6.41	1.06	1.33	Not outlier	In agreement
12	2.42	0.17	5.54	0.91	0.75	Not outlier	In agreement
13	2.45	0.24	6.85	1.13	0.65	Not outlier	In agreement
14	2.56	0.155	11.64	1.92	1.72	Not outlier	In agreement
15	2.18	0.48	-4.93	-0.81	-0.24	Not outlier	In agreement
16	2.50	0.132	9.03	1.49	1.56	Not outlier	In agreement

Table 5 - Reported results for ^{60}Co , post-disclosureNPL activity concentration = $(2.293 \pm 0.011) \text{ Bq g}^{-1}$ ($k=1$)

Reference time = 1200 UTC 1 March 2011

Participant code	Reported ^{60}Co activity concentration (Bq g^{-1})	Reported uncertainty ($k=1$) (Bq g^{-1} , unless indicated)	Deviation from NPL value (%)	Z score	Zeta score	Result of outlier test	Conclusion
4	2.84	0.03	23.86	4.31	17.12	Not outlier	Discrepant
9A	1.89	0.27	-17.58	-3.17	-1.49	Not outlier	Questionable
9B	2.1	0.29	-8.42	-1.52	-0.67	Not outlier	In agreement
9C	1.88	0.27	-18.01	-3.25	-1.53	Not outlier	Questionable
13	2.43	0.24	5.97	1.08	0.57	Not outlier	In agreement
14	2.04	0.113	-11.03	-1.99	-2.23	Not outlier	In agreement
16	2.65	0.137	15.57	2.81	2.60	Not outlier	Discrepant

Table 6 - Reported results for ^{137}Cs , pre-disclosureNPL activity concentration = $(2.053 \pm 0.016) \text{ Bq g}^{-1}$ ($k=1$)

Reference time = 1200 UTC 1 March 2011

Participant code	Reported ^{137}Cs activity concentration (Bq g^{-1})	Reported uncertainty ($k=1$) (Bq g^{-1} , unless indicated)	Deviation from NPL value (%)	Z score	Zeta score	Result of outlier test	Conclusion
1	2.2	20%	7.16	1.04	0.33	Not outlier	In agreement
2	1.21	0.04	-41.06	-5.95	-19.57	Not outlier	Discrepant
3	2.41	0.13	17.39	2.52	2.73	Not outlier	Questionable
4	2.44	0.2	18.85	2.73	1.93	Not outlier	Questionable
5	1.98	0.290	-3.56	-0.52	-0.25	Not outlier	In agreement
6A	3	0.18	46.13	6.69	5.24	Not outlier	Discrepant
6B	2.15	0.02	4.72	0.69	3.79	Not outlier	Questionable
7	2.3	0.8	12.03	1.74	0.31	Not outlier	In agreement
8A	2.31	0.33	12.52	1.82	0.78	Not outlier	In agreement
8B	2.34	0.33	13.98	2.03	0.87	Not outlier	In agreement
8C	2.19	0.29	6.67	0.97	0.47	Not outlier	In agreement
9A	1.95	0.390	-5.02	-0.73	-0.26	Not outlier	In agreement
9B	2.226	0.445	8.43	1.22	0.39	Not outlier	In agreement
9C	2.14	0.43	4.24	0.61	0.20	Not outlier	In agreement
10	1.82	0.12	-11.35	-1.65	-1.92	Not outlier	In agreement
11	2.08	0.12	1.32	0.19	0.22	Not outlier	In agreement
12	2.03	0.14	-1.12	-0.16	-0.16	Not outlier	In agreement
13	2.15	0.22	4.72	0.69	0.44	Not outlier	In agreement
14	2.28	0.11	11.06	1.60	2.04	Not outlier	In agreement
15	2.23	0.50	8.62	1.25	0.35	Not outlier	In agreement
16	2.22	0.151	8.13	1.18	1.10	Not outlier	In agreement

Table 7 - Reported results for ^{137}Cs , post-disclosureNPL activity concentration = $(2.053 \pm 0.016) \text{ Bq g}^{-1}$ ($k=1$)

Reference time = 1200 UTC 1 March 2011

Participant code	Reported ^{137}Cs activity concentration (Bq g^{-1})	Reported uncertainty ($k=1$) (Bq g^{-1} , unless indicated)	Deviation from NPL value (%)	Z score	Zeta score	Result of outlier test	Conclusion
4	2.19	0.06	6.67	0.95	2.21	Not outlier	In agreement
9A	1.78	0.34	-13.30	-1.90	-0.80	Not outlier	In agreement
9B	1.99	0.39	-3.07	-0.44	-0.16	Not outlier	In agreement
9C	1.67	0.34	-18.66	-2.66	-1.13	Not outlier	Questionable
13	2.28	0.24	11.06	1.58	0.94	Not outlier	In agreement
14	1.98	0.130	-3.56	-0.51	-0.56	Not outlier	In agreement
16	2.24	0.157	9.11	1.30	1.18	Not outlier	In agreement

Table 8 - Reported results from Participants 17 and 18

Reference time = 1200 UTC 1 March 2011

Participant code	Radionuclide	Reported activity concentration (Bq g ⁻¹)	Reported uncertainty (k=1) (%)
17	²⁴¹ Am	11.82	-
17	⁶⁰ Co	2.8	-
17	¹³⁷ Cs	2.66	-
18A	²⁴¹ Am	6.44	200.30
18A	⁶⁰ Co	2.63	30.8
18A	¹³⁷ Cs	2.06	40.6
18B	²⁴¹ Am	8.13	32.00
18B	⁶⁰ Co	2.48	26.00
18B	¹³⁷ Cs	2.19	26

Table 9 - Comparison of Segmented Gamma Scanner results with 'open geometry' results (pre-disclosure)

Radionuclide	SGS mean (Bq g ⁻¹)	Uncertainty (k=1) (Bq g ⁻¹)	Open geometry mean (Bq g ⁻¹)	Uncertainty (k=1) (Bq g ⁻¹)	Zeta score
²⁴¹ Am	3.58 ¹	0.2	5.89	2.08	1.06
⁶⁰ Co	2.34	1.09	2.53	0.28	0.16
¹³⁷ Cs	1.73	0.48	2.26	0.25	1.00

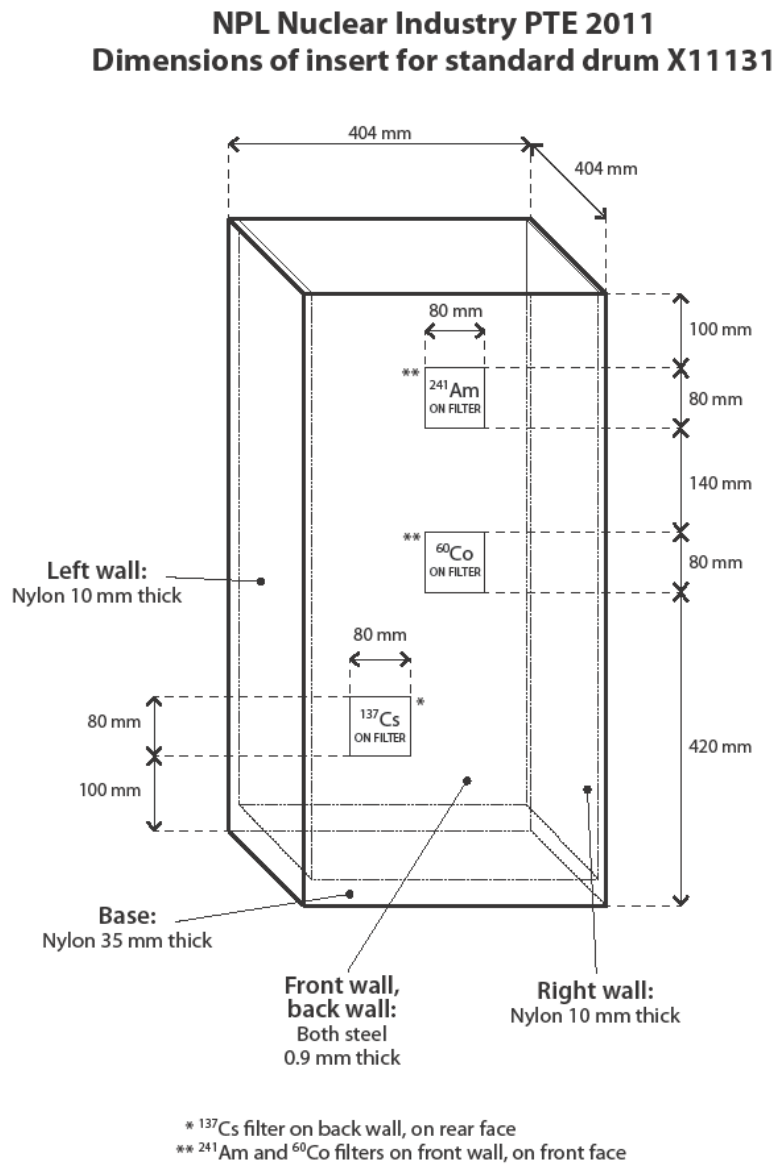
Table 10 - Comparison of ISOCS results with ISOTOPIC results (pre-disclosure)

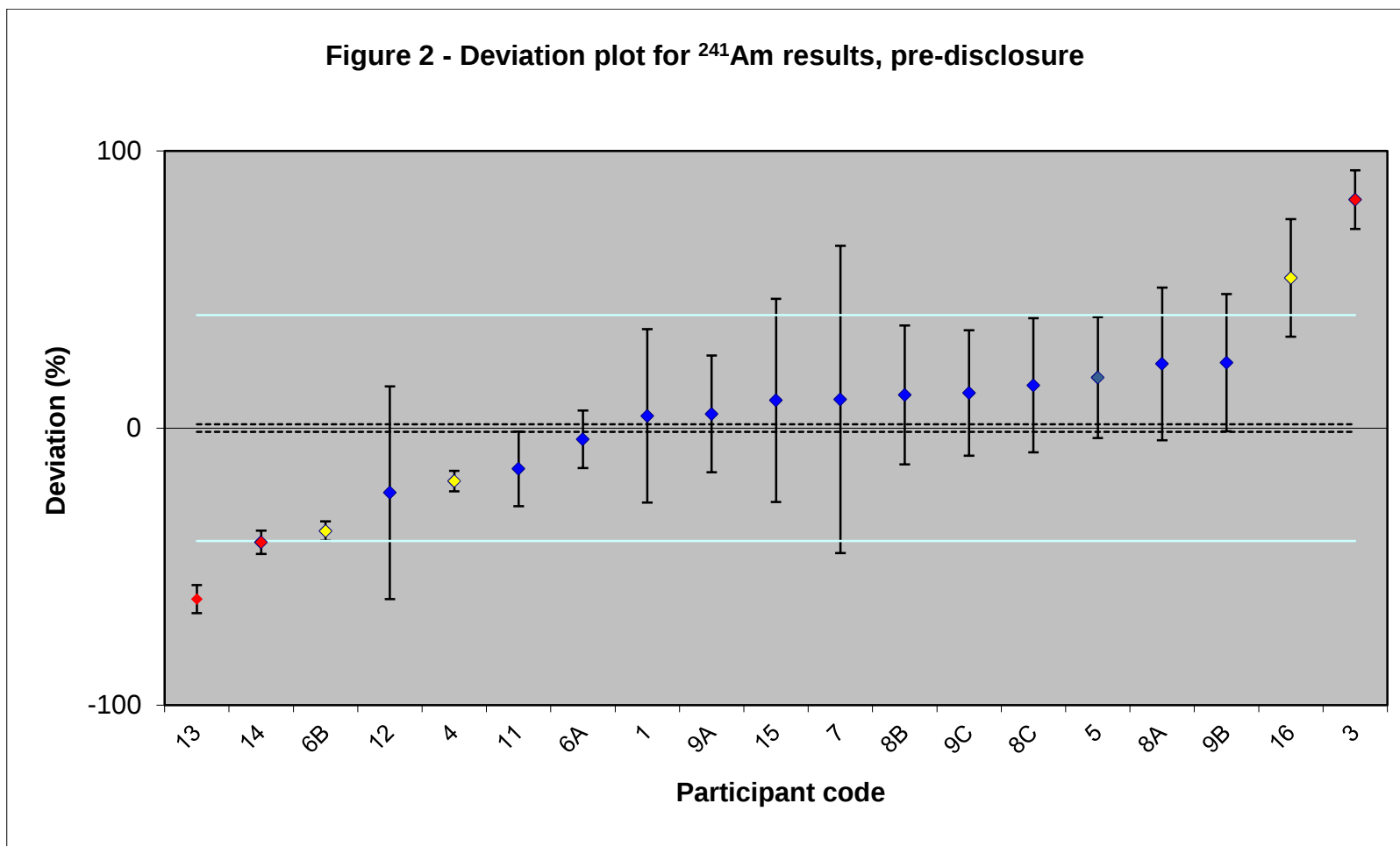
Radionuclide	ISOCS mean (Bq g ⁻¹)	Uncertainty (k=1) (Bq g ⁻¹)	ISOTOPIC mean (Bq g ⁻¹)	Uncertainty (k=1) (Bq g ⁻¹)	Zeta score
²⁴¹ Am	6.74	1.23	5.06	2.51	0.60
⁶⁰ Co	2.54	0.40	2.32	0.14	0.52
¹³⁷ Cs	2.38	0.35	2.12	0.13	0.70

¹ Single measurement.

12 FIGURES

Figure 1 - Schematic diagram of 'drum insert'





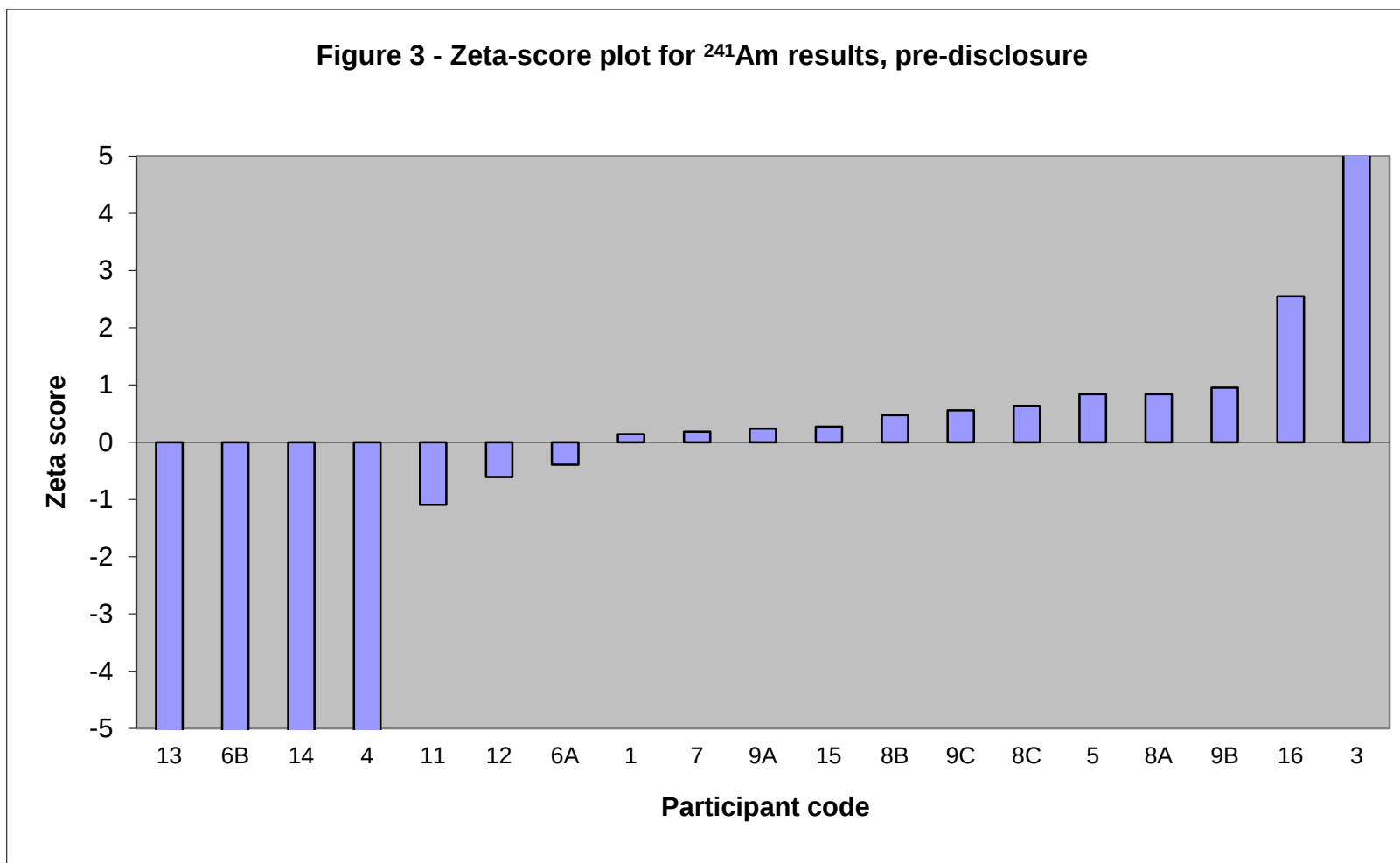
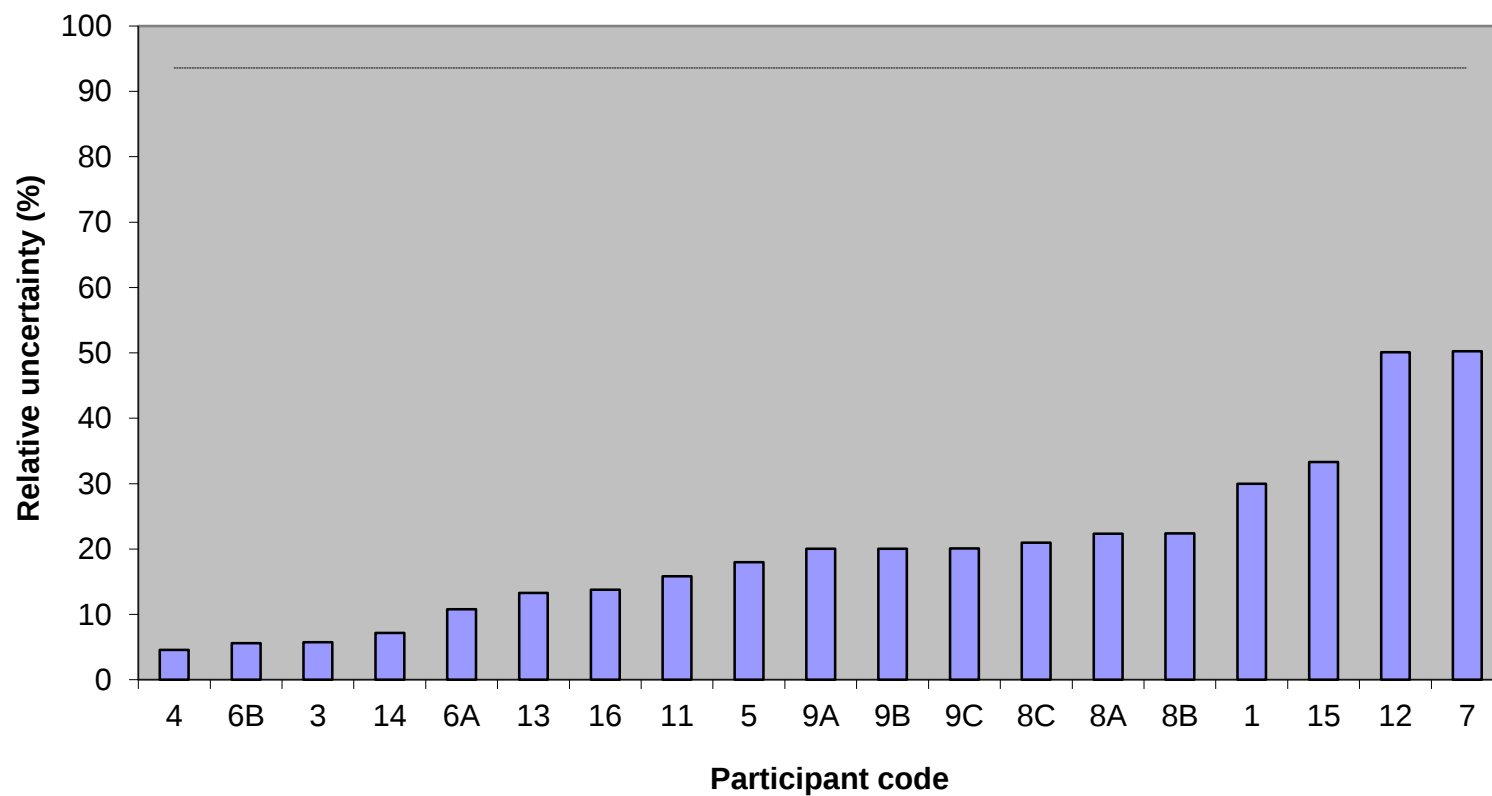
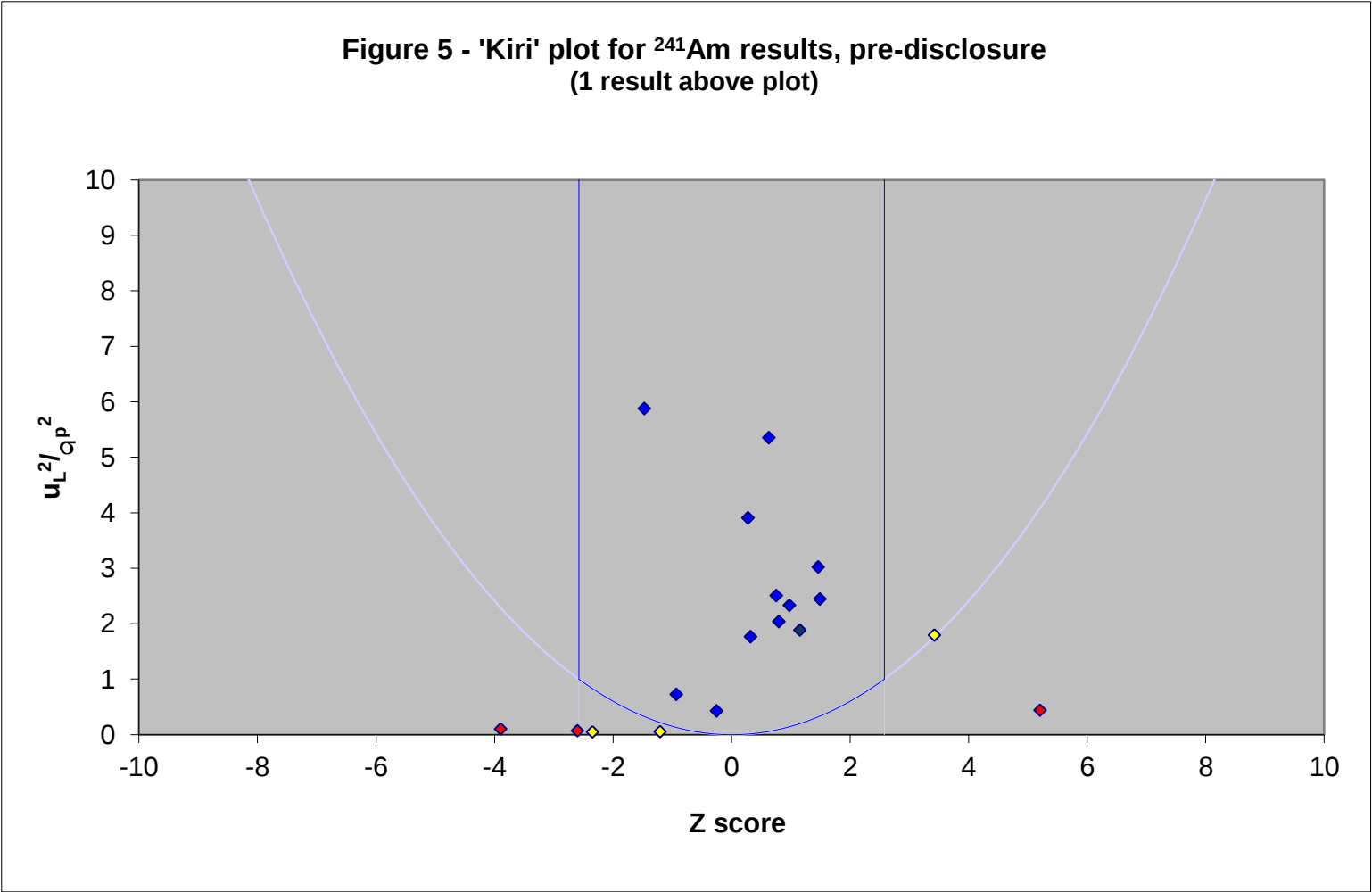
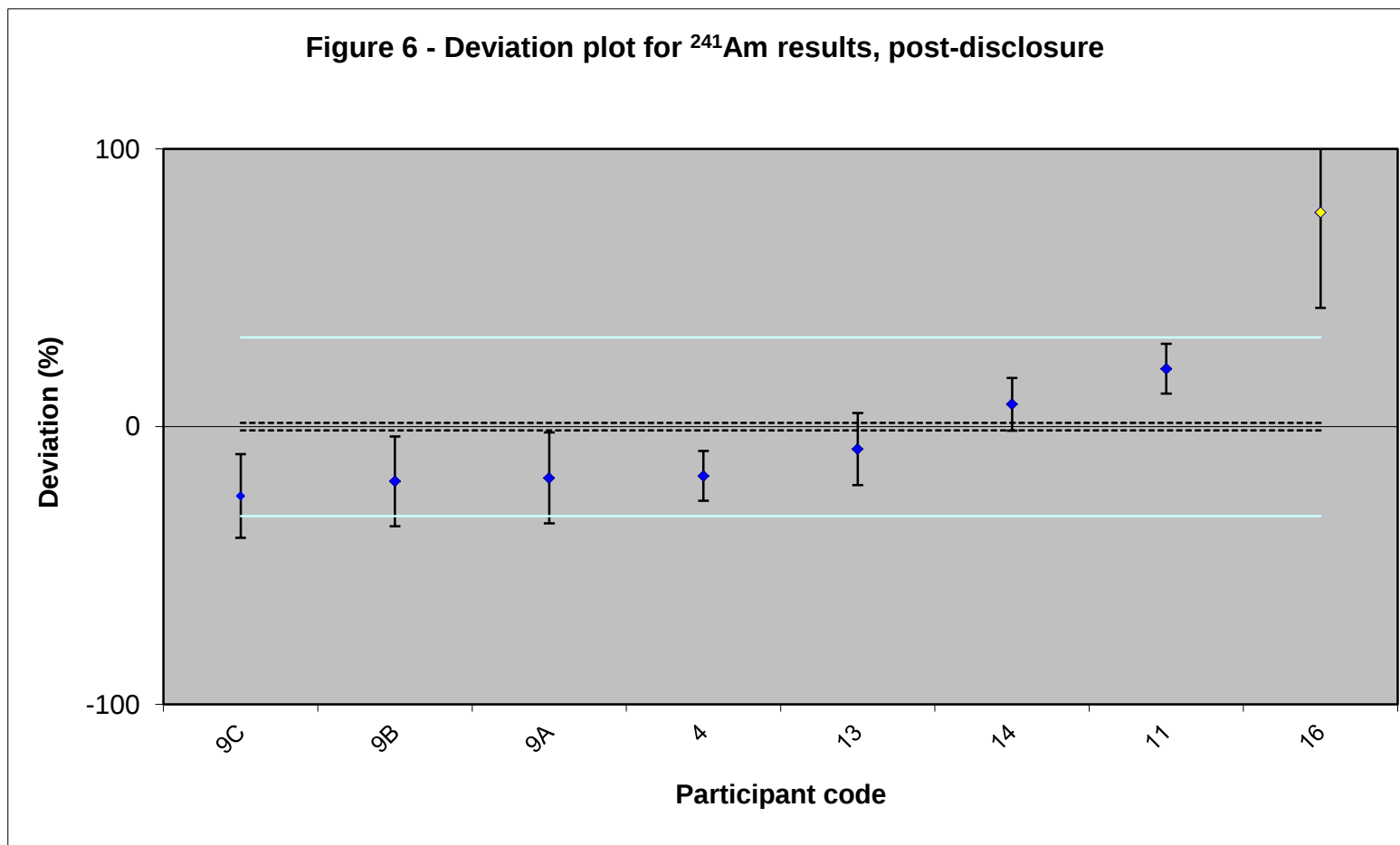
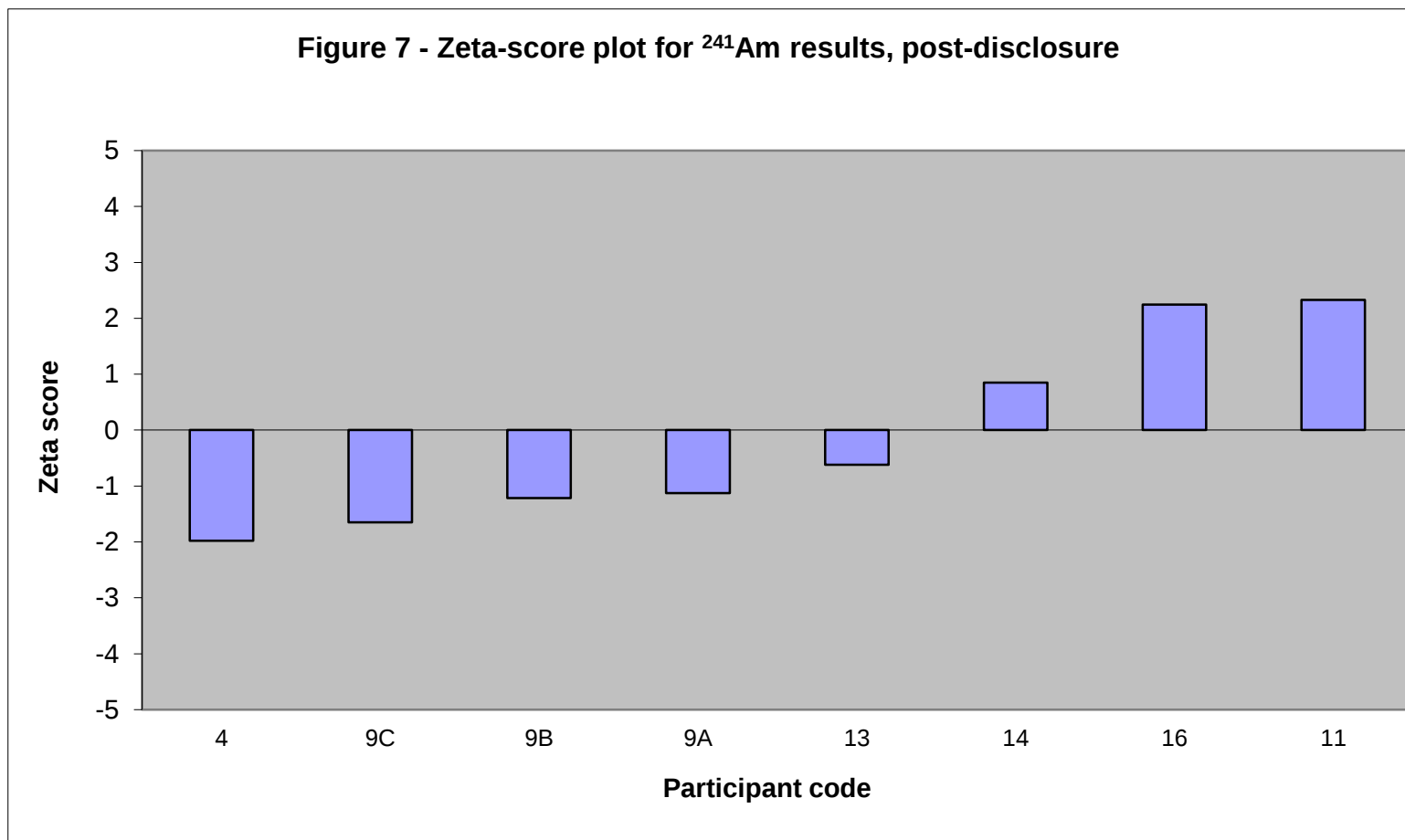
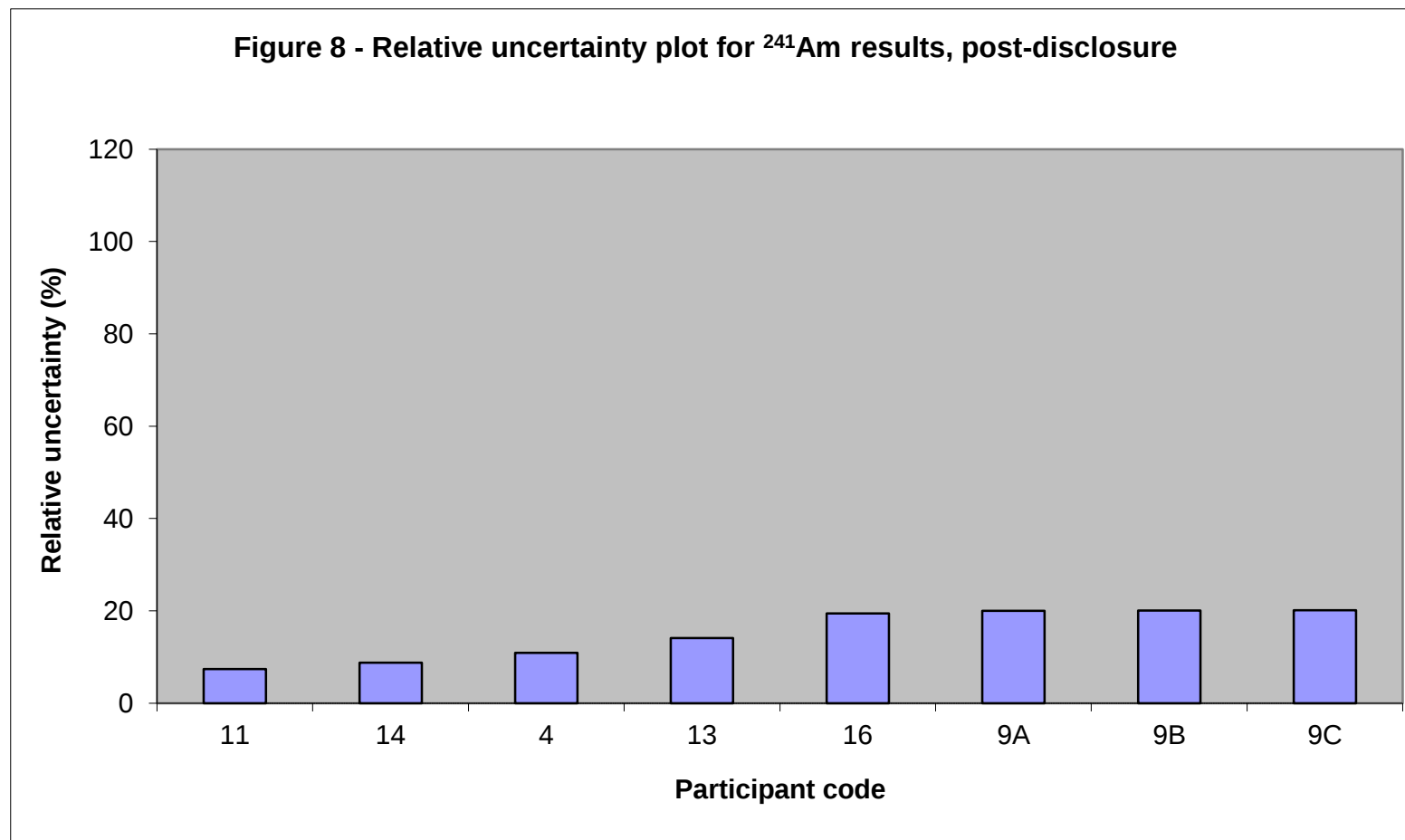


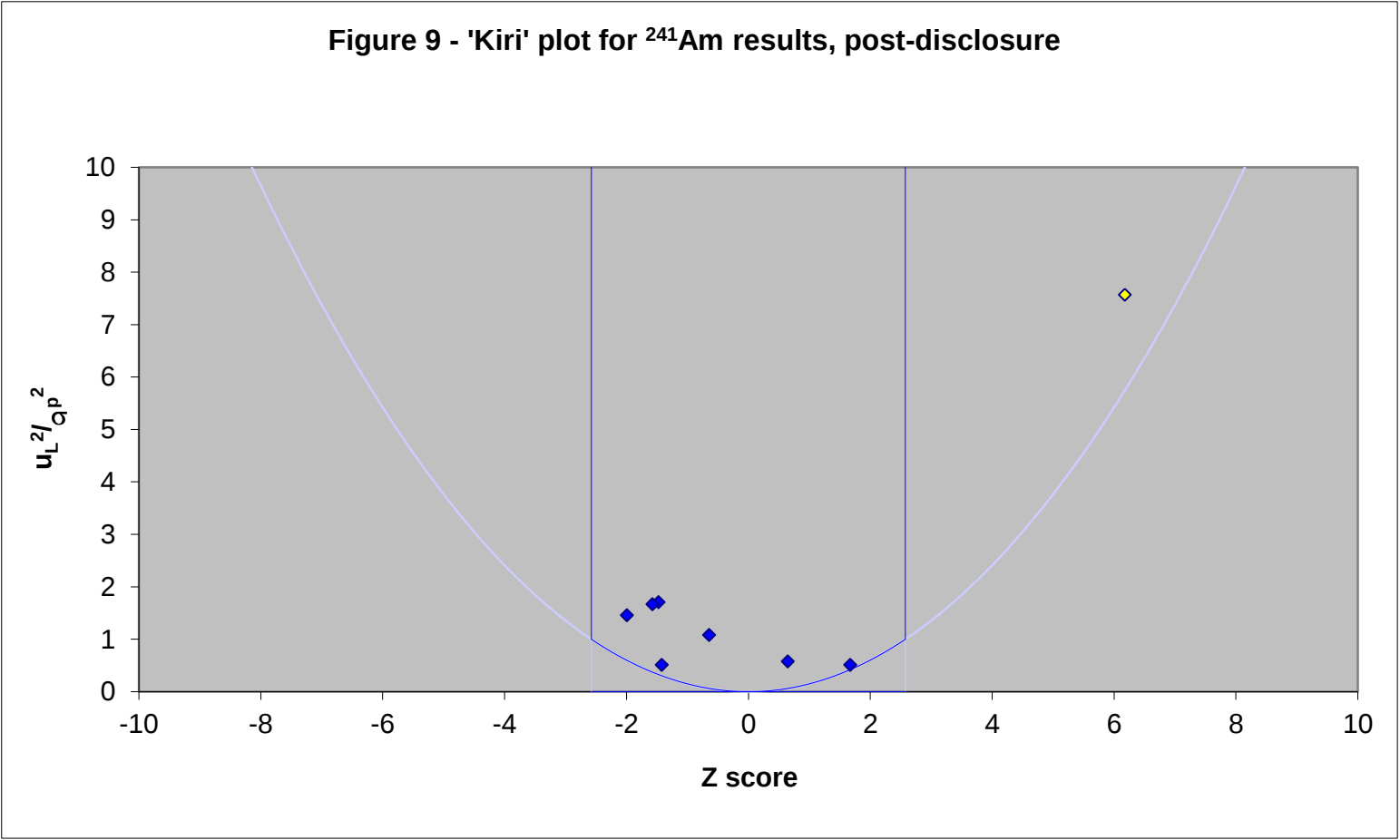
Figure 4 - Relative uncertainty plot for ^{241}Am results, pre-disclosure

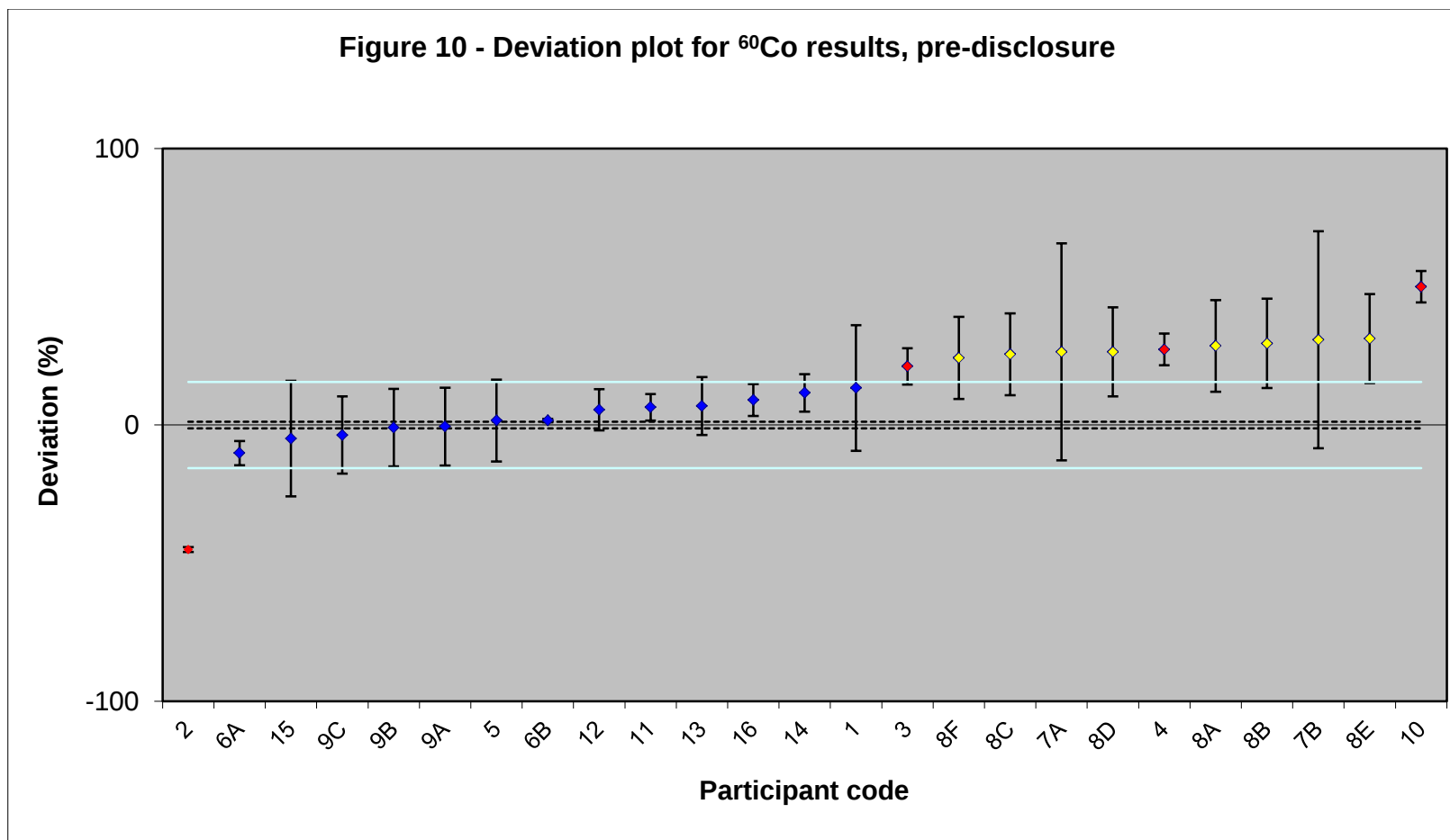


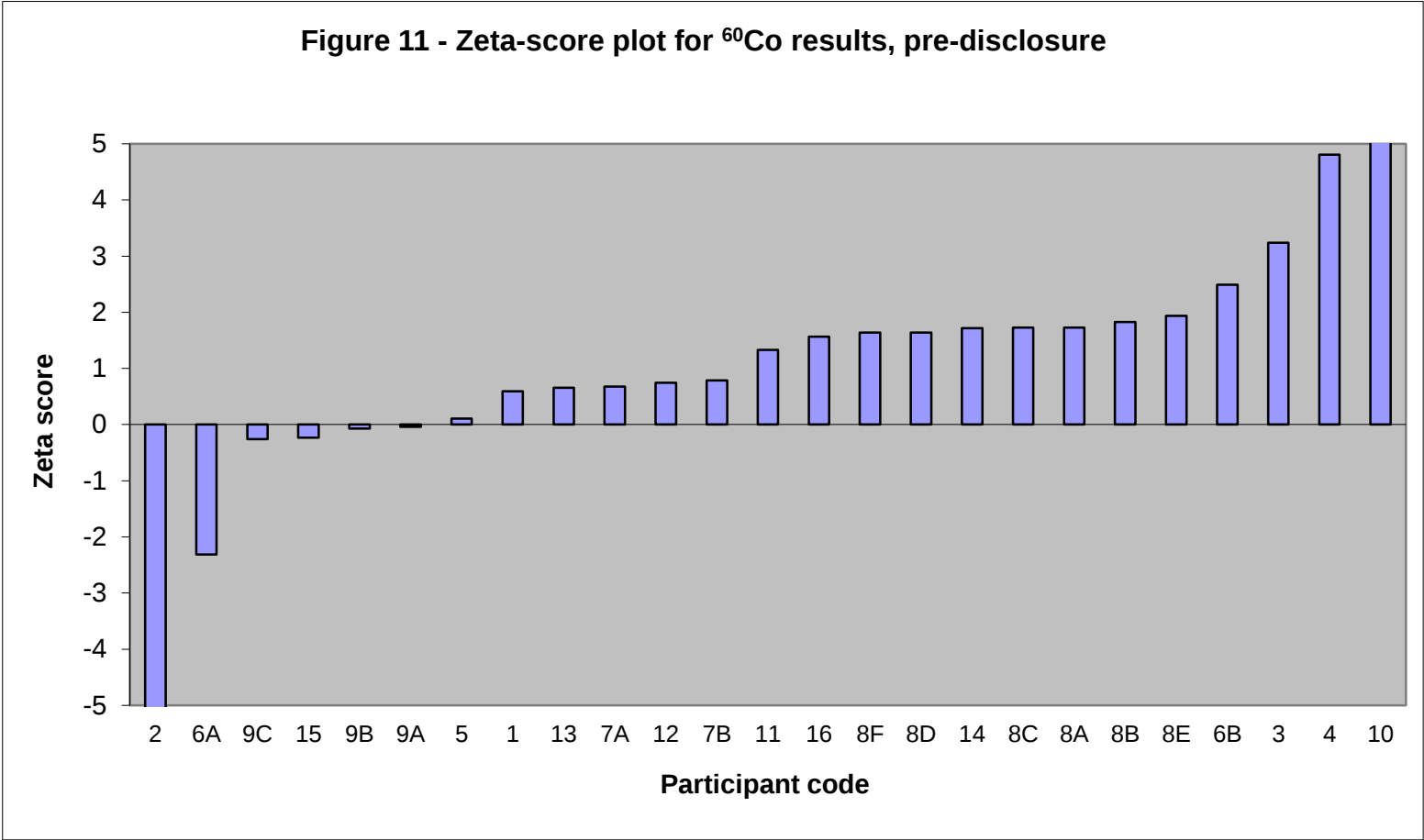


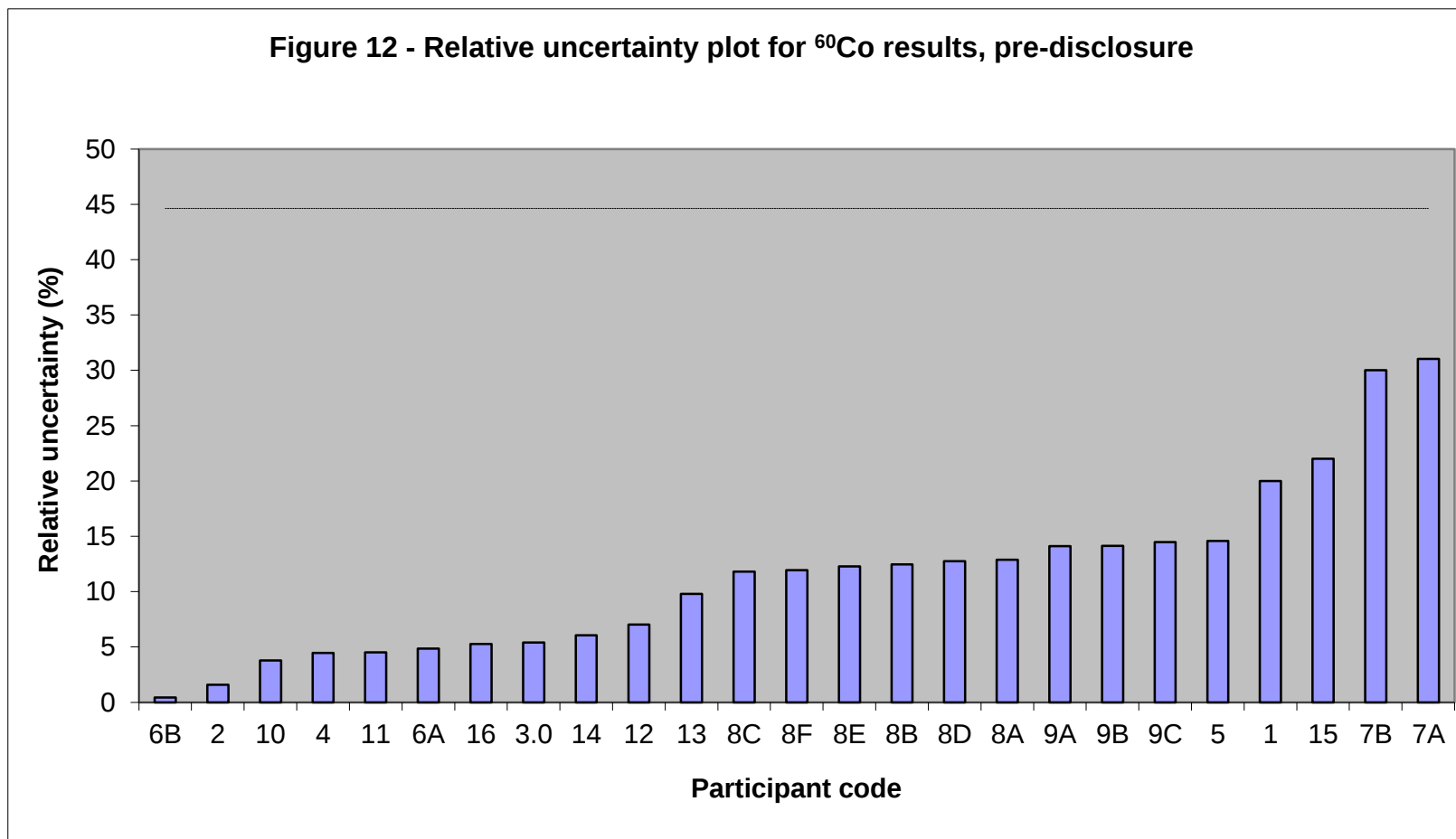


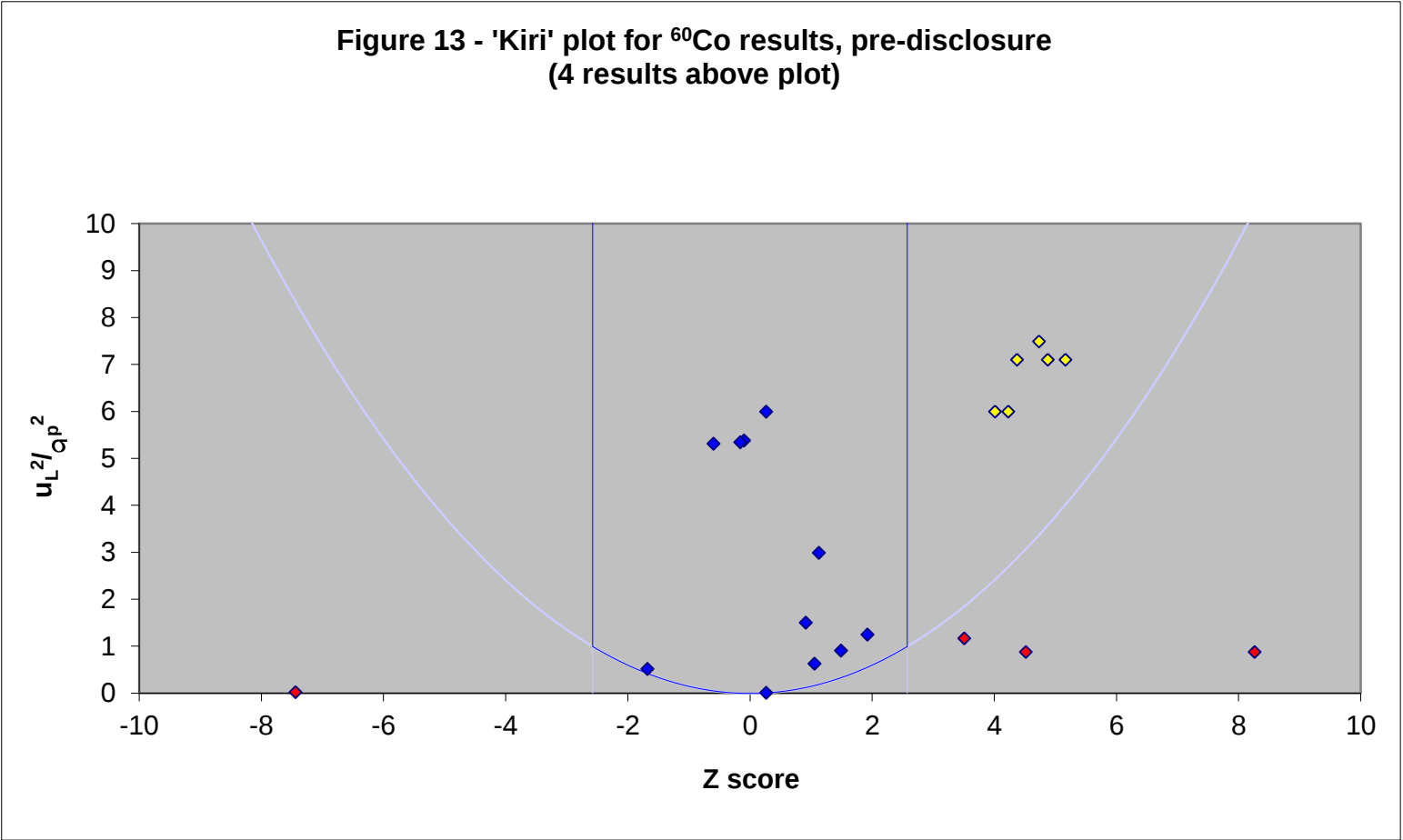


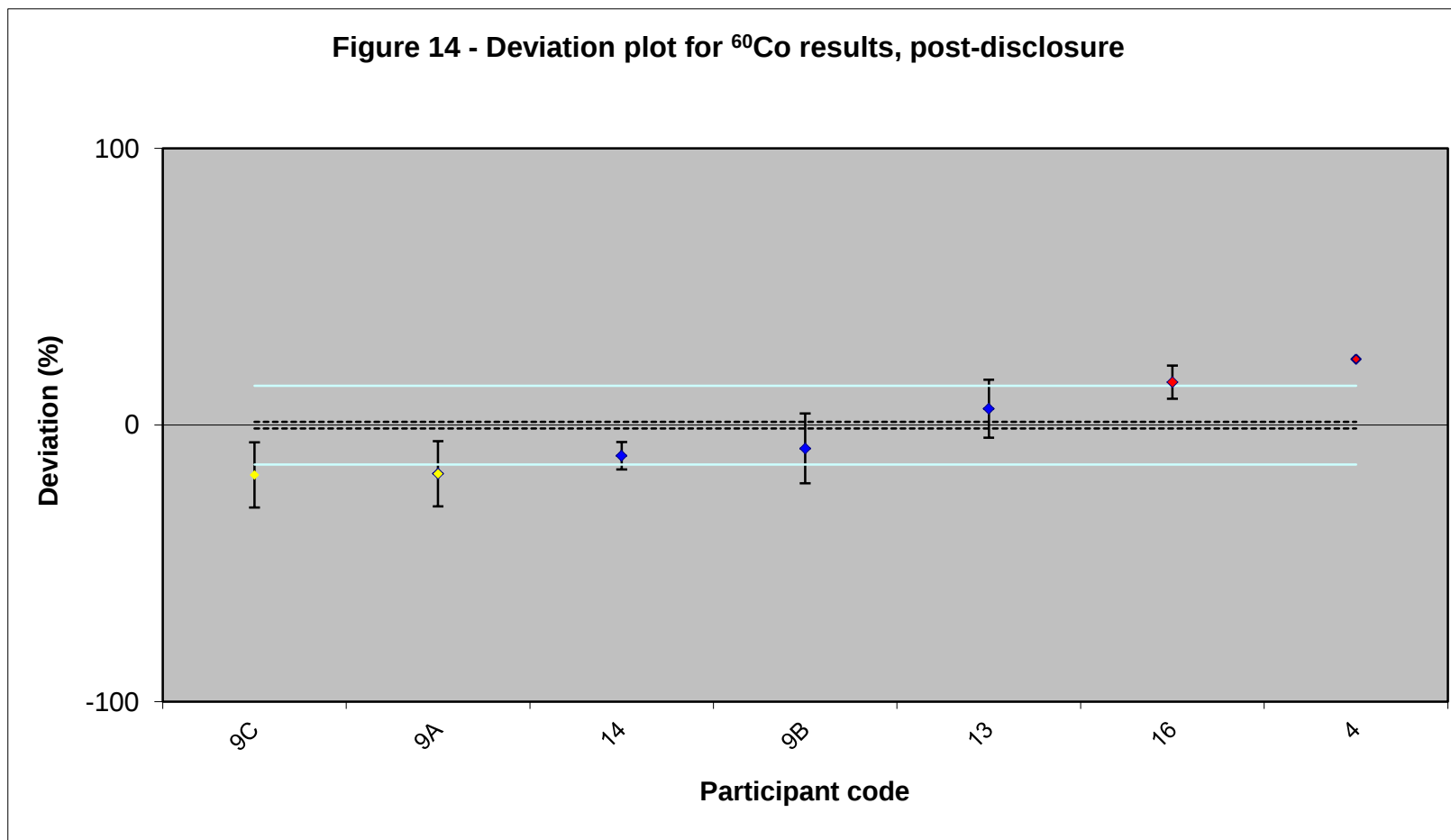


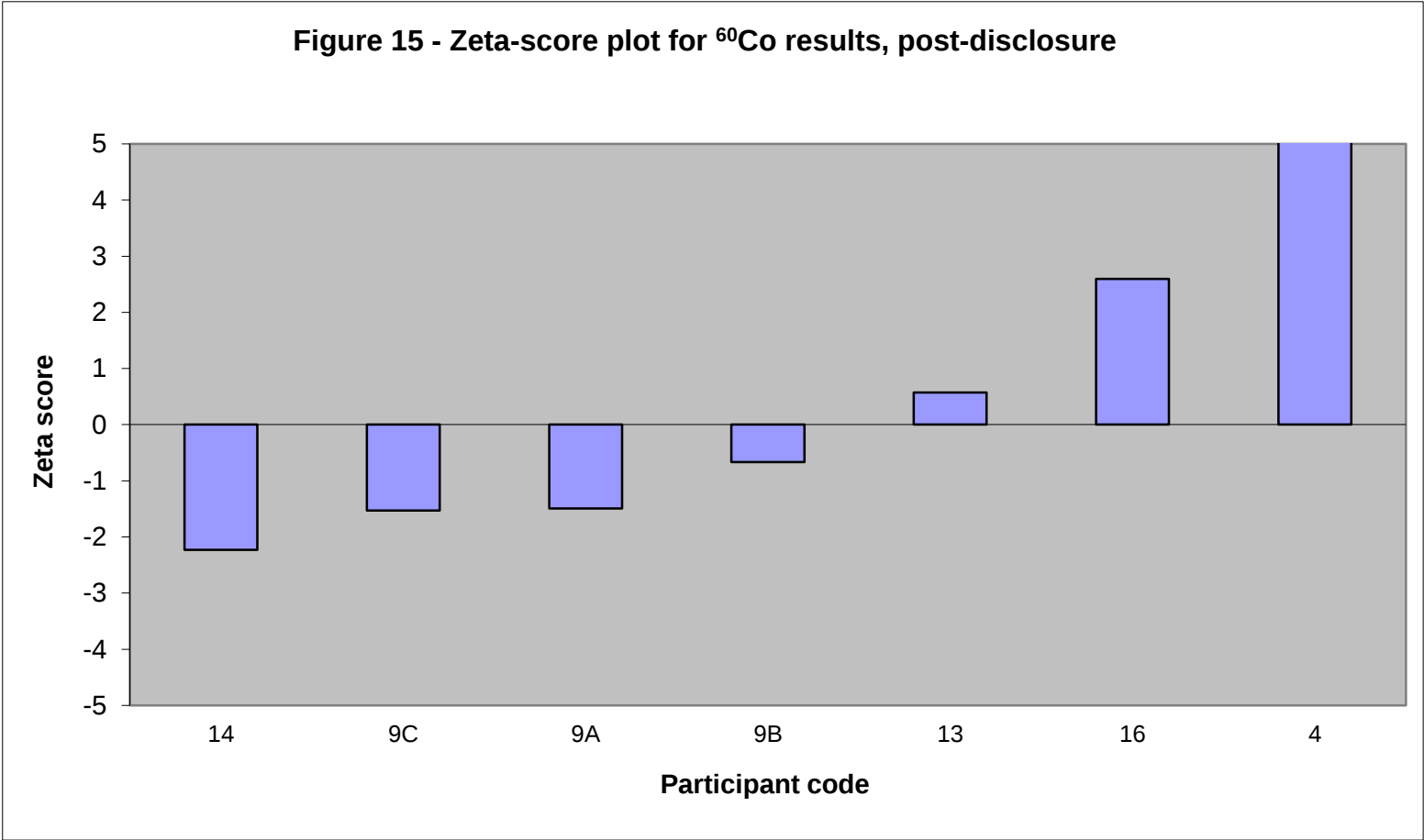


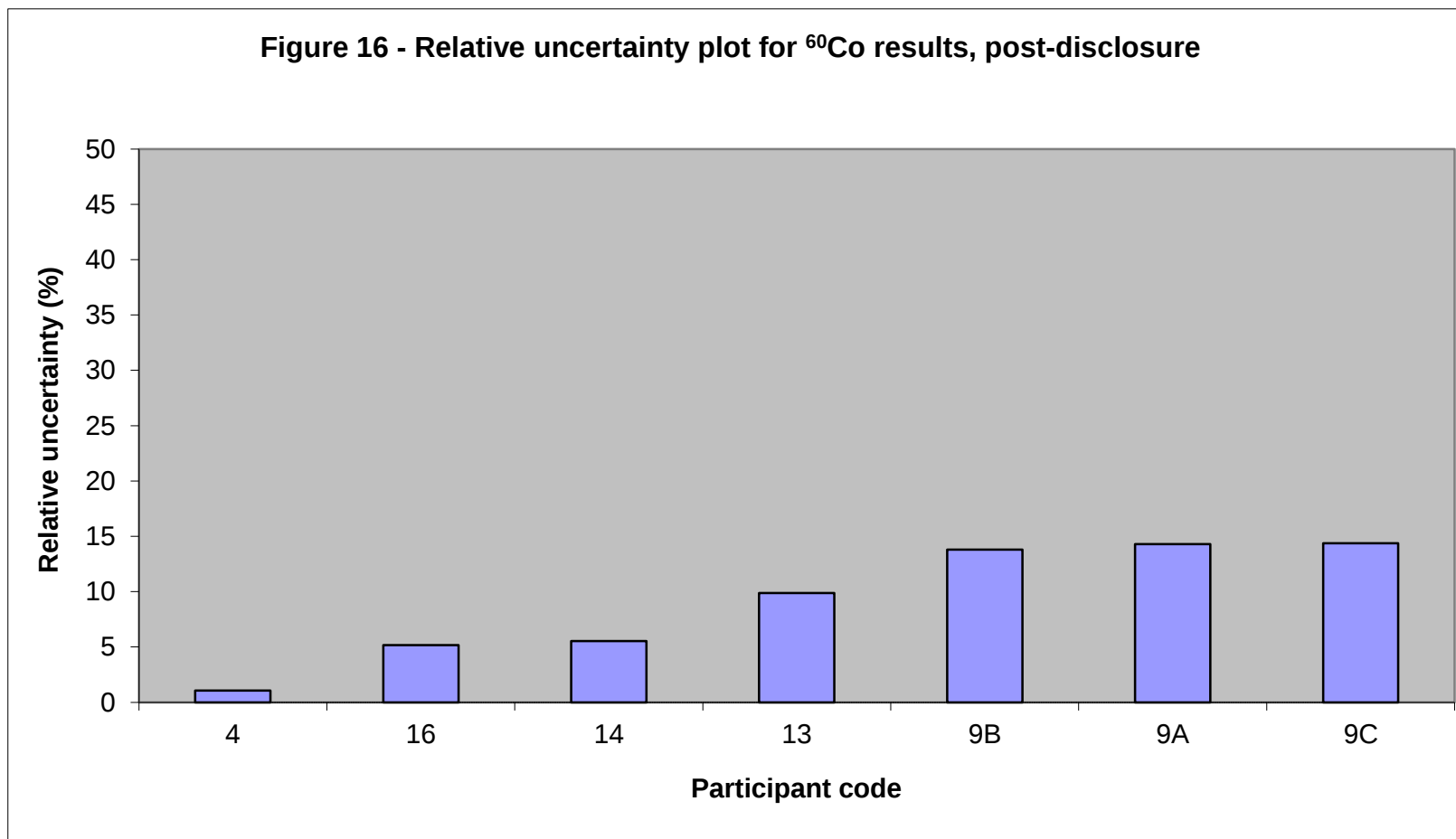


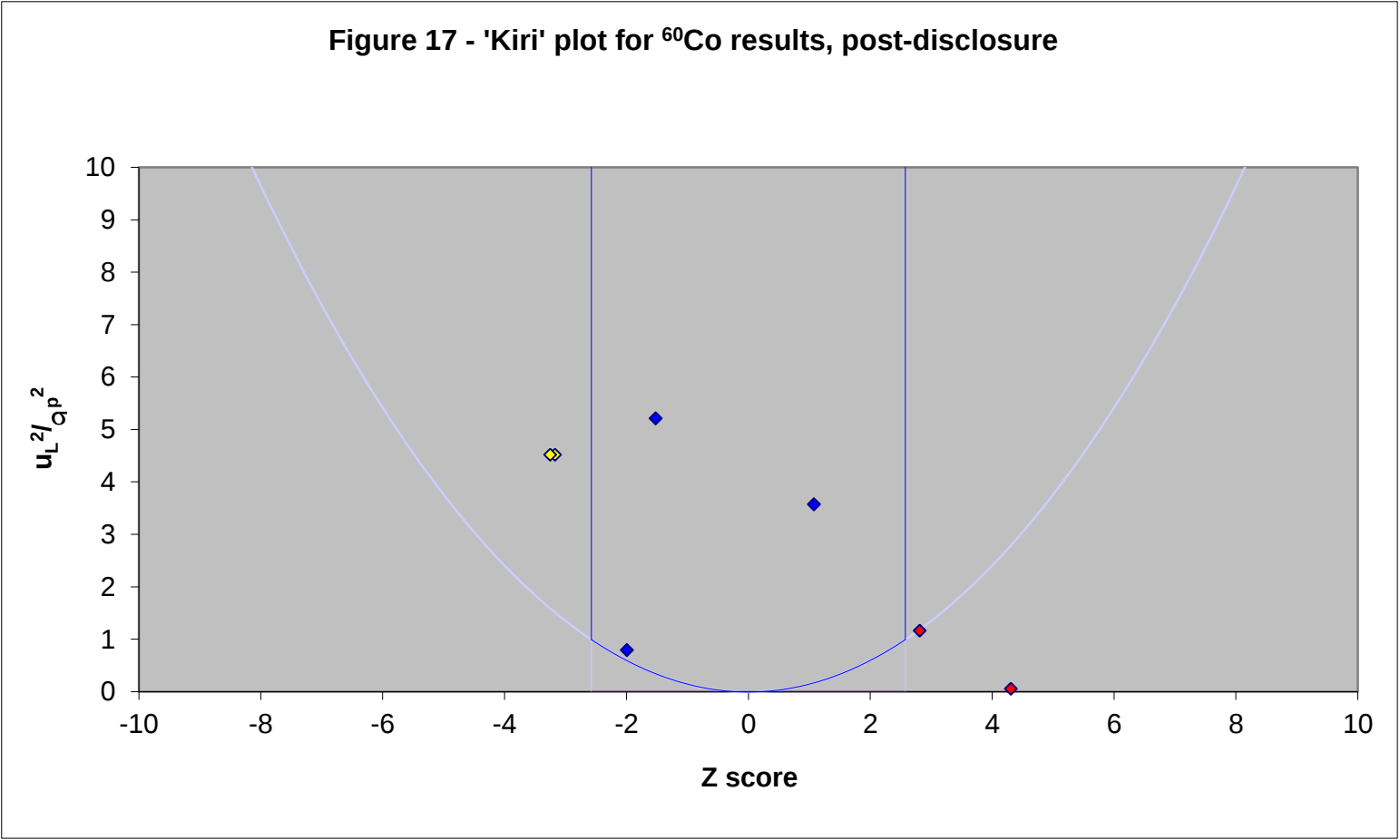


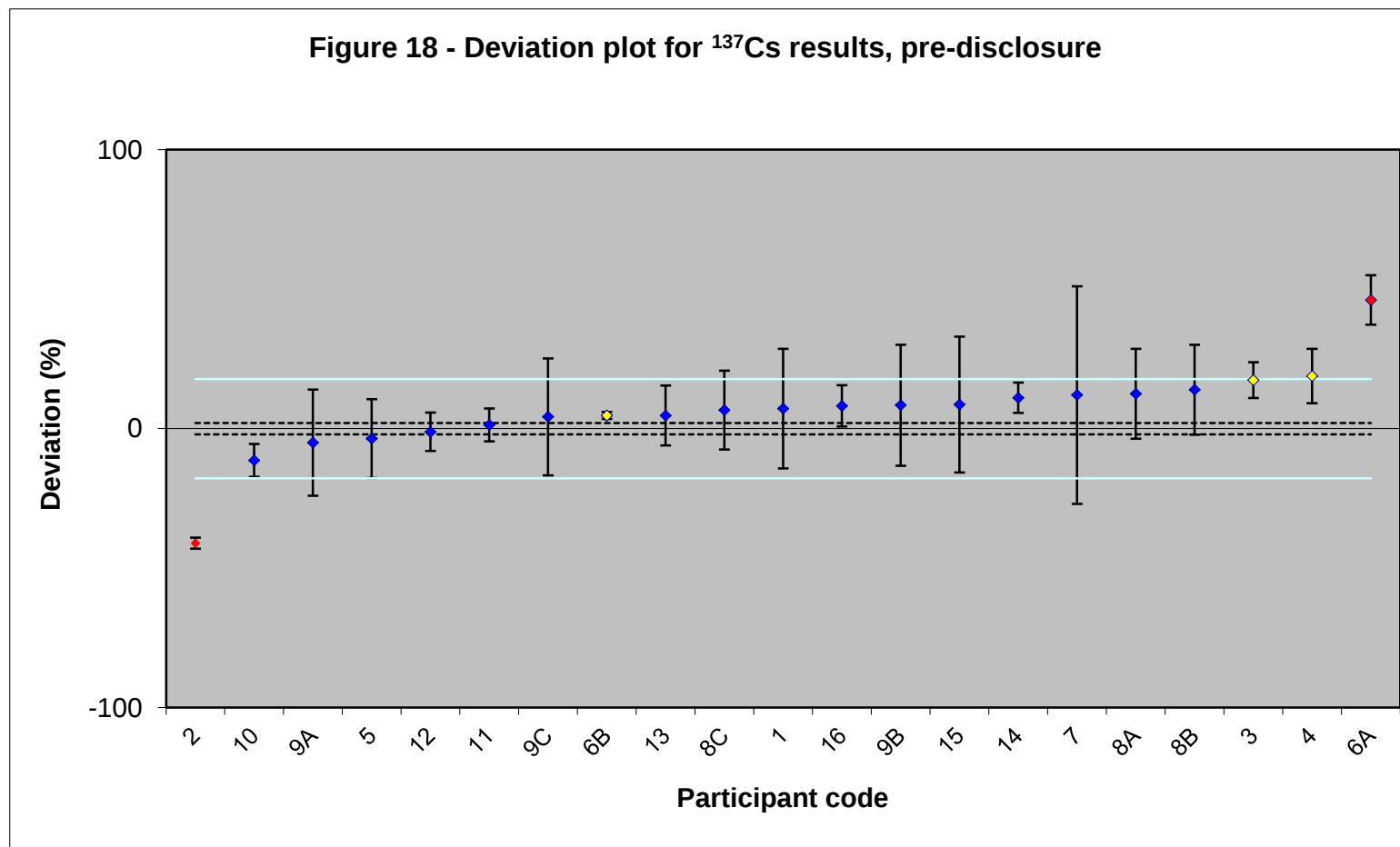












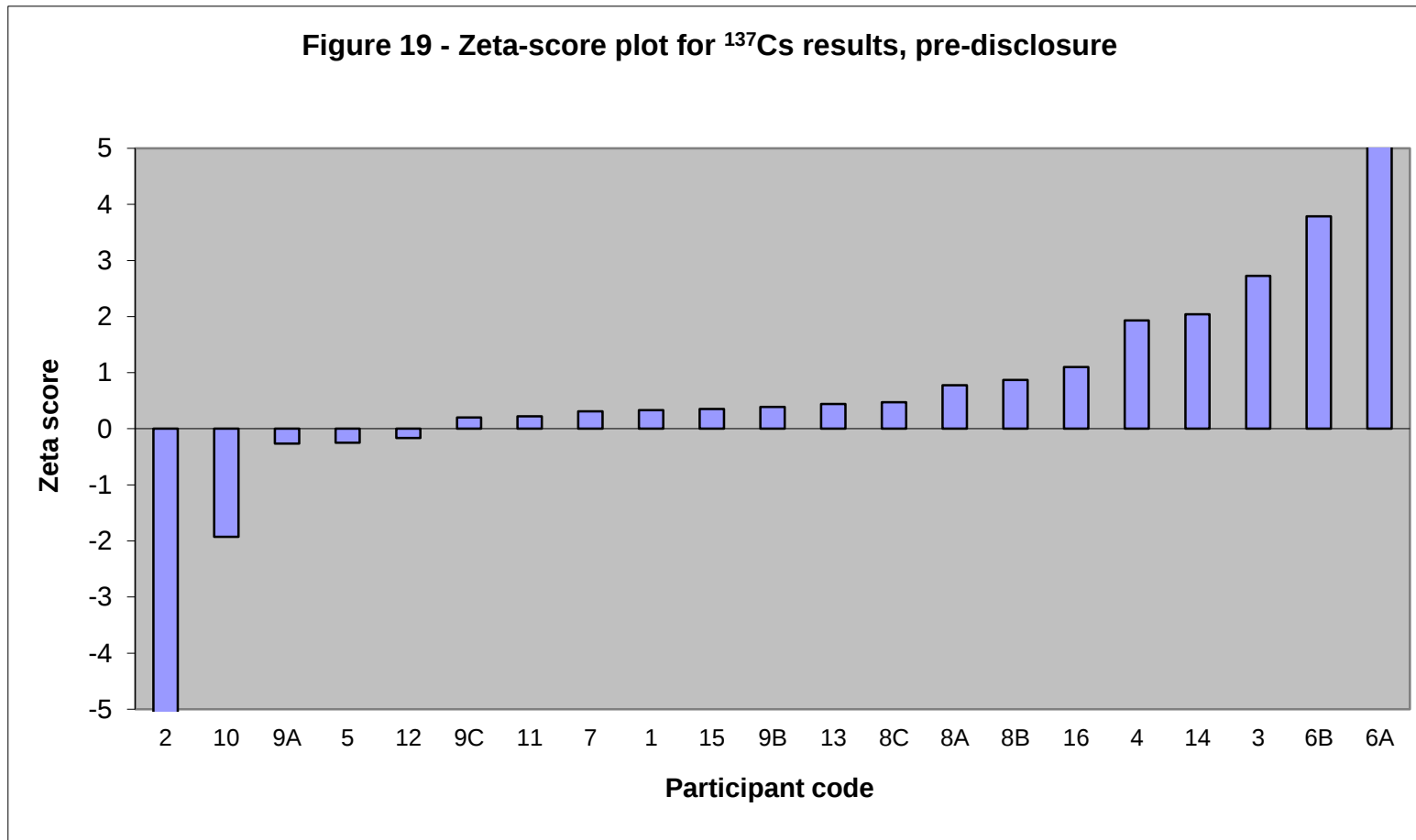
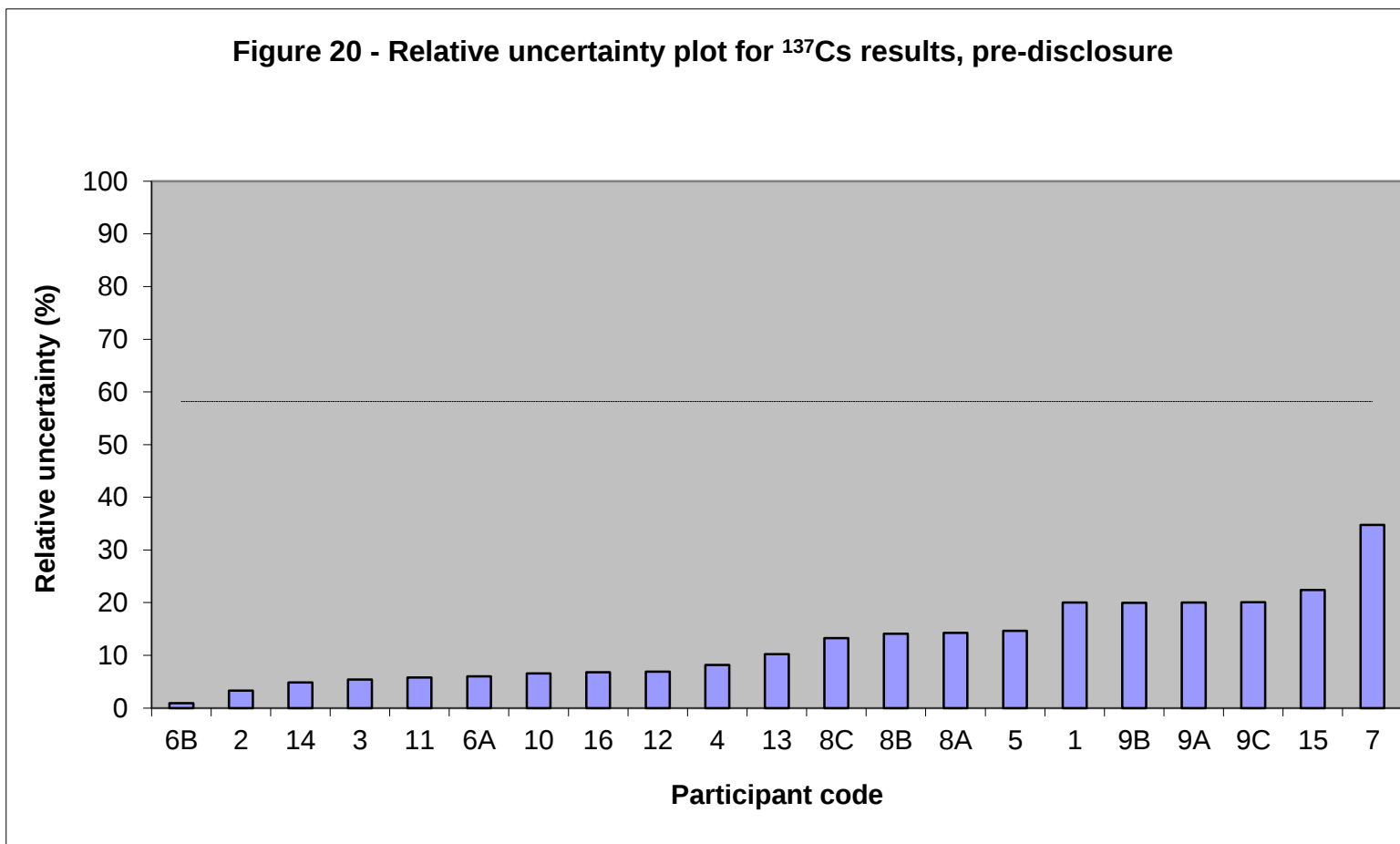
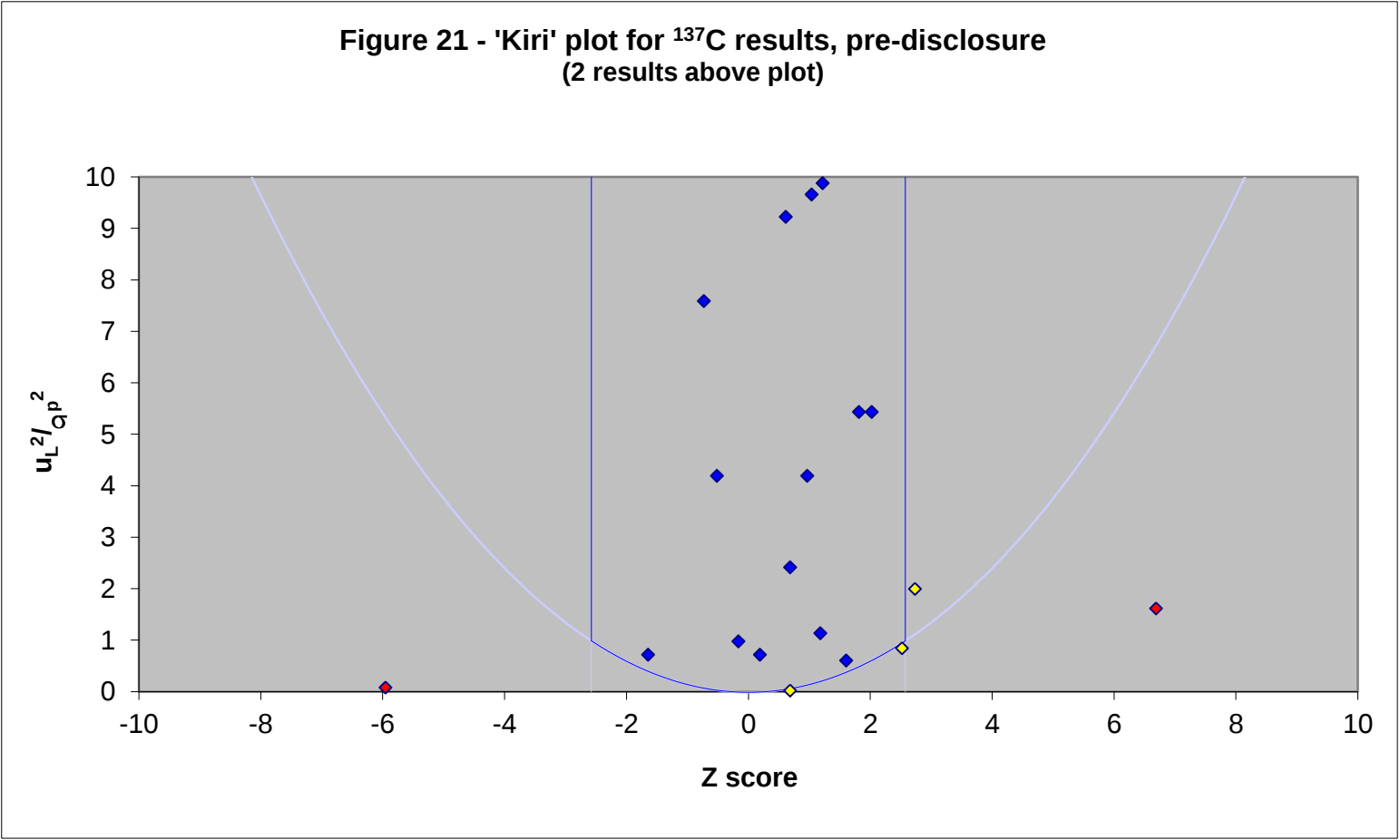
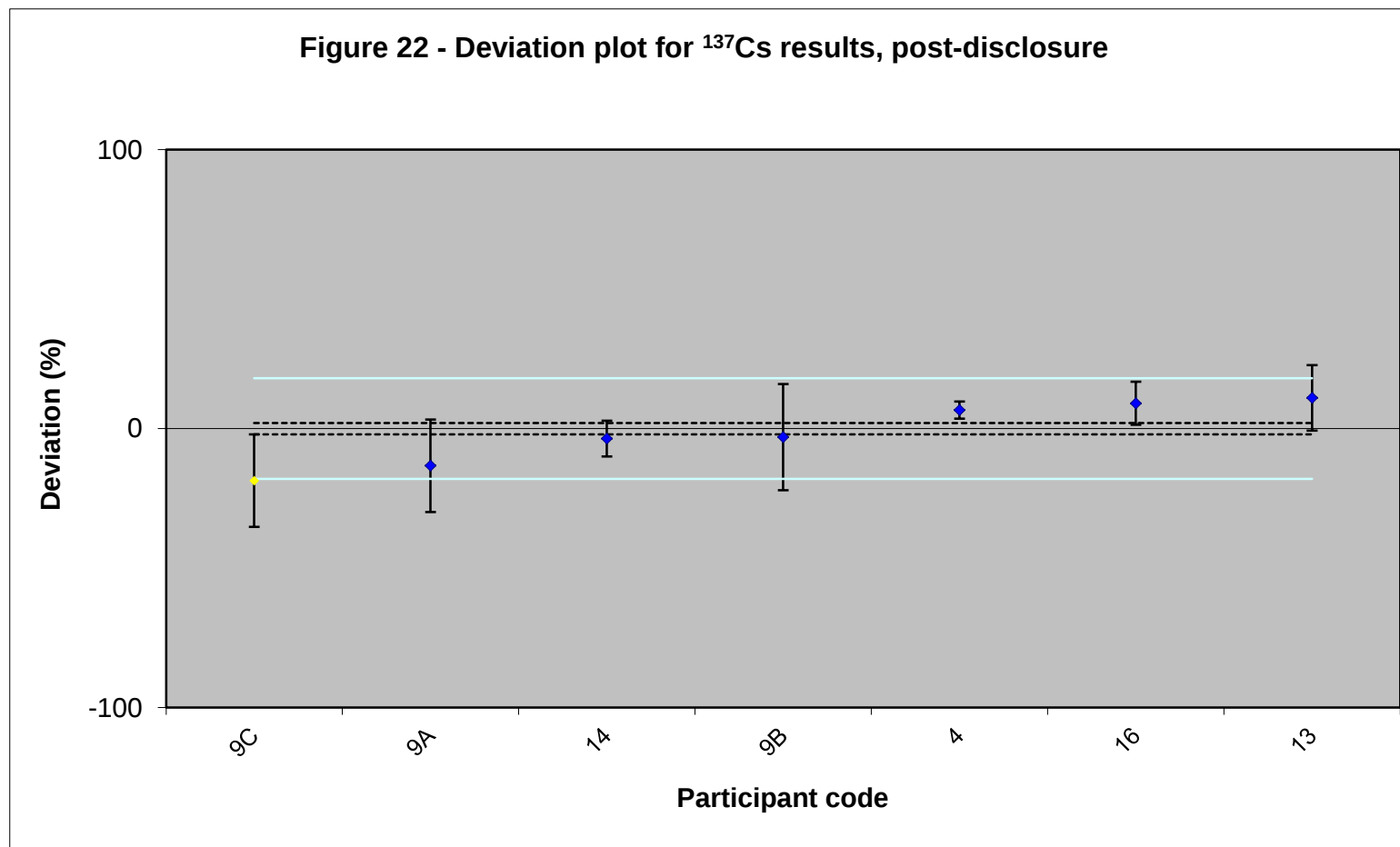
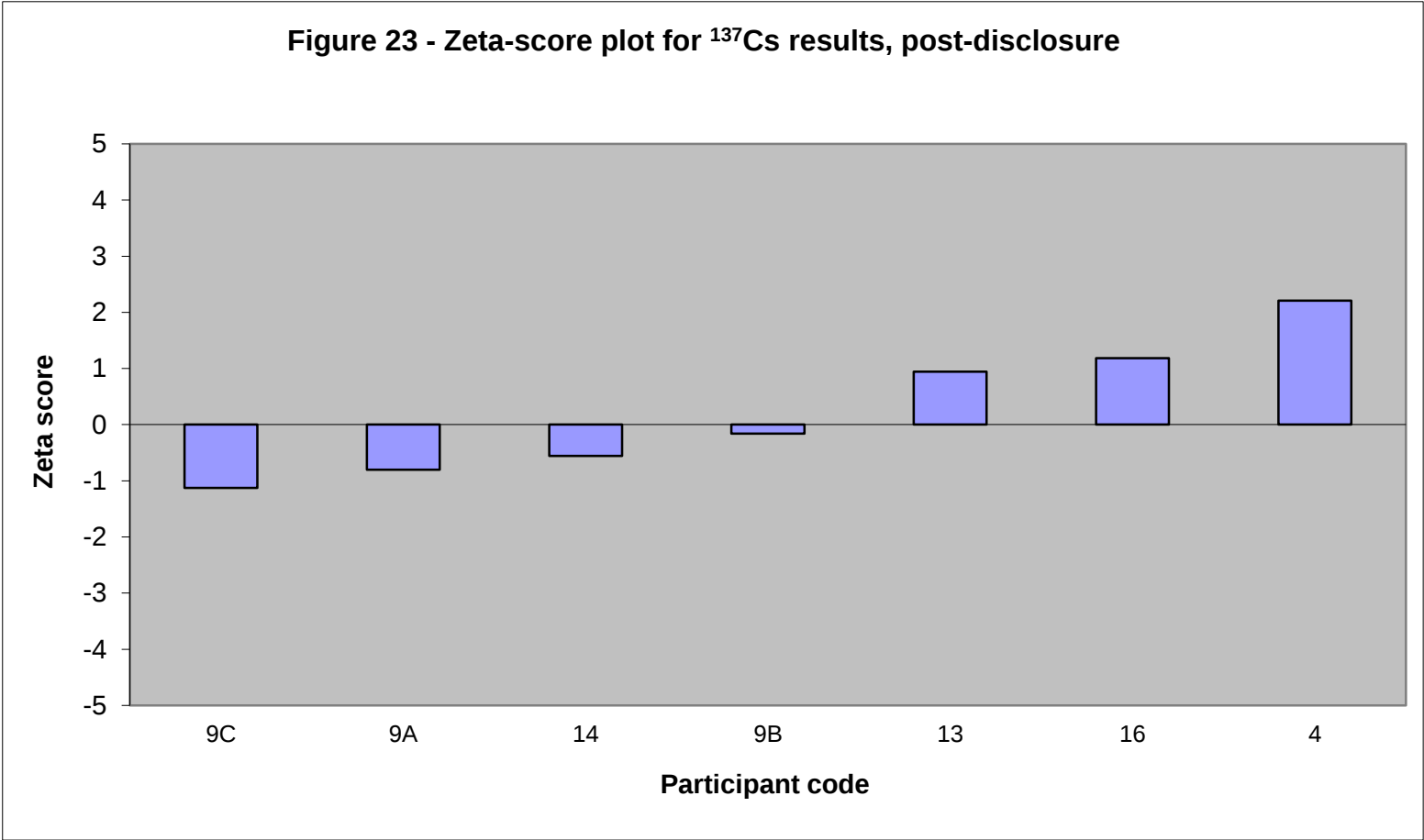
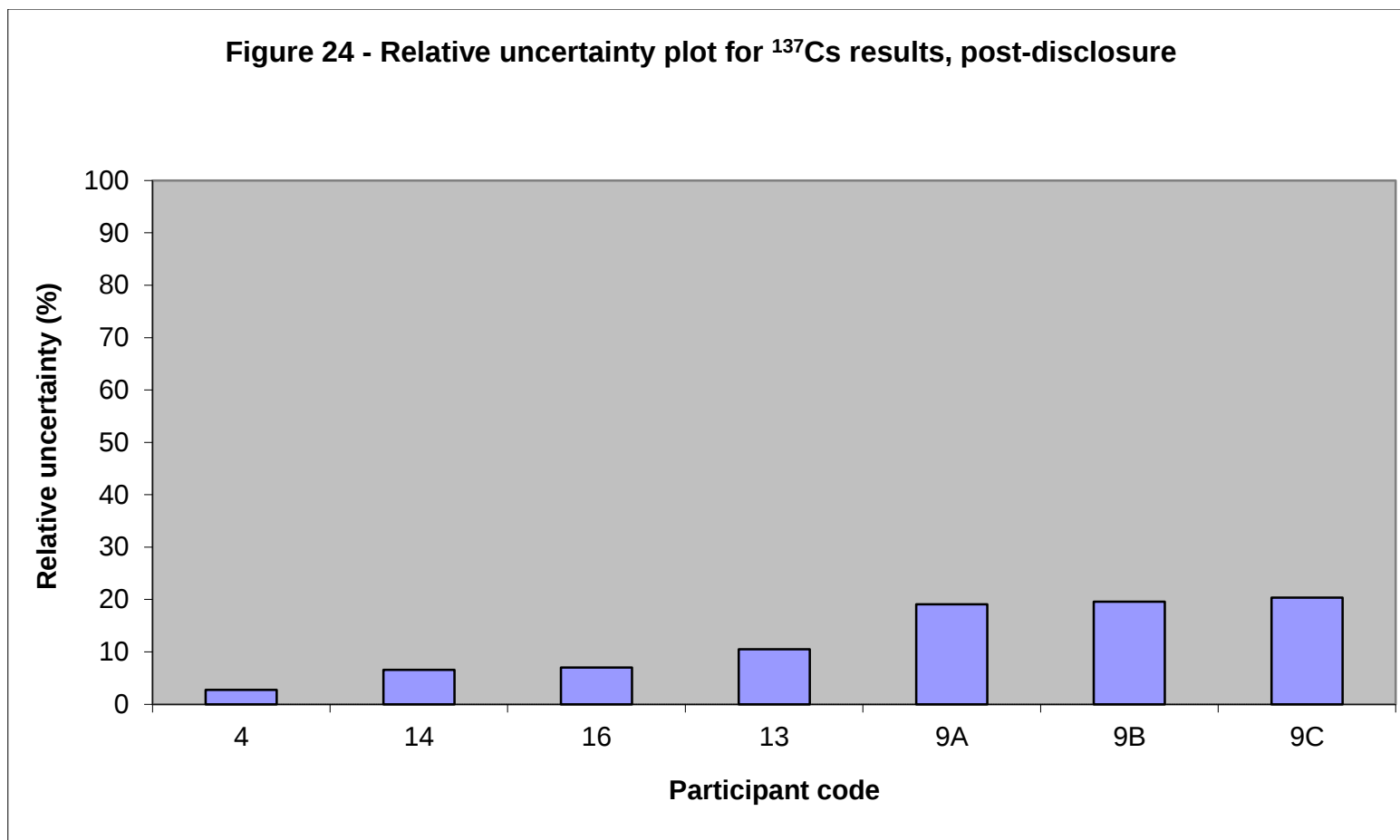


Figure 20 - Relative uncertainty plot for ^{137}Cs results, pre-disclosure









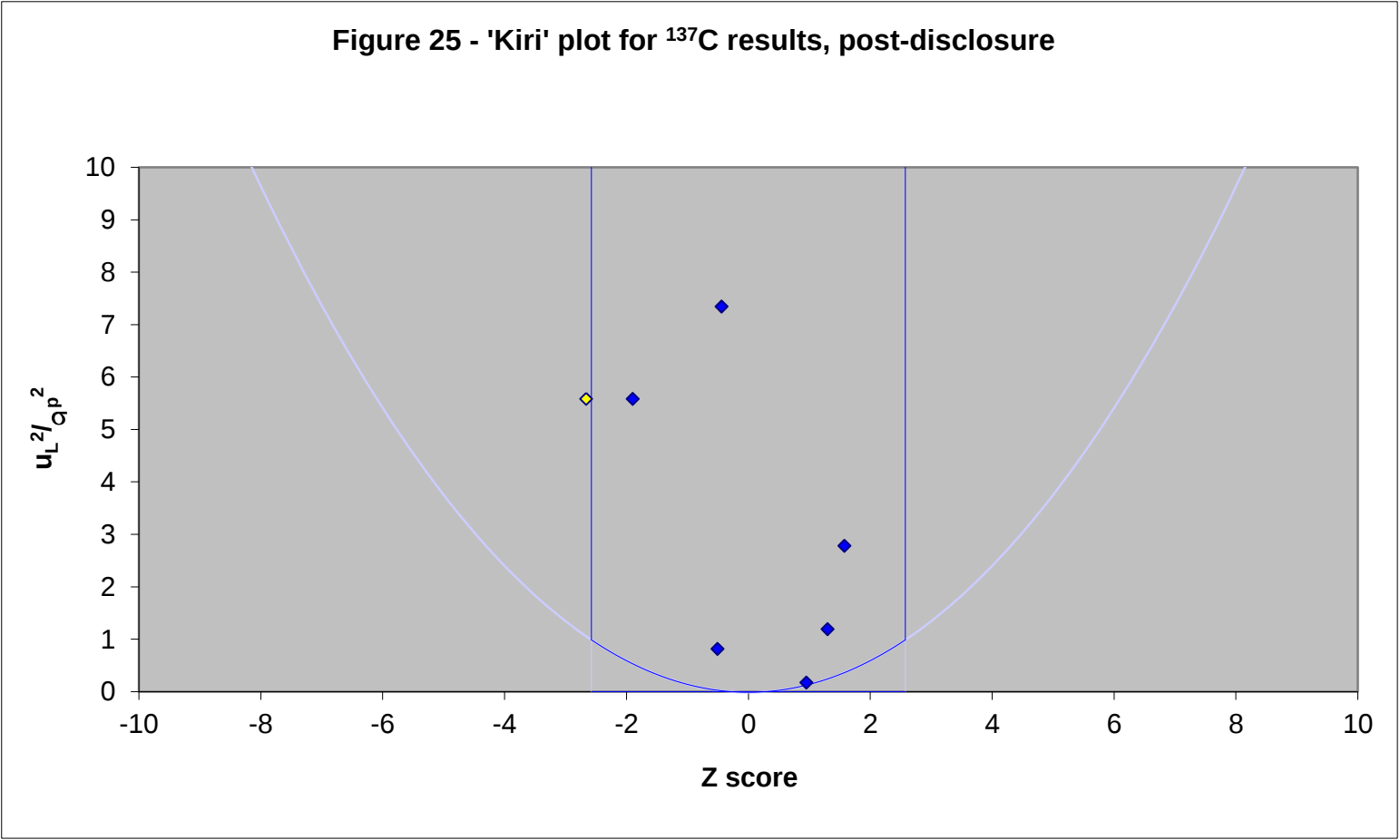
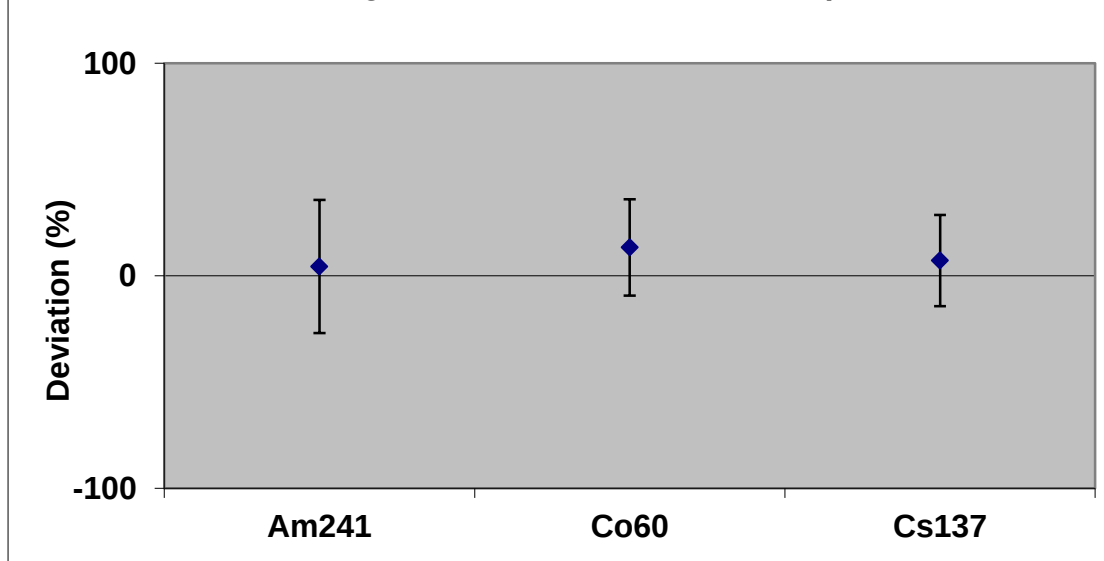
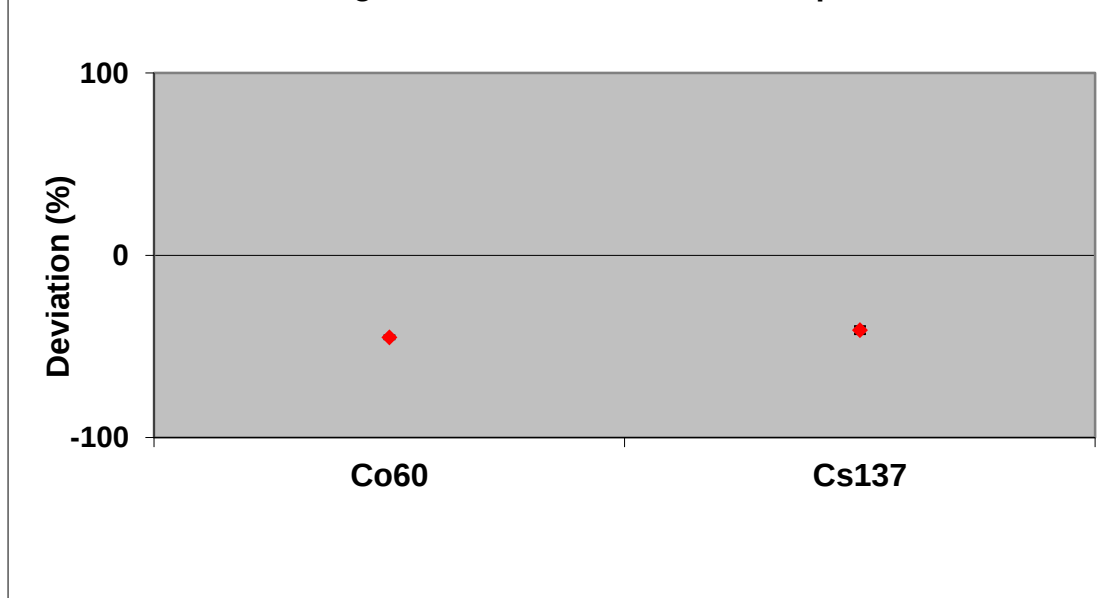
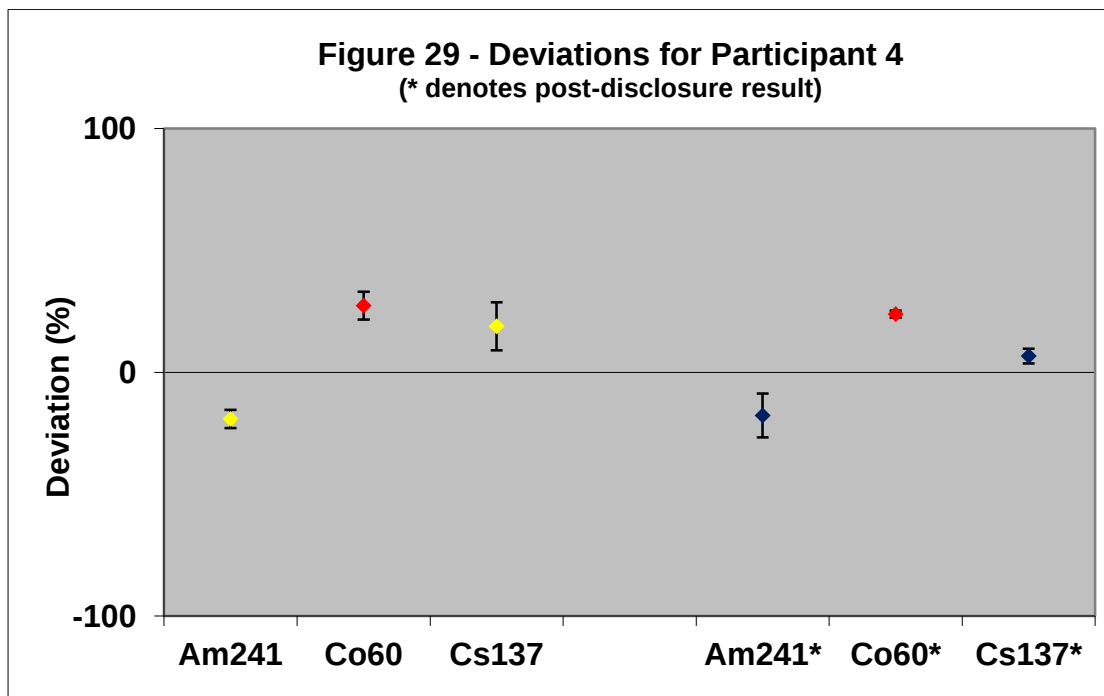
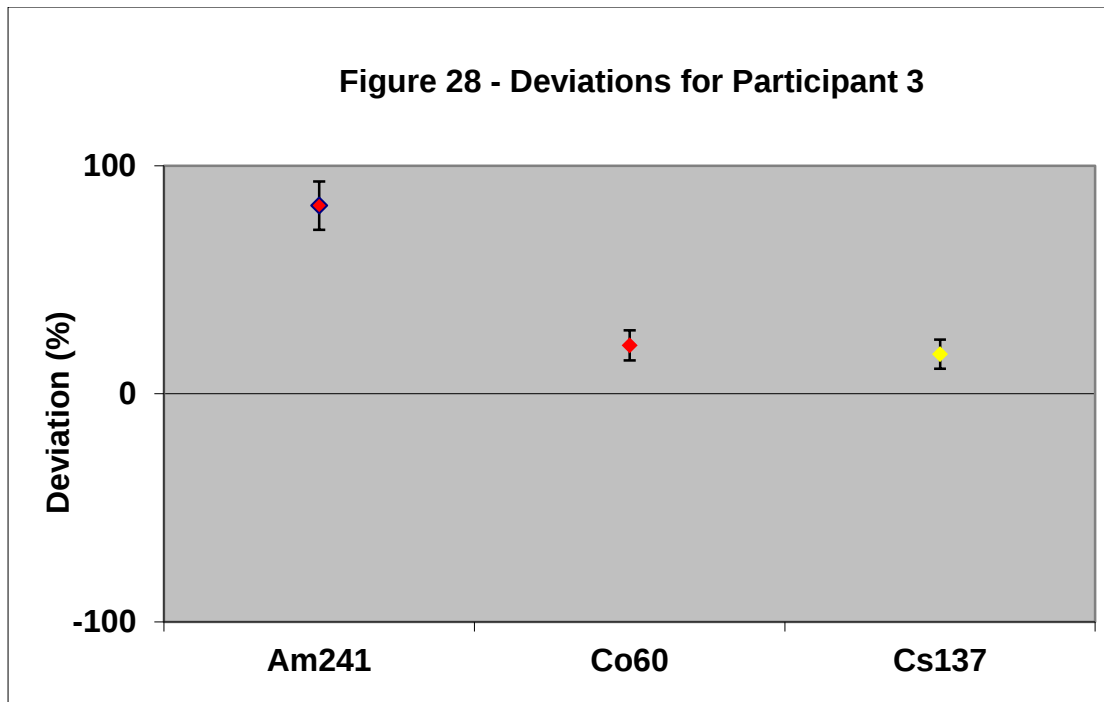
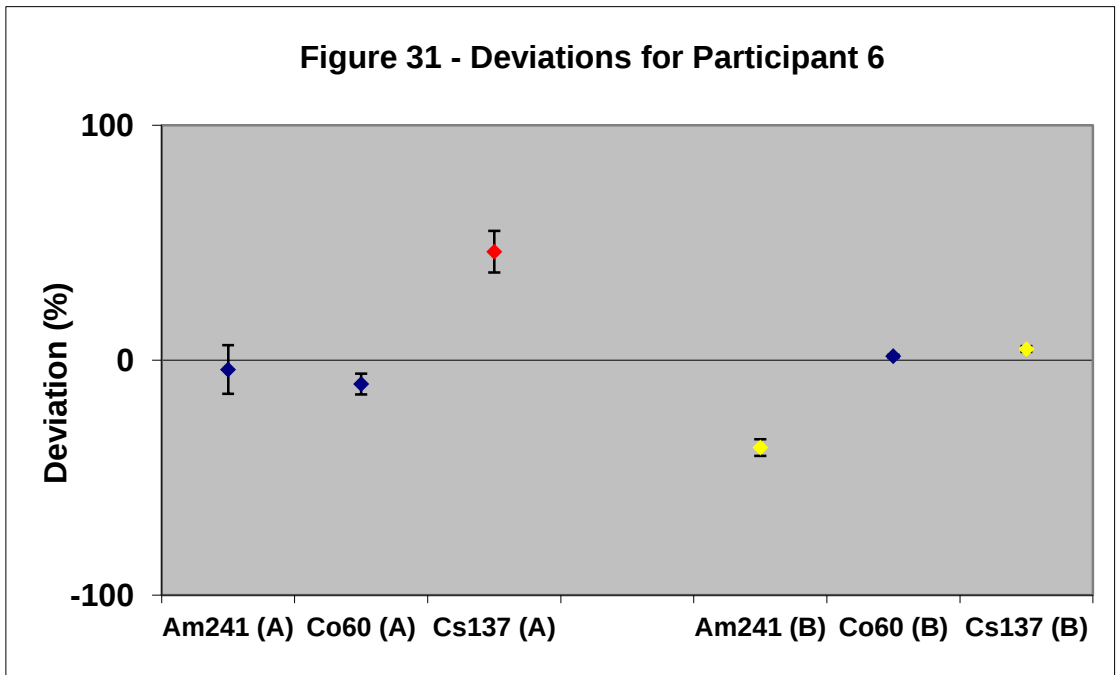
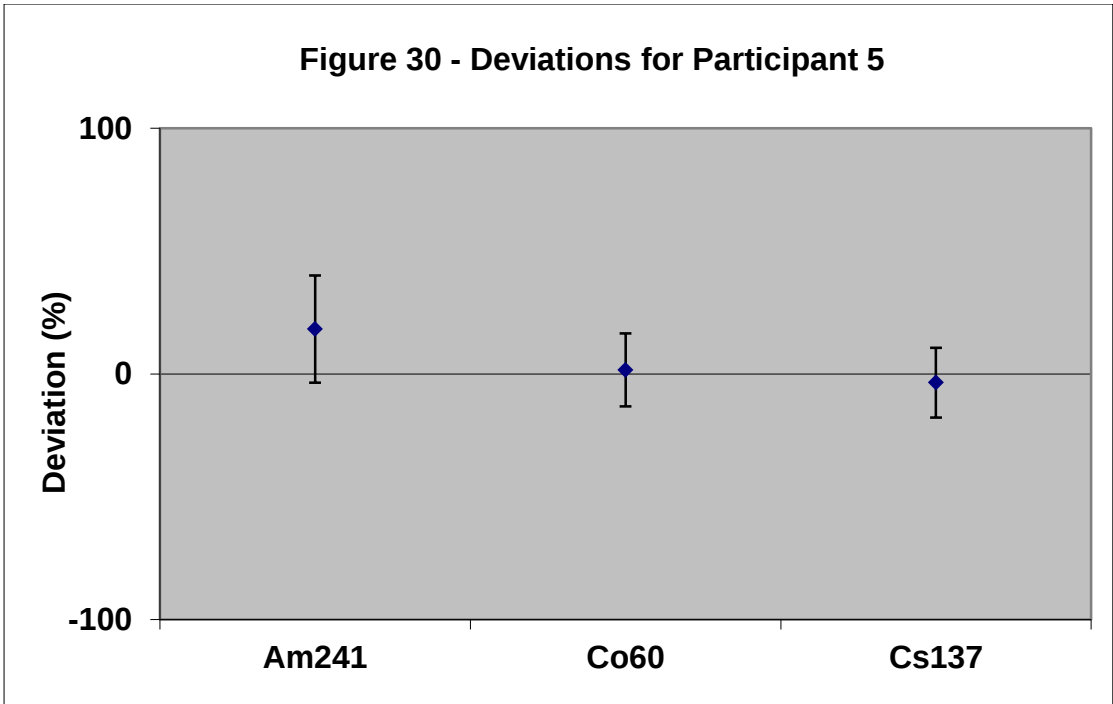
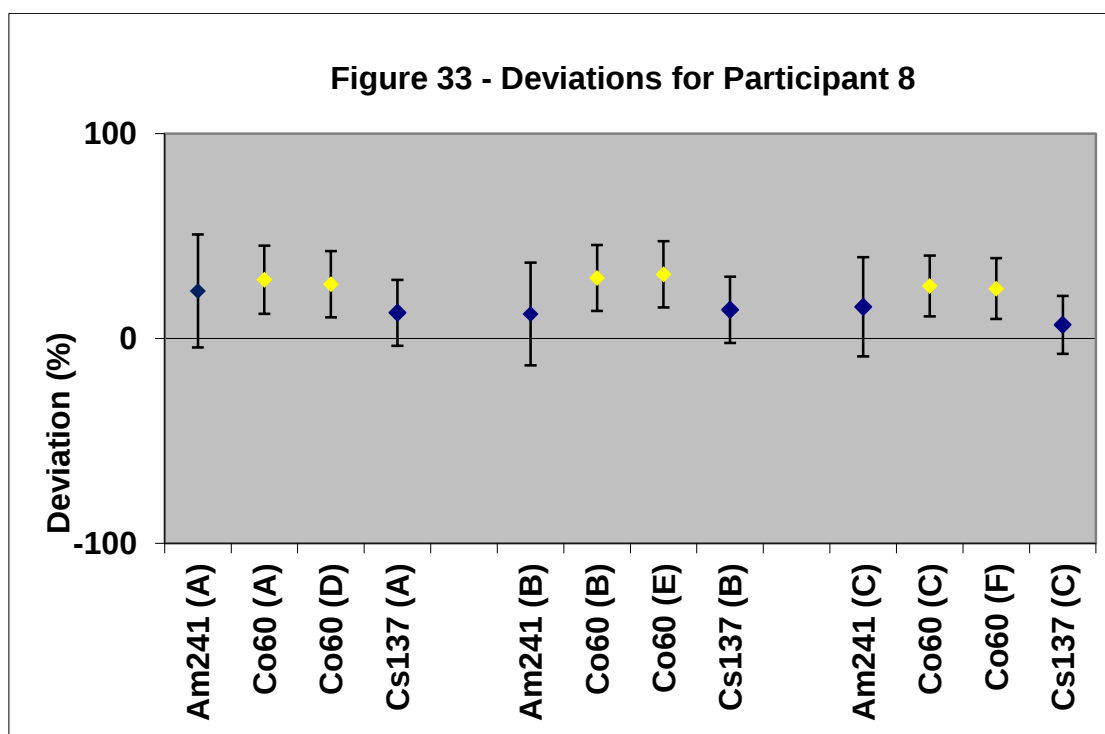
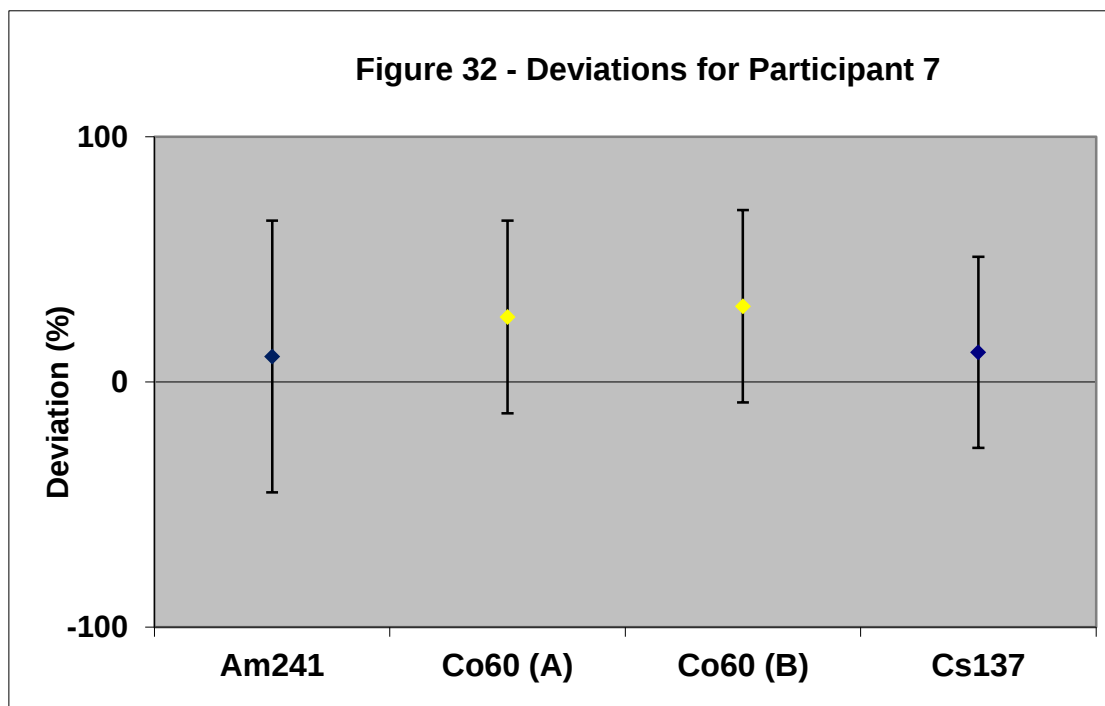
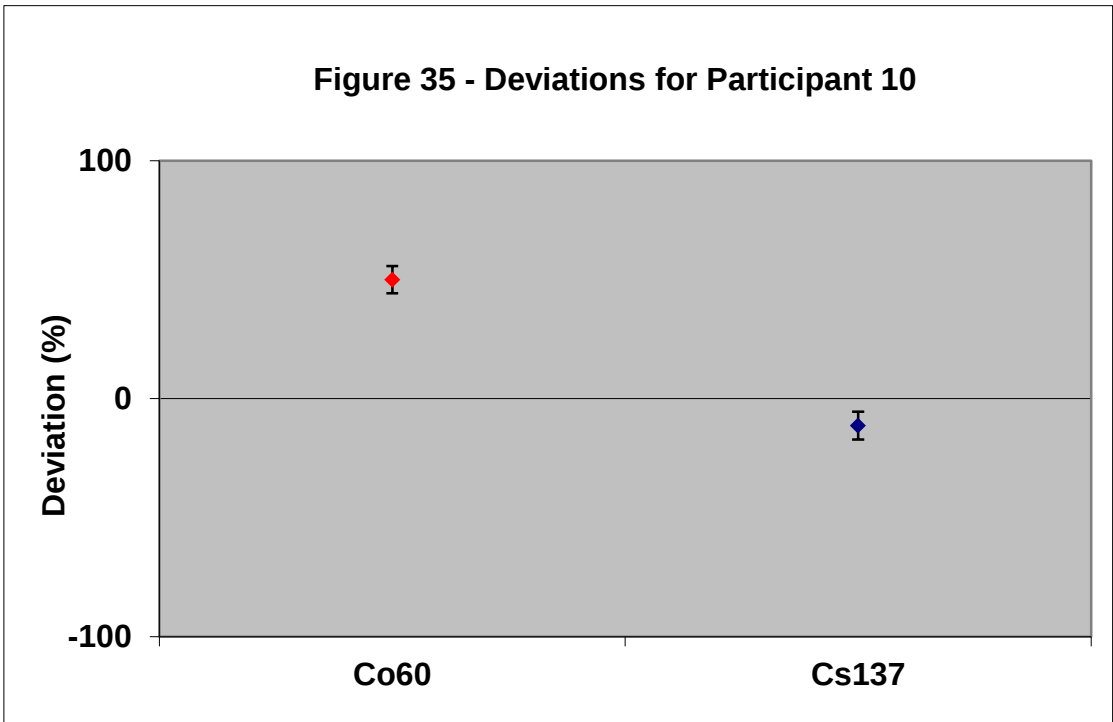
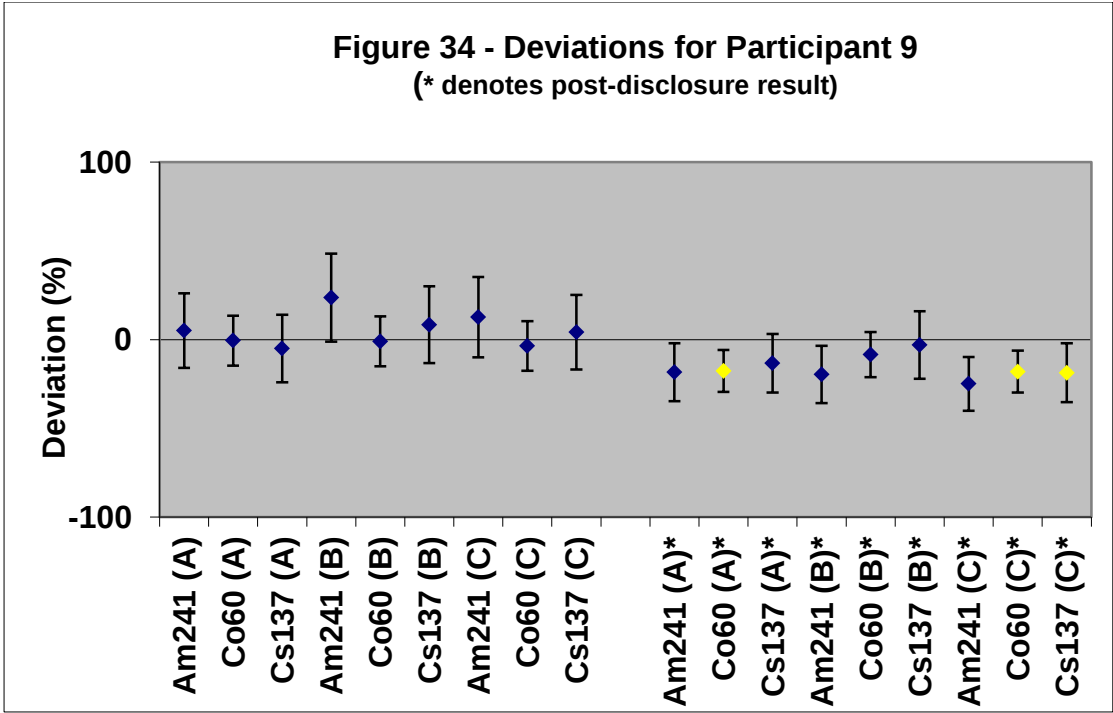


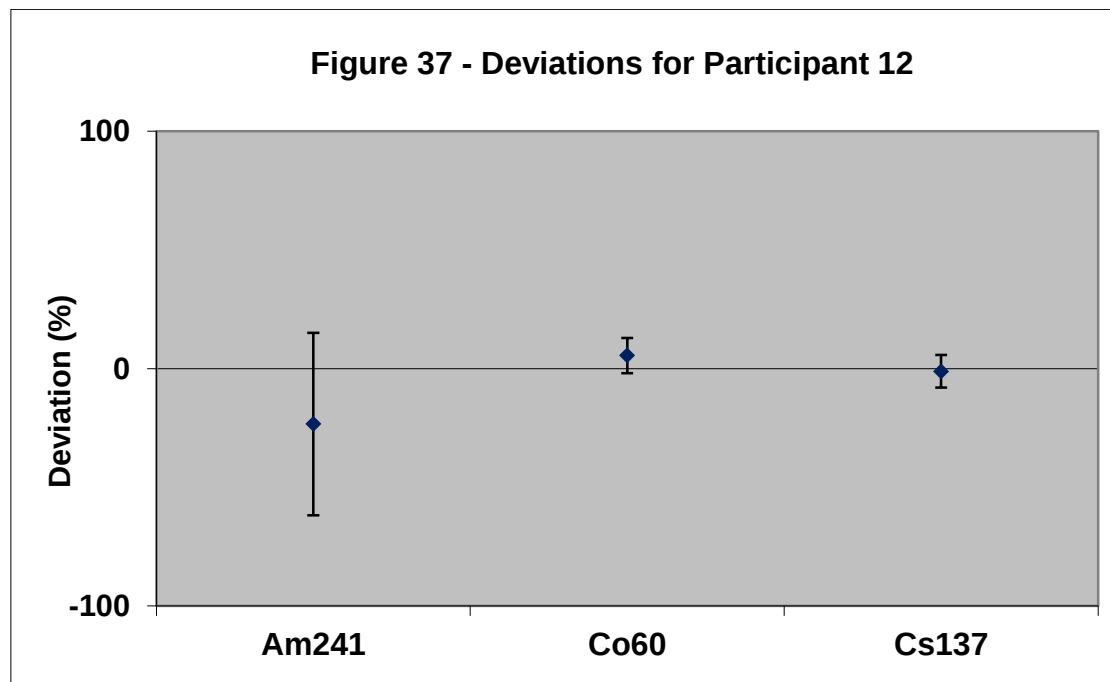
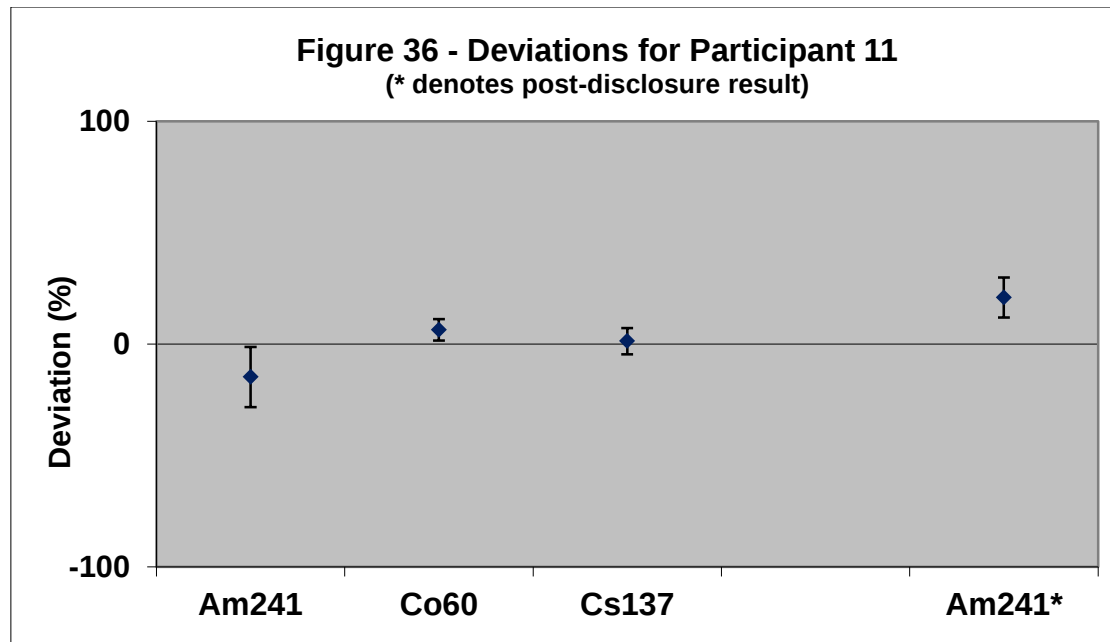
Figure 26 - Deviations for Participant 1**Figure 27 - Deviations for Participant 2**

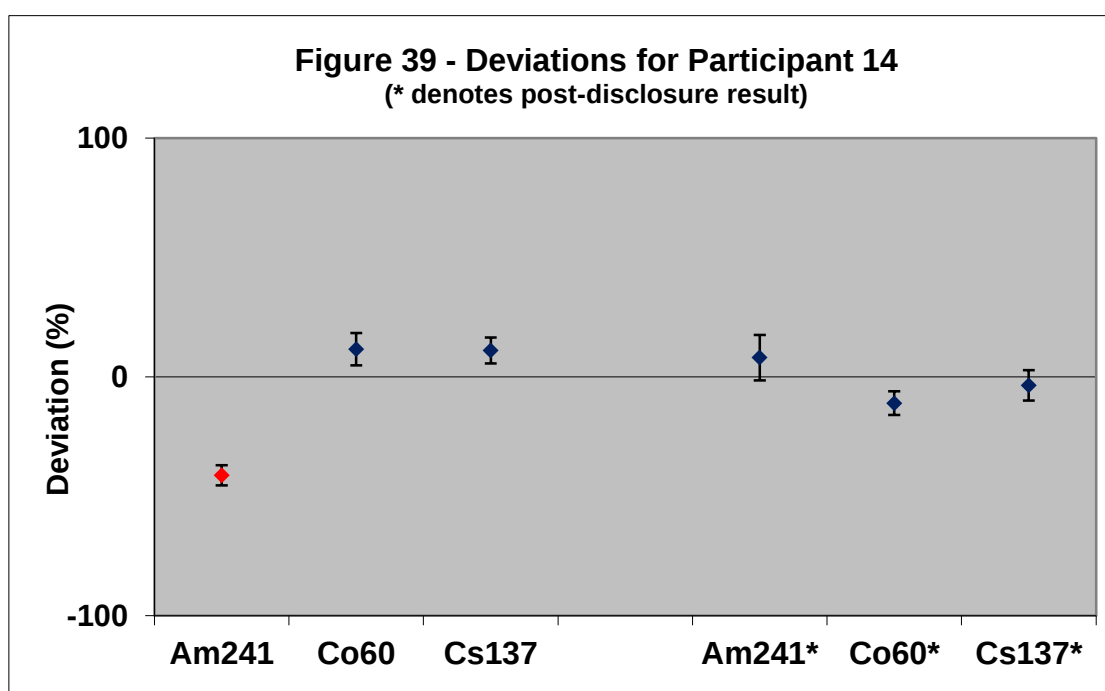
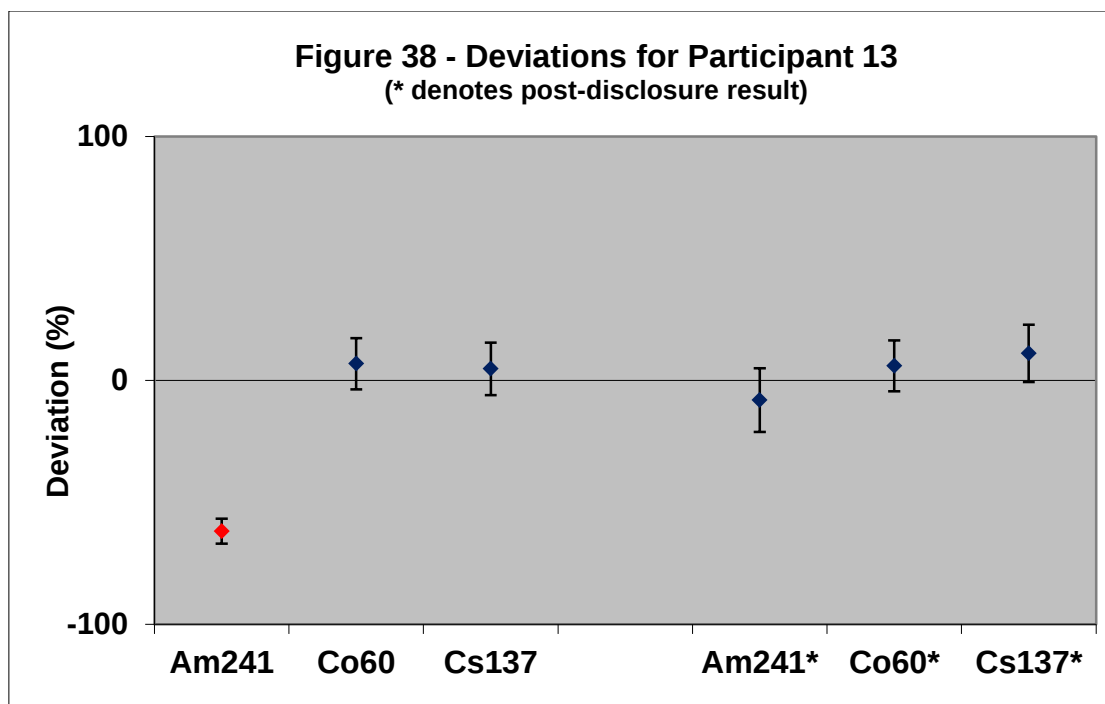


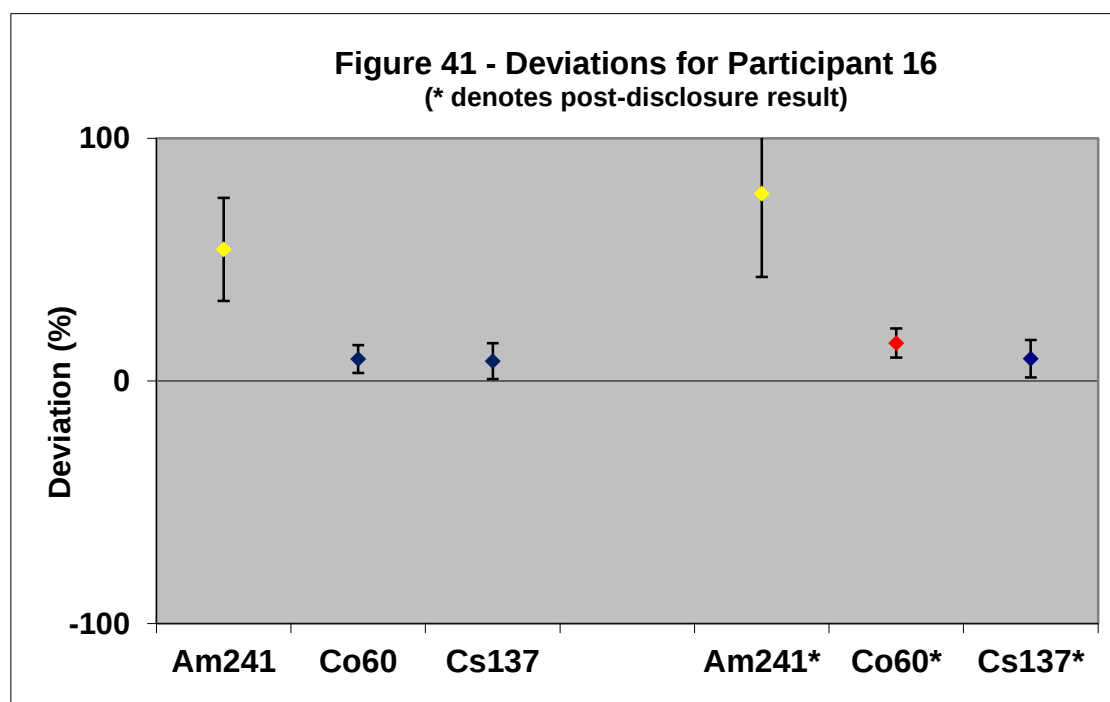
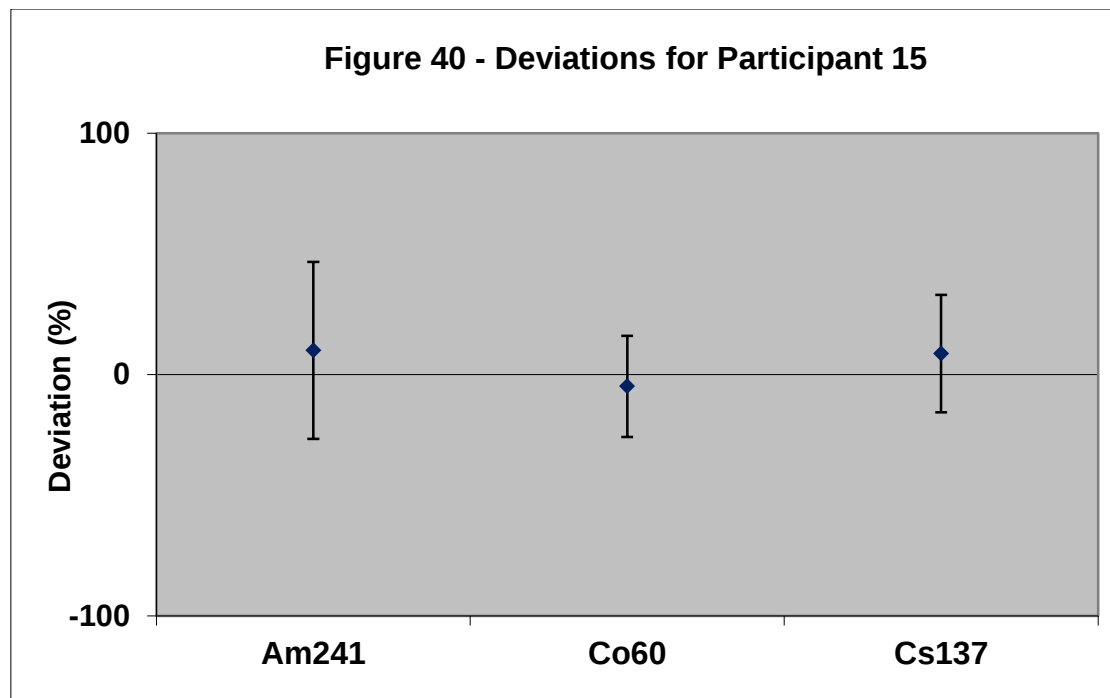












13 APPENDIX A – Initial Mailshot

Dear Colleague,

THIRD NPL NUCLEAR INDUSTRY PROFICIENCY TEST EXERCISE

In 2007 and 2009, NPL ran bulk-waste proficiency test exercises ('drum comparisons') to enable UK laboratories involved in decommissioning and site clearance to test their bulk-waste gamma measurement procedures. NPL prepared 200 litre drums (illustrated above, during loading) filled with HDPE bottles containing various matrices, some being spiked with accurately-known activities of radionuclides directly traceable to UK primary standards of radioactivity. These drums were circulated as 'blind samples' to the participants and their results were discussed at follow-up workshops. The reports on both exercises (NPL Reports IR2 and IR19) are available as free downloads from the NPL website (www.npl.co.uk).

I am now writing to invite you to participate in a third exercise. The purpose of the exercise is to provide the user community with a voluntary, independent and confidential test of their bulk waste measurement procedures.

In brief: NPL will prepare a single 'mock waste' sample in a 200-litre steel drum. The drum will contain metal, hard plastic and soft-waste material. The radionuclides ^{241}Am , ^{60}Co and ^{137}Cs will be present and the overall activity concentration (activity/total mass of material in drum) will be in the range $1 - 10 \text{ Bq g}^{-1}$. The activity will be distributed in a heterogeneous manner. The drum will be available for measurement between April and July 2011 and participants will be asked to report the activity concentrations of the individual radionuclides. A workshop to discuss the results will be held in October 2011 and a report will be published in December 2011. All results will be coded and treated as confidential.

If you are interested in participating, please read the full details given on pages 2 to 3 below and then complete and return the PTP Enquiry Form. Please note that requests to participate must be received by 11 March 2011. We hope you are interested and that you will be able to participate.

Yours faithfully

Julian Dean

February 2011

A) Plan for Third NPL Nuclear Industry PTE

NPL will prepare a single ‘mock waste’ sample in a 200-litre steel drum. The drum will contain metal, hard plastic and soft-waste material. The radionuclides ^{241}Am , ^{60}Co and ^{137}Cs will be present and the overall activity concentration (activity/total mass of material in drum) will be in the range 1 – 10 Bq g⁻¹. The activity will be distributed in a heterogeneous manner within the drum.

The drum will be delivered to (and collected from) the participants on agreed dates. Participants will be given 1 working day to measure the drum (excluding arrival and dispatch days). The drum will be an Excepted package.

Note that, prior to measurement by the participants, NPL will disclose only the above information plus:

- **the percentage by mass of each material present;**
- **the dimensions of the drum;**
- **the mass of the drum (empty and full).**

Participants will be asked to report their measurements of the individual radionuclide activity concentrations along with details of calibration procedures and instrumentation used, by a ‘first deadline’ (see Timetable below). NPL will then disclose:

- more details of the materials (e.g. which metal) and the ‘internal structure’ of the drum;
- the location(s) of the activity present.

Participants will then have the opportunity to submit (by a ‘second deadline’) additional results based on this more detailed knowledge of the drum. Note that revisions to earlier results (i.e. those submitted before the first deadline) will not be accepted during this period.

NPL will then prepare a draft report (including all results, coded for anonymity), issue it to all participants and run a follow-up one-day workshop to discuss the exercise and decide on a forward plan (e.g. another comparison or maybe investigations into any calibration or measurement problems arising). A final report will then follow.

B) Timetable

13.1.1 Action	Dates
Interested laboratories to reply to this mailshot	By 11 March 2011
NPL to contact each laboratory, advise if they can be accommodated, and, if so, agree receipt and dispatch dates	By 31 March 2011
Participants to submit Purchase Orders	Within two weeks of notification of receipt and dispatch dates
Participants to measure drum	Within agreed dates in the period April to July 2011 inclusive
Participants to submit results (<u>not knowing</u> details of drum contents)	By 5 August 2011 (First deadline)
NPL to declare further details of drum contents	8 August 2011
Participants to submit any additional results (<u>knowing</u> details of drum contents)	By 26 August 2011 (Second deadline)
NPL to disclose activity concentrations	30 August 2011
NPL to issue draft report	October 2011
NPL to hold one-day workshop	26 October 2011
NPL to issue final report	December 2011

C) How to participate

Please complete the enclosed Enquiry Form and return it by **11 March 2011** to:

radioactivity@npl.co.uk

Please indicate on the form which weeks in the period April to July you would prefer for delivery of the sample drum to your site. Note that you will be allocated one full working day to measure the drum (excluding the arrival and dispatch days).

IMPORTANT: If the exercise is oversubscribed we shall deal with requests for inclusion on a first-come-first-served basis, so please **PLEASE DO NOT SUBMIT A PURCHASE ORDER AT THIS STAGE**. We shall contact you in March to confirm whether or not we can accommodate you, and if so we will agree receipt and dispatch dates with you and ask you to submit your order at that time. Participants must ensure Purchase Orders are submitted within two weeks to ensure that they receive the drum on the allocated date.

D) Fees

The fees will be as follows:

- Participation (including loan of drum and workshop attendance): **£875**
- Delivery: **To be advised**

NPL will arrange for a courier to deliver and pick-up on the agreed dates.

14 APPENDIX B – Reporting form

NPL 200 L DRUM COMPARISON EXERCISE 2011 – RESULTS

Name:

--

Organisation:

--

Date of submission of results:

--

Details of measurement method (please include, e.g., detector type/model, source-to-detector distance, background correction method, counting time, scan pattern, collimation, density correction routine, etc.)

--

Calibration method (please include, e.g., standard sources used, traceability of standards, uncertainties on standards, any geometrical correction factors used, all associated uncertainties, etc.)

--

Efficiency models (please include, e.g., details of any software used, and any validations carried out)

--

Any other information or comments

--

NPL 200 L DRUM COMPARISON EXERCISE 2011 - RESULTS

Nuclide: **Am-241**

Reference time 1 March 2011 12:00 GMT

	(Bq/g)
Activity concentration	
Combined standard uncertainty ($k=1$)	

Uncertainty budget components	Relative uncertainty (%)
Uncertainty component associated with net count rate of nuclide	
Uncertainty component associated with detector counting efficiency	
Uncertainty component associated with emission probability	
Uncertainty component associated with decay/ingrowth correction	
Other uncertainty components (please specify)	
Combined standard uncertainty	

Nuclear data used	
Half-life (days)	
Emission probability (%)	keV
Emission probability (%)	keV

Limit of Detection	
--------------------	--

NPL 200 L DRUM COMPARISON EXERCISE 2011 – RESULTS

Nuclide:

Co-60

Reference time 1 March 2011 12:00 GMT

	(Bq/g)
Activity concentration	
Combined standard uncertainty ($k=1$)	

Uncertainty budget components	Relative uncertainty (%)
Uncertainty component associated with net count rate of nuclide	
Uncertainty component associated with detector counting efficiency	
Uncertainty component associated with emission probability	
Uncertainty component associated with decay/ingrowth correction	
Other uncertainty components (please specify)	
Combined standard uncertainty	

Nuclear data used	
Half-life (days)	
Emission probability (%)	keV
Emission probability (%)	keV

Limit of Detection	
--------------------	--

NPL 200 L DRUM COMPARISON EXERCISE 2011 – RESULTS

Nuclide: Cs-137

Reference time 1 March 2011 12:00 GMT

	(Bq/g)
Activity concentration	
Combined standard uncertainty ($k=1$)	

Uncertainty budget components	Relative uncertainty (%)
Uncertainty component associated with net count rate of nuclide	
Uncertainty component associated with detector counting efficiency	
Uncertainty component associated with emission probability	
Uncertainty component associated with decay/ingrowth correction	
Other uncertainty components (please specify)	
Combined standard uncertainty	

Nuclear data used	
Half-life (days)	
Emission probability (%)	keV
Emission probability (%)	keV

Limit of Detection	
--------------------	--

15 APPENDIX C – List of Participants

Dr H Beddow
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Didcot
Oxfordshire OX11 0TQ

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Magnox Ltd
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Mr S Defour
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NPL Report IR 25

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Mr A Wilson
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Dounreay Site Restoration Limited
Dounreay
Thurso
Caithness KW14 7TZ

Mr A Wright
Magnox Ltd
Dungeness A Site
Romney Marsh
Kent TN29 9PP

16 APPENDIX D – Minutes of post-exercise workshop, 26 October 2011

Participants

Helen Beddow	Nuvia Ltd
Julian Dean	NPL
Andrew Frith	Serco
James Gregory	G E Healthcare
Paul Harding	AWE
Martin Jackson	G E Healthcare
Cyrus Larijani	NPL
Bob Major	AMEC
Sanders McDonald	DSRL
Chris McKay	Serco
Tim Miller	AWE
Daniel Parvin	Babcock International Group
Martin Rushby	Canberra UK Ltd
Michelle Skelland	RSRL
Gareth Ward	Magnox Ltd

Welcome, domestic issues, introductions and agenda – Julian Dean, NPL

The Chairman welcomed the delegates to the meeting. The main objectives would be (i) to identify any measurement issues and possible solutions, and (ii) to identify preferences for the format of the ‘sample’ for the next PTE in this series. In relation to objective (ii), it was not yet clear whether funding would be available for an exercise in 2012 and it was estimated that the project may cost in the region of £4-5k per participant as a ‘ballpark’ figure and that funding would be a potential issue. However, this number was tentative as it would depend on the number of participants.

Nuclear Industry PTE - background, source preparation, results and discussion – Julian Dean

The PTEs are important to the participants and provide a useful reference standard. The difference in performance of the various detectors was of interest; the participants wanted to see more patterns and trends, especially the data gathered from the SGS detectors. From the discussion it was apparent that most participants used a single germanium detector and many labs did not use collimated detectors. It was noted that AWE’s NDA group had used a few different detectors and that all should be noted in the final report.

The fact that NPL had standardised the drum for individual radionuclides was seen as a positive, although the fact that NPL did not (due to not having a suitable drum scanner) measure the drum itself was seen as a negative. Also it was seen as a legitimate and worthwhile point.

The results yielded from the SGS detectors were sometimes low, and possibly the active areas within the drum were being ‘split’ between adjacent segments. This was of particular concern due to the potential underestimation of activity. When measuring a real sample, this could potentially result in a laboratory unintentionally consigning more activity than they have declared.

It was felt that the uncertainties quoted by some participants were too large. It was noted that Participants 2 and 10 both used SGSs but their results differed significantly.

It was noted that the data inputted to the software was critical.

Participant presentations

Tim Miller (AWE) presented their measurement methods, the reasons for measuring drums, typical drum samples, performance levels achieved, the reasons for participating in proficiency tests, the results of EA and NPL tests, and what AWE would like to see from future tests. The NPL PTEs are key for maintaining UKAS accreditation and AWE are very supportive of them. Regarding future exercises, a more challenging drum is required (e.g. other radionuclides could be added, or perhaps steel). **Paul Harding** (AWE) explained the NDA group's measurement methodology for the NPL drum. A combination of techniques were used to measure the drum, similar to the methods used in earlier PTEs. Radiography established the overall internal structure and a hand-held NaI(Tl) detector and a gamma imaging system were used to locate the 'hotspots'. The drum was then assayed using a hyperpure Ge crystal at different source-to-detector distances.

Daniel Parvin (Babcock) explained the Particle Swarm IMaging (PSIM) method of analysis used to measure the NPL drum, and compared it with 'conventional' Ge-based methods. Additional uncertainty analyses had been carried out since the PTE finished, and these were included. One delegate asked about data processing time for PSIM. This is of the order of 6 minutes (plus the actual measurement time). **Michelle Skelland** (RSRL) explained the SGS system used at Winfrith, its operation and calibration. It is automated and uses NDA 2000 software. The system measures ^{137}Cs and ^{60}Co as 'primary' radionuclides and other radionuclides are inferred from fingerprint. The system will be used in future to determine whether material is VLLW or Out Of Scope. **Chris McKay** (Serco) outlined their approach to measuring the drum. They participated in the PTE to conduct a blind test, to validate in-house Ge spectroscopy methods, and to experiment and develop techniques.

AOB – Julian Dean

The Chairman mentioned the upcoming 'Metrology for Decommissioning and Site Clearance' Workshop to be held at NPL on 11th January 2012 and raised the question of where we go from here. The main points raised were:

- The report should compare methods used (e.g. open geometry v SGS) and ISOCS and other modelling techniques should be taken into consideration.
- More work is needed to explain why ^{241}Am data are so different from ^{60}Co .
- The weight of the drum was a vital piece of information, but note that some participants do not routinely include the empty drum mass in their activity concentration calculations.
- Perhaps in future PTEs, participants could be required to measure total activity, or total gammas.
- The participants wanted a much more challenging drum with potentially uranium being involved. However, this may cause problems with transport or acceptance onto some sites. Concrete should be included if possible. The overall concentration should be of the order of $1 - 10 \text{ Bq g}^{-1}$.
- Participants wanted more time to measure the drum (perhaps two full days).
- The maximum mass of the drum should be 100kg.

Cyrus Larijani
Julian Dean

26 October 2011

17 APPENDIX E – Participants’ presentations from workshop



Waste Drum Assay at AWE

Timothy Miller
 Timothy.Miller@awe.co.uk
 www.awe.co.uk



OVERVIEW

- Measurement method
- Reasons for measuring drums
- Typical drums
- Performance achieved
- Reasons for participating in proficiency tests
- EA and NPL test results
- Future tests



MEASUREMENT METHOD



3



HARDWARE AND SOFTWARE

- HRGS hardware (no transmission source)
- HPGe N-Type (6 x 8 cm)
- Flush collimated (Pb/Cu)
- Digidart MCA
- Maestro 32
- SNAP

4



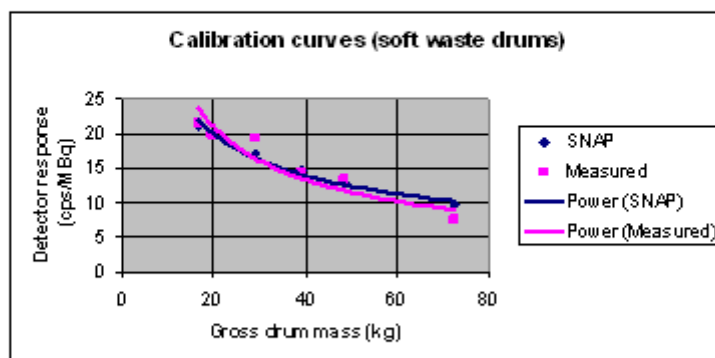
SNAP OPERATION

- Import Maestro ROI report file into SNAP
- Identify nuclides and save as RPu file
- Input modelling parameters
- Calculate activity
- DPA
- Pu and U lump correction (EA drum)
- Generate report
- Alternatively generate calibration curves

5



SNAP GENERATED Am-241 CALIBRATION



6



REASONS FOR MEASURING DRUMS

- Cost savings (ILW V Drigg LLW)
- Criticality safety
- Nuclear material accountancy (DU firing trials)
- Avoid sending VLLW/EW to Drigg
- Storage and transport regulations

7



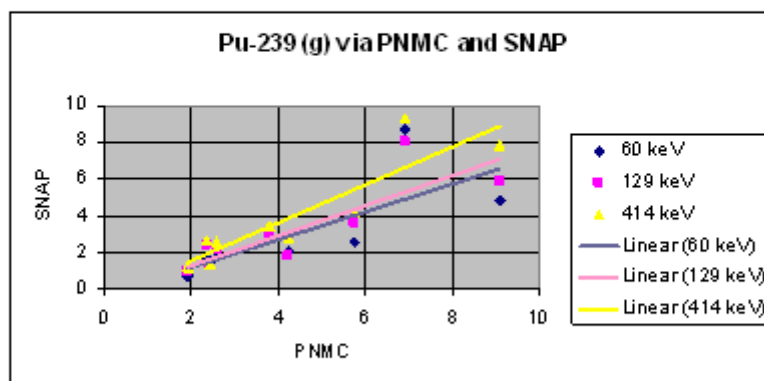
TYPICAL DRUMS

- 60 kg gross (average)
- Soft, hard or mixed
- Firing trial drums up to 300 kg
- DU up to several kg
- HEU, Pu sub mg to sub kg
- Fission products sometimes present
- SNAP UKAS accredited for full range

8



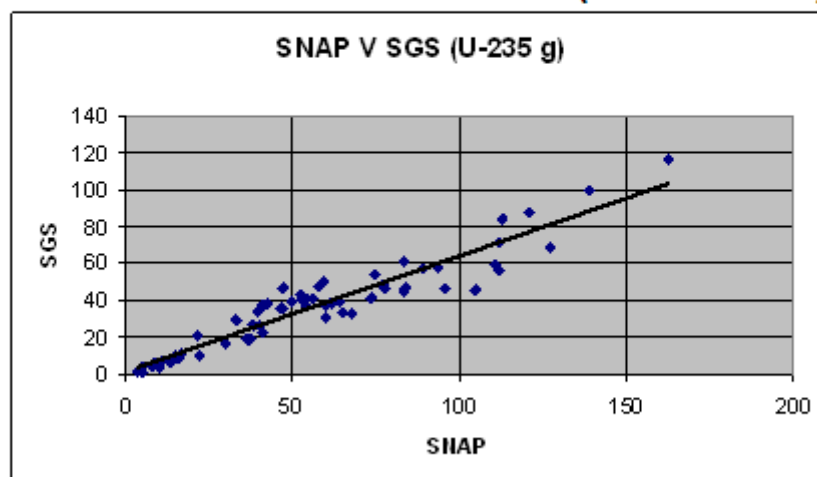
PERFORMANCE ACHIEVED (Pu/steel)



9



PERFORMANCE ACHIEVED (HEU/mixed)



10



REASONS FOR PARTICIPATING

- Achieving UKAS
- Maintaining UKAS
- Customer confidence
- EA confidence (EA drum mandatory)
- Gauge performance against other sites

11



TEST RESULTS (% OF TRUE VALUE)

Drum	Am-241	Cs-137	Co-60	U-235	U-238
NPL07	81	96	97	-	-
NPL09A	94	105	101	-	-
NPL09B	74	94	97	-	-
NPL11	104	107	113	-	-
EA10	-	-	-	100	104

12



EA DRUM

- Prepared by WQCL (Winfrith)
- 20 kg polythene shavings
- Plastic tubes/vials containing 360 g NU
- WQCL SGS (UKAS) underestimated:
 - 26 % of U-235 (SNAP 100 %)
 - 92 % of U-238 (SNAP 104 %)
- SNAP LC routine essential for U-235

13



FUTURE DRUMS

- NU, DU, or HEU plus Am-241, Cs-137, Co-60
- More typical matrix mixture
- Gross mass around 60 kg
- Surface contaminated steel
- No activity hidden in metal boxes (unrealistic)
- 2012?

14

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AWE NDA Team Measurement Methodology for the 2011 NPL Radioactivity Proficiency Test Exercise

Paul Harding

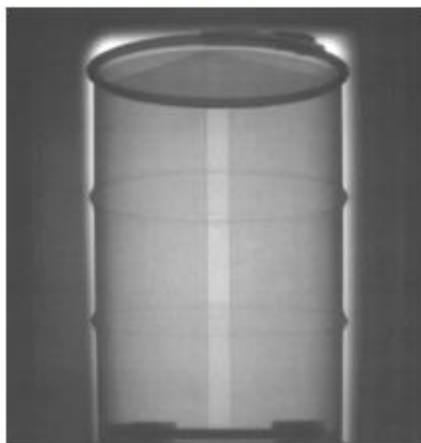
paul.harding@awe.co.uk

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Radiography



- Radiography performed upon arrival at AWE.
- 'Box' shaped object filling the height of the drum identified from the recorded images.

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2



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Transmission Measurements



- Canberra UK High Resolution Segmented Gamma-Ray Scanner (HRSGS).
- Shielded and collimated HPGe detector. Vertical translation throughout the height of the rotating drum, effectively divides the drum into 8 discrete horizontal segments.
- A Eu-152 transmission source is positioned diametrically opposite the detector and which incorporates a shielded shutter.
- At each segment the source shutter is opened and the measured Eu-152 peak information compared with reference data to provide a matrix correction factor as a function of gamma-ray energy.

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3



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Handscan



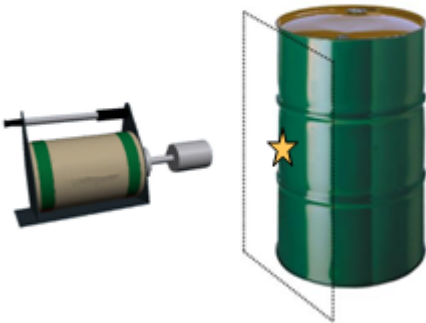
- ICX Technologies Identifinder used to perform a hand-scan of the drum.
- 36 mm dia. x 51 mm NaI (TI) crystal
- Hand-scan technique used to determine the 'hot-spots' or 'hot-face' in order to optimise the positioning of follow-on HPGe and Gamma Camera instrumentation.

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4


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HPGe Measurements



- Ortec 110% LN2 Cooled HPGe Detector.
- Lead collimator fitted to reduce the observed local background signal.
- Detector aimed at the mid-point (height) of the drum.
- Measurements performed at both the 'hot-face' and the opposing 'cold-face' at distances of 1m and 3m.

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Gamma Camera



- Gamma Ray Imaging Spectrometer (GRIS) built by LLNL.
- Coded Aperture Design.
- Gamma Detector ~ CsI(Na) [10 x 100 mm].
- Energy Range 30 to 1250 keV.
- 4 Energy Gates.

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Gamma Camera



Energy discrimination

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7

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Analysis

- Solving for an unknown from 4 measurements (2 pairs of measurements at 180°).
- Corrections made for the instrument stand-off and the attenuation of gamma-rays by both the matrix and the drum walls.
- Assumptions
 - HPGe detector stand-off sufficient to treat the unknowns as a point sources.
 - HRSGS transmission measurements representative of the real matrix attenuation encountered during HPGe determinations.

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8

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Analysis Calculated distance into drum from the 'hot-face'			
Isotope	Am-241	Co-60	Cs-137
Distance into Drum (cm)	-7.9	10.4	47.9
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9			

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Am-241 - Transmission measurements performed using a Eu-152 source over an energy range 122 to 1408 keV. Extrapolation required in order to determine the transmission for the Am-241 gamma-ray at ~60 keV. - Measurements performed in a working facility containing a large number of waste drums that included quantities of fissile material. The operational requirement to move waste drums during the measurement campaign resulted in a dynamic background.	
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10	

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Conclusions ▪ Agreement with the declared activity values for Am-241, Co-60 and Cs-137 at the 95% confidence level. ▪ Methodology worked particularly well for Cs-137. ▪ Areas of improvement identified which may be applied to similar measurement scenarios in the future. ▪ Radiography and Gamma Camera shown to be a valuable complementary techniques supporting the gamma-ray spectroscopy measurements.	
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11	

 Research Sites Restoration Ltd <i>The Segmented Gamma Scanner (SGS) at RSRL Winfrith</i> <i>Michelle Skelland</i>	
 Research Sites Restoration Ltd	
1	

The Winfrith SGS



- Used to measure the activity of low level waste in standard 220 l drums
- Utilises automatic loading and unloading via a conveyor system
- Has a transmission source for matrix attenuation measurement
- Can be used in "batch" or fully automatic mode by use of a barcode reader

The Winfrith SGS – Hardware – Mechanism



- Each 220 l drum is analysed in 4 segments
- The drum rotates on a turntable during counting to even out the effects of uneven activity distribution within the drum
- Each drum is loaded and unloaded via a conveyor system, of which the turntable forms an integral part
- The turntable includes a load cell to measure the mass of the drum

The Winfrith SGS – Hardware – Mechanism



The detector (housed within a lead collimator) moves to each segment using a motor and leadscrew



The transmission source is mechanically linked to the detector lift by cable and pulleys



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4

The Winfrith SGS – Hardware – Mechanism

The whole system is controlled by PLC (Programmable Logic Controller)

- Commands from and status to NDA 2000
- Manual control panel



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The Winfrith SGS – Hardware – Detector



- The system uses a high purity germanium (Ge) detector
- Liquid nitrogen (contained in a dewar) is used to keep it cold
- It requires a high voltage bias (a few thousand volts)
- A solid lead collimator assembly provides all round shielding for the detector crystal
- The collimator restricts the detector field of view to a single quarter of the drum segment

The Winfrith SGS – Hardware – Transmission Source

- The transmission source is Eu-152 (59.2 MBq on 1 October 1993)
- Half-life = 13.3 years
- The range of main gamma energies of Eu-152 covers the typical gamma energies within waste produced at RSRL Winfrith

Energy (keV)	Intensity (%)		Energy (keV)	Intensity (%)
121.78	28.4		867.32	4.2
244.69	7.5		964.01	14.4
344.27	26.5		1085.78	10
411.11	2.2		1112.02	13.3
443.98	2.8		1407.95	20.7
778.89	13			

The Winfrith SGS – Calibration

- The system is subject to energy/resolution, transmission and efficiency calibration procedures on an annual basis
- The energy/resolution and efficiency calibration procedures are performed with a Eu-152 source (63.2 MBq on 1 October 1993)
- The transmission calibration procedure is performed with the in-situ Eu-152 transmission source
- The load cell on the turntable is calibrated on an annual basis



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The Winfrith SGS – Software

- The system uses NDA 2000
- It is typically operated in automatic (continuous) mode and requires minimal operator input
- The system waits for a drum to arrive on the turntable and then reads the barcode on the drum label
- The barcode provides the system with both the unique drum ID and the waste stream (fingerprint)
- A drum dose-rate is also measured



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The Winfrith SGS – Software

- Before each drum assay, a background count in Segment 1 is performed
- This data is used to correct the subsequent analysis data in each of the 4 segments
- A “Verifier Drum” measurement must be performed every 7 days and the system will refuse to function without this check measurement
- The typical count times per segment for Winfrith LLW drums are:
 - Background counttime = 180 seconds
 - Transmission counttime = 60 seconds
 - Segment counttime = 180 seconds
 - Total counttime = 19 minutes



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The Winfrith SGS – Software

- The standard drum report shows the radionuclides that have been detected
- The results are reported in terms of Activity (MBq) and Specific Activity (MBq/Te) for the whole drum
- The main radionuclides that are detected within Winfrith waste are Cs-137 and Co-60 and these are called the “Primary Nuclides”
- The “Secondary Nuclides” are then calculated by using the relevant fingerprint for a particular waste stream
- In general, fission products are determined by ratio to Cs-137 and activation products are determined by ratio to Co-60



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The Winfrith SGS – Future Use

- The SGS will be used in future to determine whether material may be sentenced as VLLW or exempt (out of scope)
- In order to achieve a higher level of confidence in the activity results, longer count times in the segments will be used

Bringing service to life

serco



Use of the PTE for Part Validation of Serco Radiation Services In-Situ Assay Service

Presentation to
Nuclear Industry PTE Workshop

Presented by
C McKay

25th October 2011

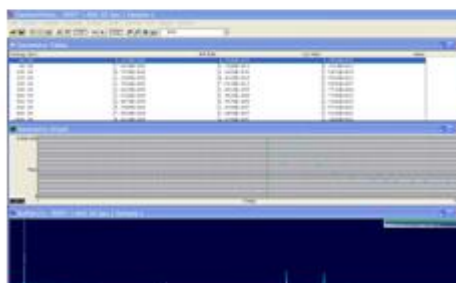
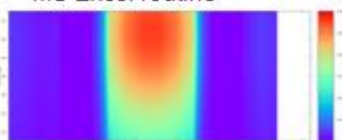
Serco's Objectives

- Serco have NaI and HPGe Gamma spectroscopy capability.
- Principally used for in-situ, investigative and environmental measurements
 - Measurement of vessel/pipe contents etc..
 - Characterisation of unidentified objects
 - Environmental surveys
- Rarely measure drums with HPGe
 - Portable HPGe systems not ideal for routine waste monitoring due to complexity and set up time.
 - Main benefit of HPGe is resolution, allowing identification of radionuclides and separation of sample from background.
- However, a drum is just a small vessel, so similar to in-situ work
- Serco's objectives for the PTE
 - Convenient opportunity to conduct blind test of measurement capability
 - Useful in validating our HPGe spectroscopy methods against industry standards
 - Opportunity to experiment and develop internal techniques

serco

The Equipment

- Ortec Portable Gamma Gage
 - n-type HPGe detector
 - 55% relative efficiency
 - Aluminium cryostat casing
 - Carbon fibre low energy window
- Analysis
 - Spectroscopic analysis: GammaVision
 - Modelling: ISOTOPICS
 - Regression analysis with custom MS Excel routine



Analysis Methods

- The estimated activity from gamma spectroscopy is highly dependant on the assumptions used in the computational modelling.
- Initial gamma survey of the drum identified significant anisotropy in the gamma flux around the drum
- So for the pre-disclosure analysis we decided to test two differing approaches
 1. Assumption of single point source located at centre of drum (conservative option)
 2. Allow model parameters to be inferred from correlation with measurements
- We chose to report results from option 2, as these were judged to be best estimate.



Option 1: Centred Source

- Measurements made of drum as rotated through different angles
- Geometrical and attenuation correction made on assumption that source is located at the centre of a drum with a homogeneous contents.
- Activity calculated as mean of all measurements
 - Measurements must be geometrical symmetrical (or rotate fast relative to measurement duration) for variations to counter each other.

Radionuclide	Measured Activity Concentration	Actual Activity
Am-241	7.8 (± 3.5) Bq/g	5.7 Bq/g
Cs-137	2.43(± 0.41) Bq/g	2.29 Bq/g
Co-60	2.91(± 0.28) Bq/g	2.05 Bq/g

- Measurements are conservative but still reasonably accurate
 - Uncertainty on Am-241 is large and could make a significant difference to waste measurement



Option 2: The Model

- The observed intensity changes with position of the detector
- Can use a simple equation to predict observed count rate

$$I = \left[A \varepsilon(E_\gamma) \right] \left[\frac{R_C^2}{R_S^2} \right] \left[e^{-\left(\sum_i R_i \mu_i(E_\gamma) \right)} \right]$$

Efficiency Correction Geometry Correction Attenuation Correction

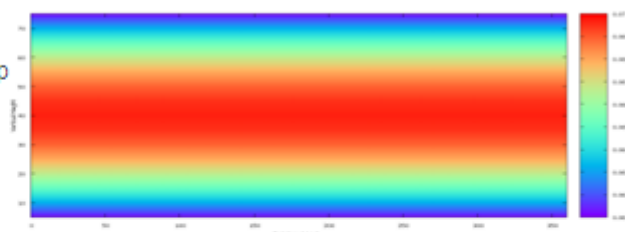
- A = Source Emission Rate
- $\varepsilon(E_\gamma)$ = Detector standard efficiency
- R_C = Calibrated source detector distance
- R_S = Source detector distance
- R_i = distance through material i
- $\mu_i(E_\gamma)$ = linear attenuation coefficient for material i

- By using simple computational methods allows rapid recalculation
- By iteratively adjusting modelling parameters, comparison of the resulting model predictions against multiple measurements allows unknown modelling parameters to be determined.

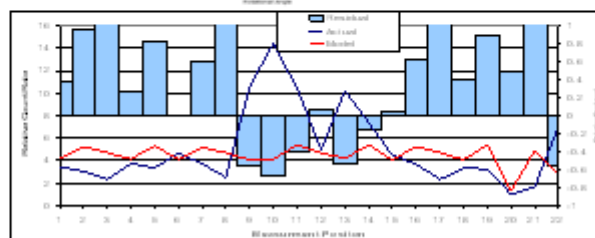
serco

Option 2: Comparison with the Model

- Cs-137 Relative photon intensity map around drum



- Comparison of measurement Vs model predictions



- With an assumed point source at the centre the correlation is rubbish!
– Hardly surprising

Option 2: Solving the Problem

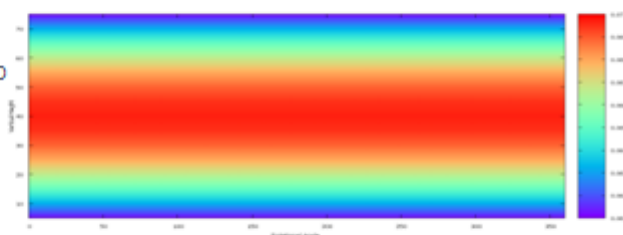
- We allowed certain modelling parameters to 'float', such as source position, and used a non-linear gradient reduction regression to derive those parameters.
- How does this work...
 - First define a scoring system to quantify the 'goodness of fit'
 - ▶ Sum Square of Residuals (weighted/unweighted)
 - Metrologist makes an initial guess at likely parameters
 - Computer introduces variation and observes relative change in 'goodness of fit'
 - Based on the relative change an estimate for the best parameters is made
 - Repeated with each estimate used as the initial guess for the next iteration.
 - The process stops when the system converges on a solution and the change from one iteration to the next is insignificant.

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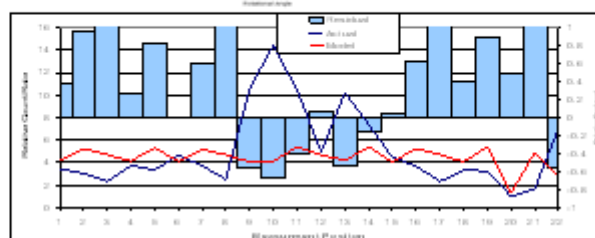
Option 2: Solving the Problem

- Allowing number of sources and their position to be determined by regression we go from the following to get ...

- Cs-137 Relative photon intensity map around drum



- Comparison of measurement Vs model predictions



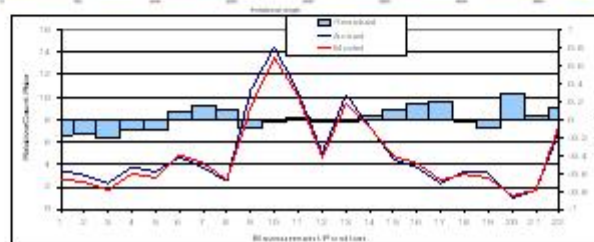
Option 2: Solving the Problem

- ... a much better correlation
- And knowledge of likely source location

- Cs-137 Relative photon intensity map around drum



- Comparison of measurement Vs model predictions



Option 2: Results

- For the different radio nuclides the following results were obtained

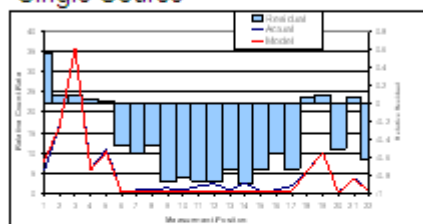
Radionuclide	Measured Activity Concentration	Actual Activity
Am-241	2.18 (± 0.29) Bq/g	5.7 Bq/g
Cs-137	2.15(± 0.22) Bq/g	2.29 Bq/g
Co-60	2.45(± 0.24) Bq/g	2.05 Bq/g

- Marginal improvement in accuracy and precision of Cs-137 & Co-60
 - Also allowed source locations to be determined within ± 2 cm within the drum
- Am-241 is way off, what happened there?

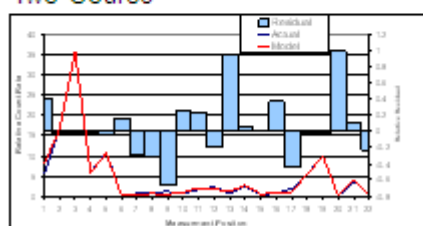
What happened with the Am-241?

- The inhomogeneity in attenuation within the drum results in a distortion of the measurement results.
 - Most severely impacted Am-241 due to large attenuation of its low energy gamma emission
 - Result is that regression fitting suggested **two** point sources
- However, as we later found there was only one source
- Post disclosure it was clearly a result of attenuation from the two metal sheets in the drum
- Comparison of fixing model to 1 or 2 sources
 - One source = $7.68(\pm 1.1)$ Bq/g
 - Two source = $2.18(\pm 0.29)$ Bq/g
 - Actual = 5.7 Bq/g

Single Source



Two Source



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Post disclosure results

- Post disclosure we knew precise source locations and the structure of the drum contents.
- The attenuation models were updated
- Unsurprisingly, the results are much improved with deviations of <11%
- Following disclosure, it was clear that our pre-disclosure results had identified the location of the Cs-137 and Co-60 sources within ± 2 cm and the Am-241 within ± 15 cm

Radionuclide	Measured Activity Concentration	Actual Activity
Am-241	$5.24 (\pm 0.74)$ Bq/g	5.7 Bq/g
Cs-137	$2.43 (\pm 0.24)$ Bq/g	2.29 Bq/g
Co-60	$2.28 (\pm 0.24)$ Bq/g	2.05 Bq/g

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Conclusions about our measurements

- Iterative determination of model parameters could be useful for simple geometries. But easily confounded by modelling inaccuracies
 - Accurate input data for modelling is vital
- Iterative modelling has potential to increase precision
 - Comparison of attenuation of different gamma rays from same nuclide remains useful.
 - Useful in providing check of assumptions
- Simple is usually the best
 - For most in-situ measurements the key is to know whether it meets an action level threshold
 - Knowledge of the activity is relatively unimportant if criteria are met.
 - For most applications simple conservative assumptions are all that's needed.
- Complexity usually only warranted when trying to increase precision

Comments on NPL PTE exercise

- Very useful exercise
 - Assisted with internal validation, provides confidence that our methods work
 - Allows comparison with others in industry
 - Useful excuse to experiment a bit.
- Useful for industry to assess confidence in measurement techniques
 - Would be useful to broadly categorise results by technique used
- Question whether approach is really representative of industry reality
 - For waste, measurements and assumptions should not be based on a best estimate approach.
 - Whilst statistical uncertainty can be assessed. Assumptions about modelling parameters would need to be conservative.
 - If measurements were being performed in a method similar to those actually being used, most reported measurements should have had a positive deviation.

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