NPL
National Physical Laboratory

NPL REPORT IR 35

NPL Nuclear Industry Proficiency Test Exercise 2014

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National
Measurement
System

NPL Nuclear Industry Proficiency Test Exercise 2014

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ABSTRACT

A fifth Nuclear Industry Proficiency Test Exercise has been run by NPL. One sample was prepared, consisting of a nominal 200 L drum loaded with bottles containing ion exchange resin. The bottles were spiked with known weights of a standard solution of ^{241}Am, ^{60}Co and ^{137}Cs. The activity concentrations (averaged across the drum contents) were approximately 3.4 Bq g^{-1}, 1.2 Bq g^{-1} and 1.2 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, an activity concentration range, details of the empty drum (e.g. its mass and dimensions) and the percentage of each material type present. The drum was measured by 8 UK and 5 overseas laboratories. Approximately 42 % of the results were statistically 'in agreement' with the NPL value. A high bias was observed for the ^{241}Am data.

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Approved on behalf of the NPLML by Dr Steven Judge, Radioactivity Group Leader, Acoustics and Ionising Radiation Division

CONTENTS

1 INTRODUCTION

The accurate measurement of radioactivity in potentially active materials produced in nuclear site decommissioning is essential for correct waste categorisation. This is important for public safety, to reduce the industry's costs and to minimise LLW volumes. The National Physical Laboratory (NPL) runs Nuclear Industry Proficiency Test Exercises ('drum comparisons') [Dean, 2007, 2010, 2012, 2013] to enable laboratories involved in the clearance and sentencing of bulk γ-emitting waste to test their measurement procedures. The exercises provide a check on the modelling techniques used for calculating detection efficiencies and enable participants to demonstrate measurement capability to third parties.

This report describes the fifth PTE in this series, covering:

- the preparation of the 'standard drum';
- the circulation of the drum and reporting of data;
- the reported results and data analysis.

2 PREPARATION OF STANDARD DRUM

The following materials were procured / prepared:

- Standardised radionuclide solutions of ^{241}Am, ^{60}Co and ^{137}Cs;
- 100 kg of ion-exchange resin;
- 240 x 500 ml HDPE bottles;
- 1 x 200 L steel drum.

The activity concentrations of the individual radionuclide solutions had been previously determined by assay in a re-entrant secondary standard ionisation chamber. Each ampoule had also been measured using a γ-spectrometer to check for the presence of any γ-emitting impurities; none were detected. The solutions were combined and the concentrations in the mixed solution were determined gravimetrically and confirmed by γ-spectrometry.

Ion-exchange resin (400 g nominal) was dispensed to each empty 500 ml bottle. The mixed radionuclide solution was dispensed to the bottles from a plastic pycnometer; the pycnometer was weighed throughout the dispensing sequence in such a way that the total mass of solution dispensed to each subset of 48 bottles was known.

The empty drum was loaded with the bottles in 5 layers of 48 bottles. Thus, the total activity in each layer was known. The drum was certificated for the activity concentrations of each radionuclide present (see Table 1) and was designated as source X140079.

3 CIRCULATION OF DRUM AND REPORTING OF DATA

An initial mailshot was prepared (see Appendix A) and circulated to potential participants.

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

- The activity concentration range (1 - 9 Bq/g);
- Details of the empty drum from the manufacturer;
- The mass of the drum empty (18.0 kg nominal) and full (132.3 kg nominal);
- The percentage by mass of the drum's contents:
    - HDPE (14 % nominal)
    - Ion-exchange resin (86 % nominal).

A list of participants is given in Appendix C.

4 RESULTS AND DATA ANALYSIS

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

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

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

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

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 plotted in Figures 1 – 3. The error bars in the graphs represent the standard uncertainty (k=1) of the deviation:

$$u_{\mathrm{D}} = 100\frac{L}{N}\sqrt{\left(\frac{u_{\mathrm{L}}}{L}\right)^2+\left(\frac{u_{\mathrm{N}}}{N}\right)^2} \qquad [2]$$

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:

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

$$R_{\mathrm{L}} = \frac{u_{\mathrm{L}}}{L} \quad [4]$$

$$z = \frac{L - N}{\sigma_p} = \frac{L - N}{0.05823\,N} \quad [5]$$

where:

ζ = zeta score
R_L = relative uncertainty of the participant's value
Z = z-score
σ_p = standard deviation for proficiency assessment

The standard deviation for proficiency assessment represents the level of performance that NPL would wish laboratories to be able to achieve. It corresponds to a deviation 'D' of 15% at the 99% confidence level.

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 results are plotted in Figures 4 - 6. The zeta scores, z-scores and outlier results are also given in Tables 2 - 4.

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.

5 DISCUSSION

All the participants used hyperpure Ge γ-spectrometers of various types. Participants 2, 7 and 9 used detectors with 3 Ge crystals and participants 3 and 5A used Segmented Gamma Scanners. A range of methods were used to determine detection efficiencies including ISOCS, ISOTOPIC, SNAP and Winner Track software. Some participants used in-house calibration sources.

Overall, 42% of the results submitted were in agreement with the NPL Assigned Values. The percentages in agreement by nuclide were ^{241}Am (5%), ^{60}Co (58%) and ^{137}Cs (63%). Apart from two results, all the results for ^{241}Am were higher than the NPL value, with positive deviations typically between 10 % and 50 % but approaching an order of magnitude higher in two cases. It may be significant that the activity in the drum was in effect distributed as 5 layers of diffuse point sources, although a

similar drum had been prepared for the first PTE in this series (Dean, 2007) and much better results had been obtained for ^{241}Am in that exercise.

In comment with earlier exercises in this series, there was some evidence that Segmented Gamma Scanners were producing values lower than the true value.

6 ACKNOWLEDGEMENTS

The author wishes to thank the participating organisations for the time and effort they have put into analysing the drum, and for the information provided. Also, thanks go to Daniel Ainsworth for organising the transport of the drum and Peter Ivanov for reviewing this report. Finally, the author gratefully acknowledges the financial support of the National Measurement System.

7 REFERENCES

Dean, J.C.J. A Comparison of Procedures Used at UK Nuclear Sites for Gamma Assays of Potentially Contaminated or Activated Materials. NPL Report IR2, 2007.

Dean, J.C.J. A Second Comparison of Procedures for the Assay of Low Levels of Gamma-emitters in Nuclear Site Waste. NPL Report IR19, 2010.

Dean, J.C.J. NPL Nuclear Industry Proficiency Test Exercise 2011. NPL Report IR25, 2012.

Dean, J.C.J. NPL Nuclear Industry Proficiency Test Exercise 2012. NPL Report IR30, 2013.

Harms, A.V. A New Approach for Proficiency Test Exercise Data Evaluation. Accreditation and Quality Assurance, 14, 253-261, 2009.

8 TABLES

Table 1 - Principal radionuclides in X140079

Radionuclide	Decay mode	Activity concentration (averaged across contents of drum) @ 2014-05-01 1200 UTC• (Bq g^{-1})	Standard uncertainty (k=1) (Bq g^{-1})	Standard uncertainty (k=1) (%)
^{241}Am	α/γ	3.346	0.013	0.39
^{60}Co	β^-/γ	1.2303	0.0050	0.41
^{137}Cs	β^-, β^-/γ	1.232	0.009	0.73

• Universal Time, Co-ordinated. This replaced Greenwich Mean Time in 1972. Format of date isYYYY-MM-DD.

Table 2 - Reported results for ^{241}Am

NPL activity concentration = (3.346 ± 0.013) Bq g^{-1} (k=1) Reference time: 2014-05-01 1200 UTC

Participant code	Reported activity concentration (Bq/g)	Reported uncertainty (%)	Zeta score	Z score	D (%)	Outlier test result	Classification
1A	4.91	21.7	1.47	8.03	46.74	Not outlier	Questionable
1B	3.72	283	0.04	1.92	11.18	Outlier	Questionable
2	3.91	29.64	0.49	2.89	16.86	Not outlier	Questionable
3	2.98	14.9	-0.82	-1.88	-10.94	Not outlier	In agreement
4A	4.24	20.23	1.04	4.59	26.72	Not outlier	Questionable
4B	3.92	20.37	0.72	2.95	17.15	Not outlier	Questionable
4C	4.28	20.36	1.07	4.79	27.91	Not outlier	Questionable
4D	4.13	20.34	0.93	4.02	23.43	Not outlier	Questionable
5A	3.85	30.5	0.43	2.59	15.06	Not outlier	Questionable
5B	4.46	24	1.04	5.72	33.29	Not outlier	Questionable
6A	4.28	40.0	0.55	4.79	27.91	Not outlier	Questionable
6B	4.27	39.8	0.54	4.74	27.62	Not outlier	Questionable
6C	4.19	39.4	0.51	4.33	25.22	Not outlier	Questionable
7	28.027	10.322	8.53	127	737.6	Not outlier	Discrepant
8	4.13	20.34	6.73	136	794.5	Not outlier	Discrepant
9	2.22	50	-1.01	-5.78	-33.65	Not outlier	Questionable
10	8.55	5	12.17	26.7	155.5	Not outlier	Discrepant
11	4.36	26	0.89	5.20	30.30	Not outlier	Questionable
12	5.44	59	0.65	10.8	62.58	Not outlier	Questionable

Table 3 - Reported results for ^{60}Co

NPL activity concentration = (1.2303 ± 0.0050) Bq g^{-1} (k=1) Reference time: 2014-05-01 1200 UTC

Participant code	Reported activity concentration (Bq/g)	Reported uncertainty (%)	Zeta score	Z score	D (%)	Outlier test result	Classification
1A	1.55	11.3	1.83	4.47	26.0	Not outlier	Questionable
1B	1.22	42.5	-0.02	-0.14	-0.81	Not outlier	In agreement
2	1.21	27.22	-0.06	-0.28	-1.63	Not outlier	In agreement
3	1.18	1.8	-2.29	-0.70	-4.07	Not outlier	In agreement
4A	1.19	14.40	-0.23	-0.56	-3.25	Not outlier	In agreement
4B	1.14	14.47	-0.55	-1.26	-7.32	Not outlier	In agreement
4C	1.13	14.45	-0.61	-1.40	-8.13	Not outlier	In agreement
4D	1.10	14.46	-0.82	-1.82	-10.6	Not outlier	In agreement
5A	1.04	10.0	-1.82	-2.65	-15.4	Not outlier	Questionable
5B	1.05	10.4	-1.65	-2.51	-14.6	Not outlier	In agreement
6A	1.42	16.9	0.79	2.65	15.5	Not outlier	Questionable
6B	1.43	16.9	0.83	2.79	16.3	Not outlier	Questionable
6C	1.40	16.8	0.72	2.37	13.8	Not outlier	In agreement
7	2.114	0.254	120.48	12.34	71.9	Not outlier	Discrepant
8	1.40	1.5	7.88	2.37	13.8	Not outlier	Questionable
9	1.07	7	-2.13	-2.23	-13.0	Not outlier	In agreement
10	1.62	4.06	5.91	5.45	31.7	Not outlier	Discrepant
11	1.38	12	0.91	2.09	12.2	Not outlier	In agreement
12	1.44	16	0.91	2.93	17.1	Not outlier	Questionable

Table 4 - Reported results for ^{137}Cs

NPL activity concentration = (1.232 ± 0.009) Bq g^{-1} (k=1) Reference time: 2014-05-01 1200 UTC

Participant code	Reported activity concentration (Bq/g)	Reported uncertainty (%)	Zeta score	Z score	D (%)	Outlier test result	Classification
1A	1.47	11.7	1.38	3.32	19.3	Not outlier	Questionable
1B	1.23	66.3	0.00	-0.03	-0.162	Not outlier	In agreement
2	1.25	27.24	0.05	0.25	1.46	Not outlier	In agreement
3	1.07	3.5	-4.21	-2.26	-13.1	Not outlier	Questionable
4A	1.18	20.34	-0.22	-0.72	-4.2	Not outlier	In agreement
4B	1.16	20.36	-0.30	-1.00	-5.84	Not outlier	In agreement
4C	1.19	20.34	-0.17	-0.59	-3.41	Not outlier	In agreement
4D	1.15	20.34	-0.35	-1.14	-6.66	Not outlier	In agreement
5A	1.04	20.1	-0.92	-2.68	-15.6	Not outlier	Questionable
5B	1.09	16.2	-0.80	-1.98	-11.5	Not outlier	In agreement
6A	1.38	12.5	0.86	2.06	12.0	Not outlier	In agreement
6B	1.33	12.2	0.60	1.37	7.95	Not outlier	In agreement
6C	1.34	11.9	0.68	1.51	8.77	Not outlier	In agreement
7	2.713	0.559	83.98	20.64	120	Not outlier	Discrepant
8	1.42	2.3	5.55	2.62	15.3	Not outlier	Discrepant
9	1.09	7	-1.85	-1.98	-11.5	Not outlier	In agreement
10	1.80	4.4	7.13	7.92	46.1	Not outlier	Discrepant
11	1.36	18	0.52	1.78	10.4	Not outlier	In agreement
12	1.42	21	0.63	2.62	15.3	Not outlier	Questionable

9 FIGURES

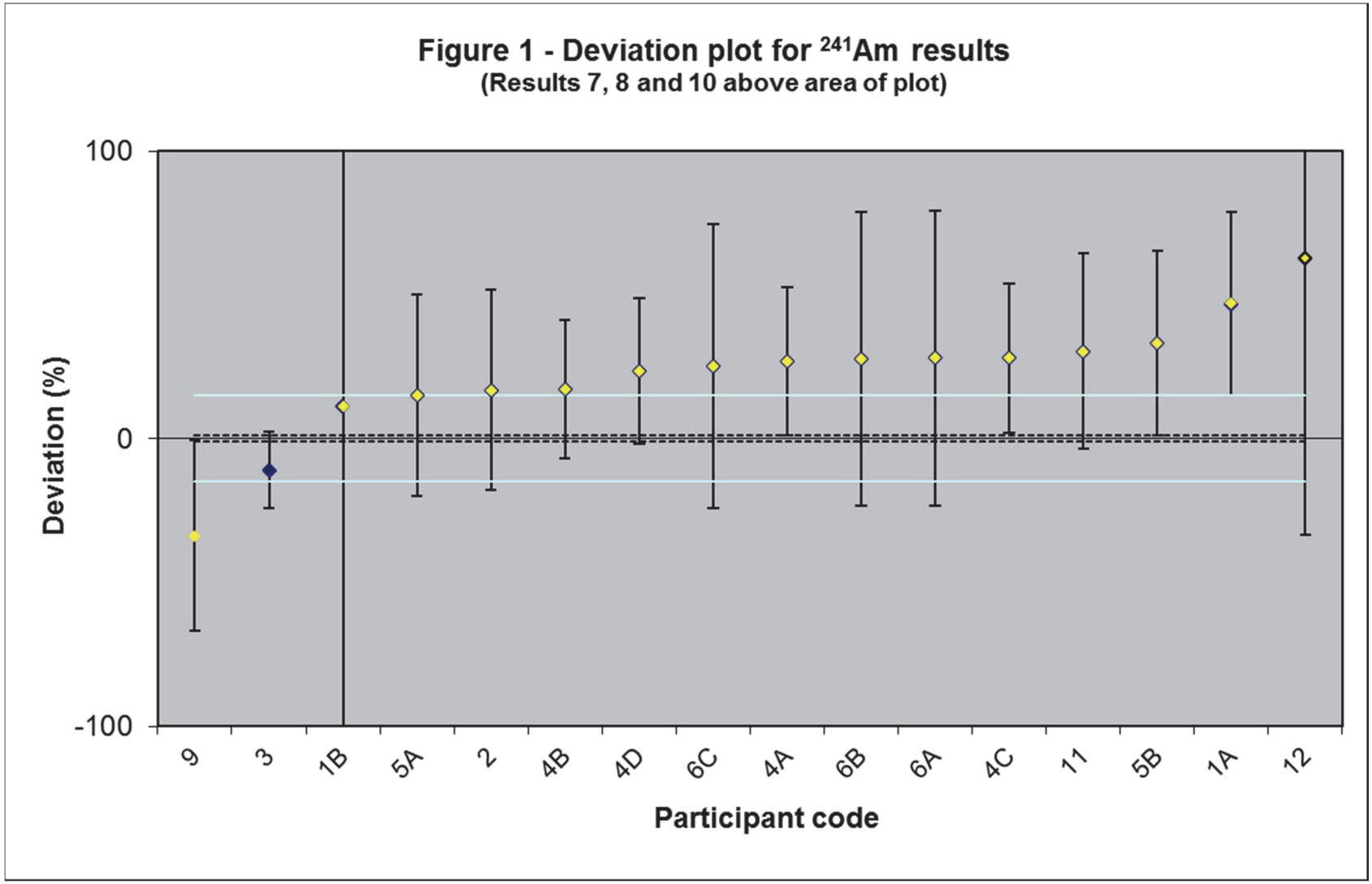
Figure 1 - Deviation plot for 241Am results
(Results 7, 8 and 10 above area of plot)
100
0
-100
Deviation (%)
9
3
1B
5A
2
4B
4D
6C
4A
6B
6A
4C
11
5B
1A
12
Participant code

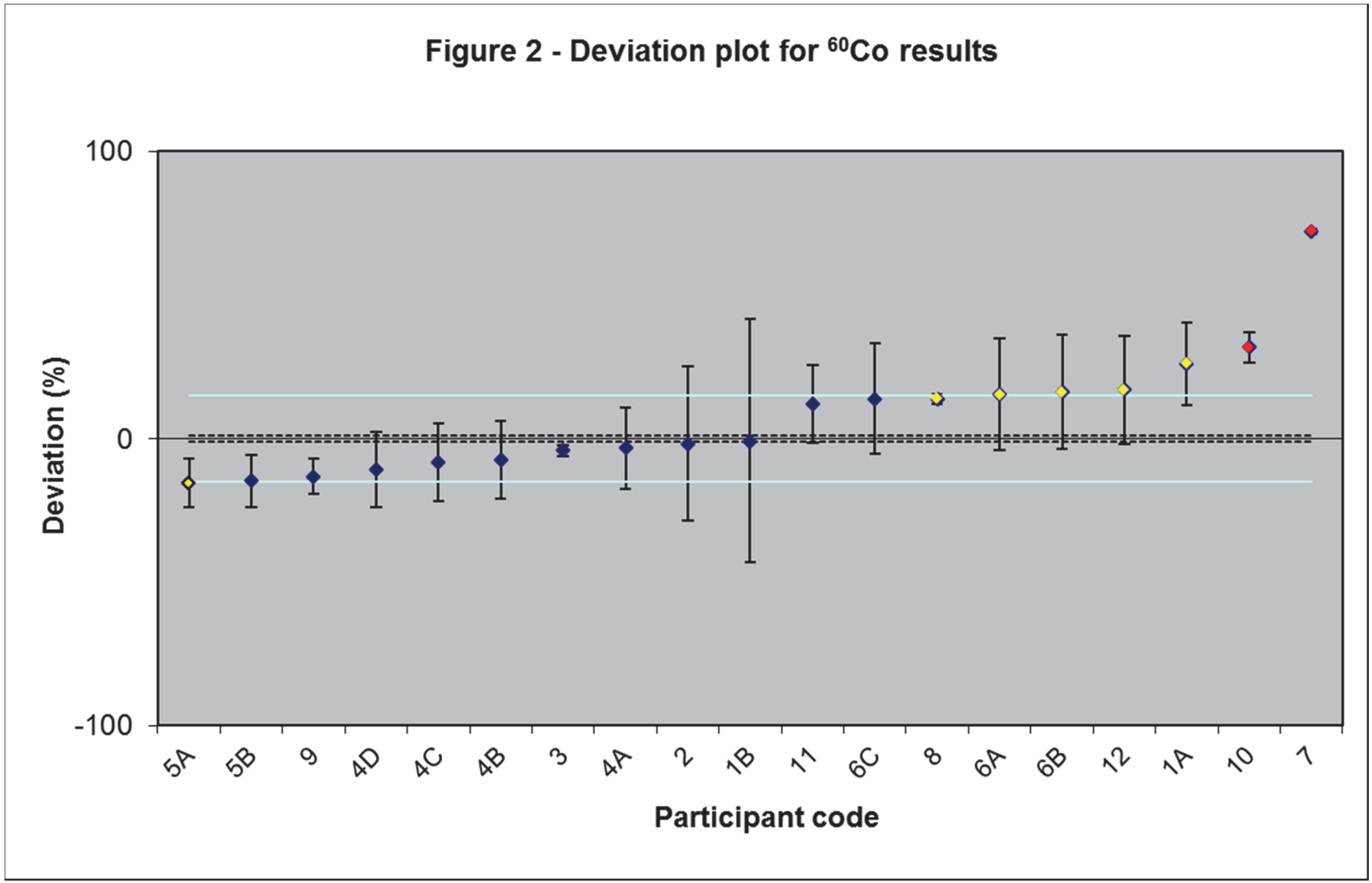
Figure 2 - Deviation plot for 60Co results
100
0
-100
Deviation (%)
5A
5B
9
4D
4C
4B
3
4A
2
1B
11
6C
8
6A
6B
12
1A
10
7
Participant code

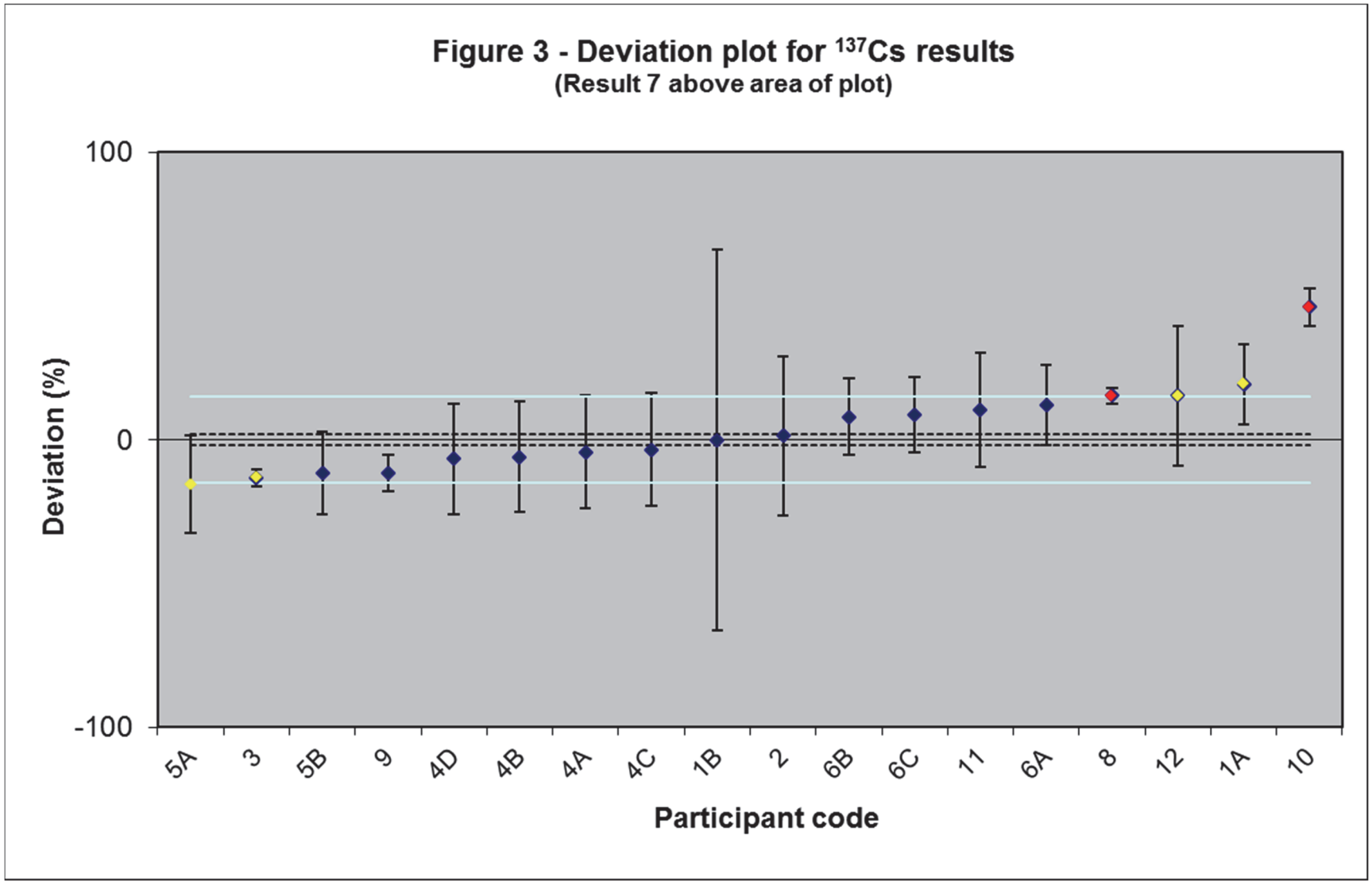
Figure 3 - Deviation plot for 137Cs results
(Result 7 above area of plot)
100
0
-100
Deviation (%)
5A
3
5B
9
4D
4B
4A
4C
1B
2
6B
6C
11
6A
8
12
1A
10
Participant code

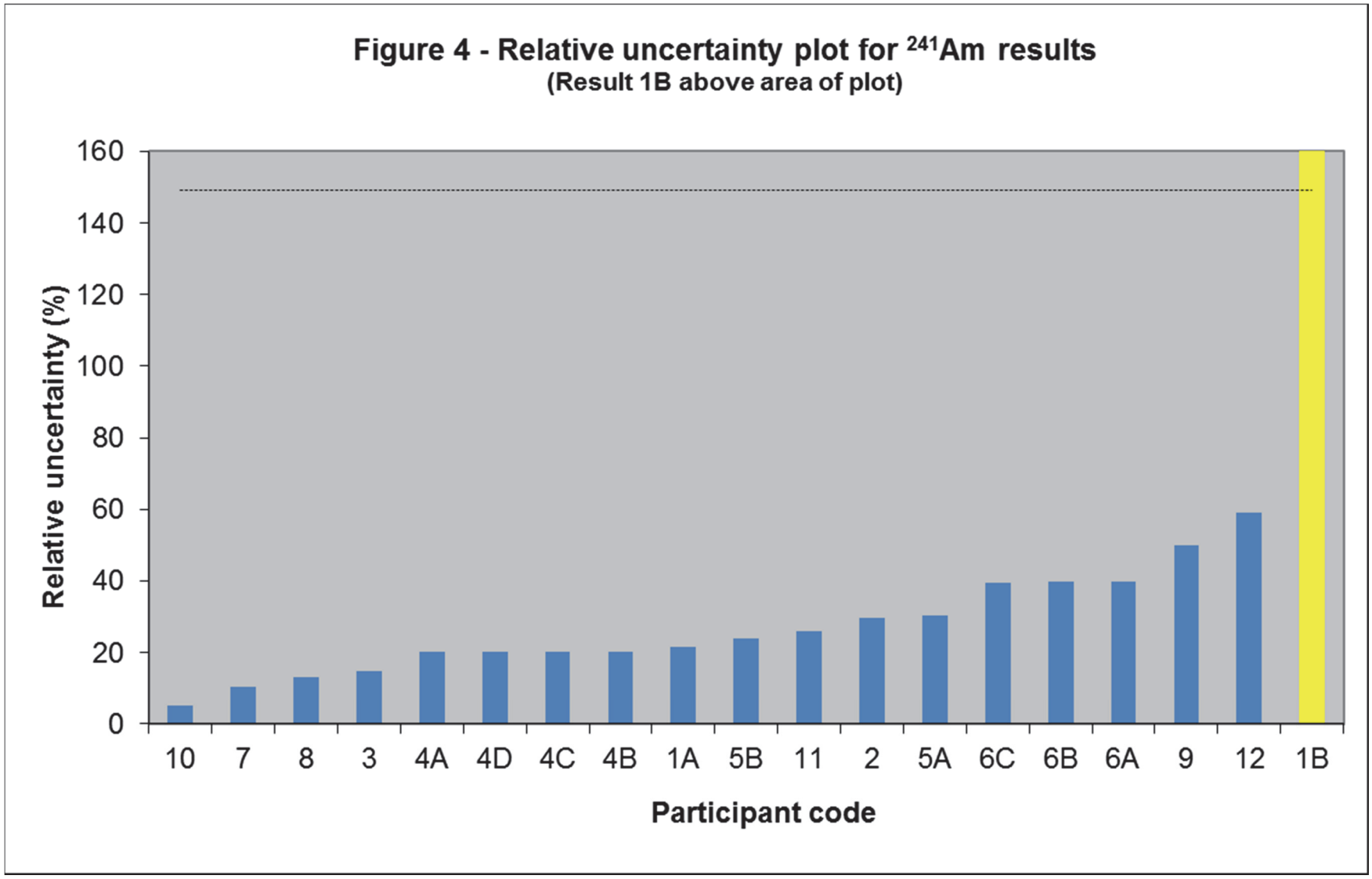
Figure 4 - Relative uncertainty plot for 241Am results
(Result 1B above area of plot)
Relative uncertainty (%)
160
140
120
100
80
60
40
20
0
10
7
8
3
4A
4D
4C
4B
1A
5B
11
2
5A
6C
6B
6A
9
12
1B
Participant code

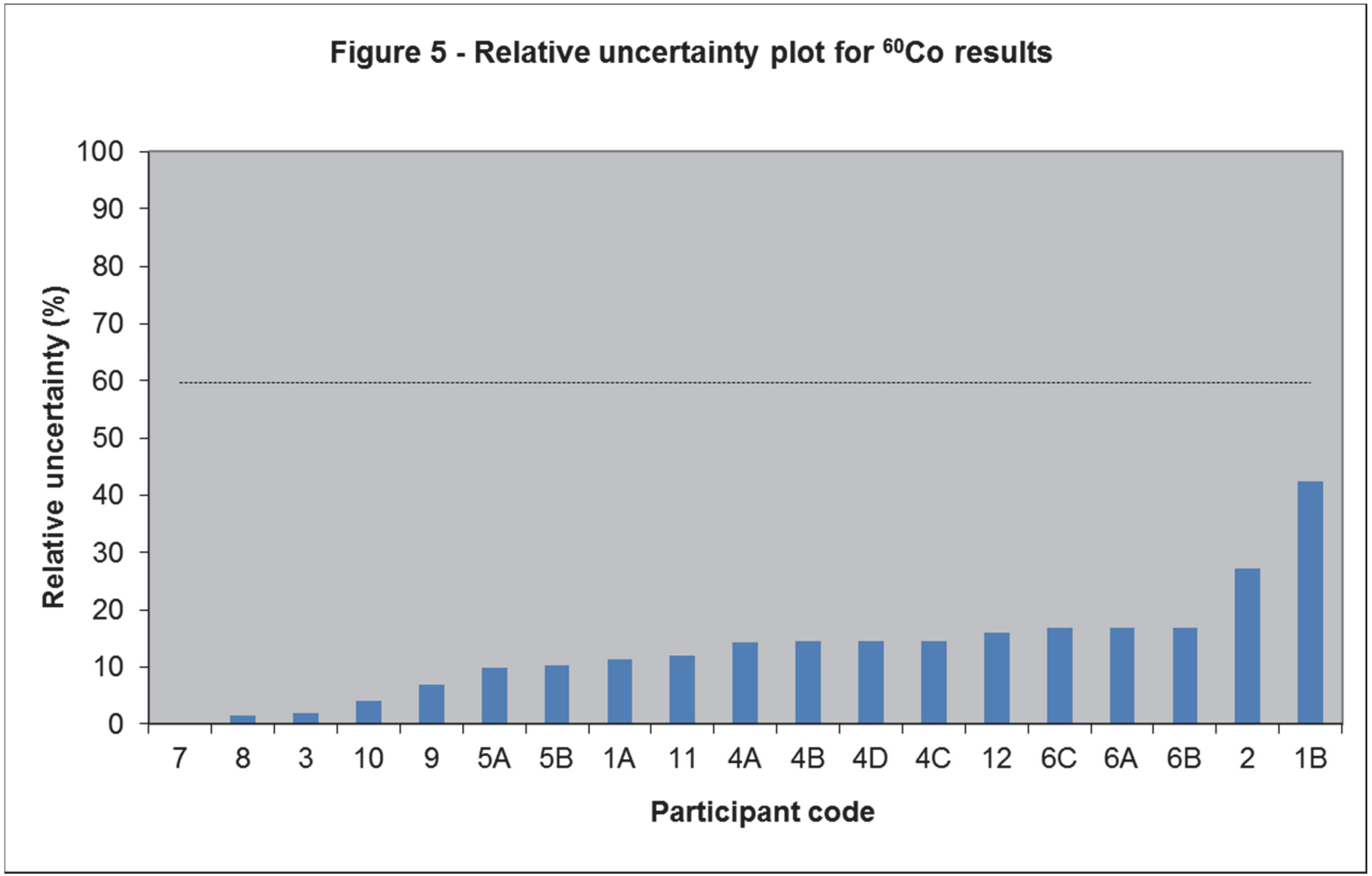
Figure 5 - Relative uncertainty plot for ^{60}Co results
Relative uncertainty (%)
100
90
80
70
60
50
40
30
20
10
0
7
8
3
10
9
5A
5B
1A
11
4A
4B
4D
4C
12
6C
6A
6B
2
1B
Participant code

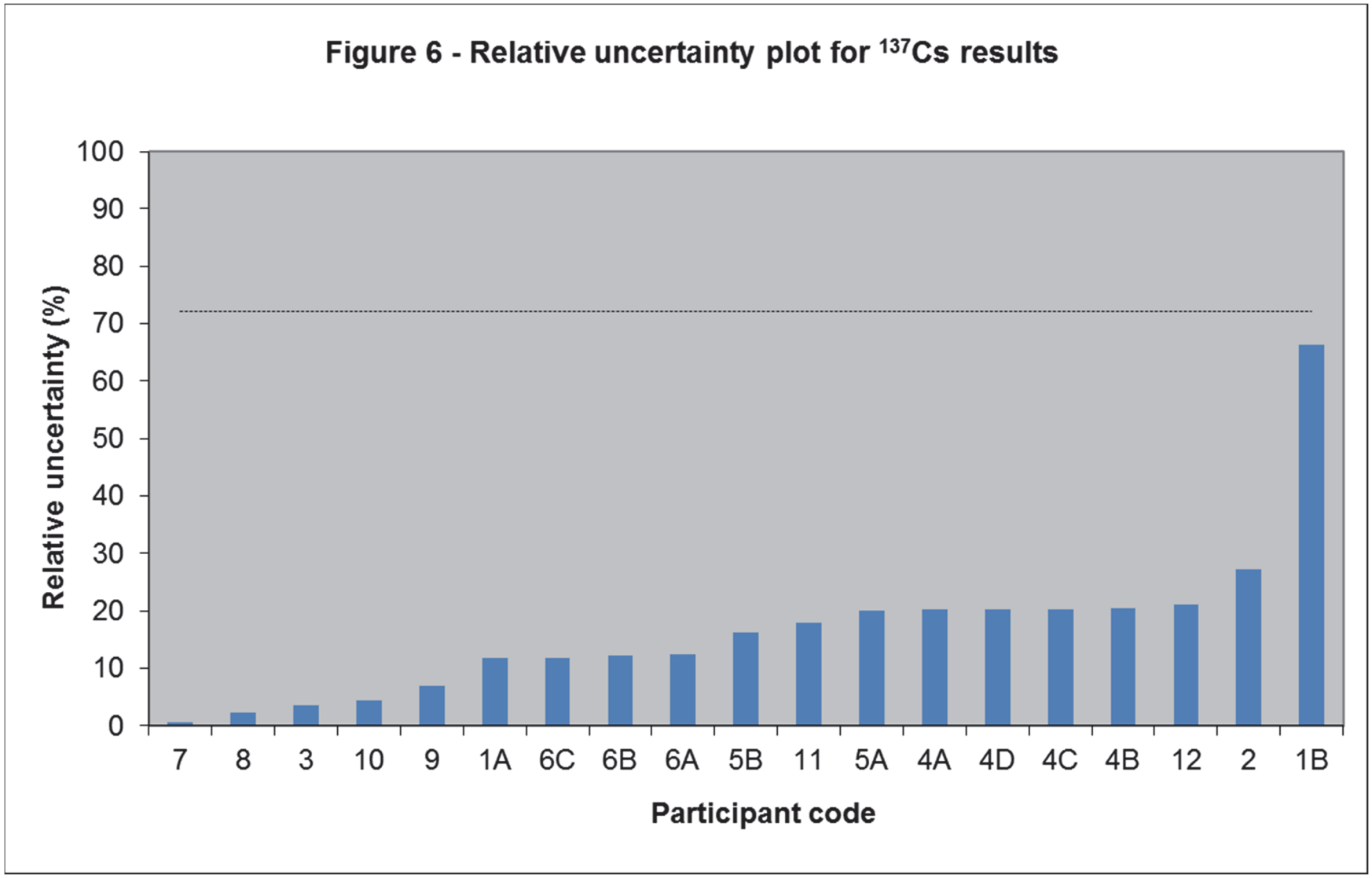
Figure 6 - Relative uncertainty plot for 137Cs results
Relative uncertainty (%)
100
90
80
70
60
50
40
30
20
10
0
7
8
3
10
9
1A
6C
6B
6A
5B
11
5A
4A
4D
4C
4B
12
2
1B
Participant code

10 APPENDIX A – Initial Mailshot

Dear Colleague,

NPL NUCLEAR INDUSTRY PROFICIENCY TEST EXERCISE 2014

NPL is about to run its fifth bulk-waste proficiency test exercise ('drum comparison') to enable laboratories worldwide involved in decommissioning and site clearance to test their bulk-waste gamma measurement procedures. As in previous exercises, the purpose 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 plastic bottles filled ion-exchange resin. The radionuclides ^{241}Am, ^{60}Co and ^{137}Cs will be present in each bottle and the overall activity concentration will be in the range 1 – 10 Bg g^{-1}.

The drum will be available for measurement between 7 July and 24 November 2014 and participants will be asked to report the activity concentrations of the individual radionuclides. A short exercise report will be published in February 2015. All results will be coded and treated as confidential.

n.b. Please note that, due to limited available funding, there will be no post-exercise meeting.

If you are interested in participating, please read the full details given on pages 2 to 3 below and then complete and return the PTE Enquiry Form. Please note that requests to participate must be received by close of business on 25 April 2014.

We hope you are interested and that you will be able to participate.

Yours faithfully

Julian Dean March 2014

A) Plan for NPL Nuclear Industry PTE 2014

NPL will prepare a single 'mock waste' sample in a 200-litre steel drum. The drum will contain 240 plastic bottles (in 5 'layers' of 48) filled with ion-exchange resin. The radionuclides ^{241}Am, ^{60}Co and ^{137}Cs will be present in each bottle. The activity will be added to each bottle as a few weighed drops of standardised solution, so the distribution of activity in a bottle will approximate to a point source. The overall activity concentration of the drum (i.e. activity / total mass of material in drum) will be in the range 1 – 10 Bg g^{-1}.

The drum will be delivered to (and collected from) the participants on agreed dates. Participants will be given 3 working days to measure the drum (excluding arrival and dispatch days). The drum will be an Excepted Package. NPL will disclose the percentage by mass of plastic and resin present, the dimensions of the drum and the mass of the drum (empty and full).

Note that the filled drum will have a mass of approximately 130 kg and will be loaded on a pallet. Participants must have arrangements in place for moving the drum from and to the courier's

van; they must also ensure that the drum is secured to the pallet and bears the address label provided by NPL before it is returned to the courier.

Participants will be asked to report their measurements of the individual radionuclide activity concentrations along with details of calibration procedures and instrumentation used. NPL will then prepare a short draft report (including all results, coded for anonymity) and issue to all participants.

B) Timetable

Action	Dates
Interested laboratories to reply to this mailshot	By 25 April 2014
NPL to contact each laboratory, advise if they can be accommodated, and, if so, agree receipt and dispatch dates	By 20 June 2014
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 7 July and 24 November 2014
Participants to submit results	By 8 December 2014
NPL to issue report	February 2015

C) How to participate

Please complete the enclosed Enquiry Form and return it by close of business on **25 April 2014** to radioactivity@npl.co.uk . Please indicate on the form which weeks in the period 7 July to 24 November 2014 you would prefer for delivery of the sample drum to your site. Note that you will be allocated three full working days 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 by 20 June 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: **£1075**
- Delivery: **To be advised**

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

11 APPENDIX B – Reporting form

NUCLEAR INDUSTRY PTE 2014 REPORTING FORM	
Participating laboratory	
Contact person	
Address	
Submission date	

Radionuclide	Activity concentration (Bq g^{-1}) RT 1200 UTC, 01/05/14	Type A uncertainty (%, k=1)	Type B uncertainty (%, k=1)	Combined standard uncertainty (%, k=1)
^{241}Am				
^{60}Co				
^{137}Cs				

Please provide brief details of the following:

Detector type	
Scanning method used (including, e.g., detector collimation, segmented scanning and rotation of drum)	
Matrix density correction applied	
Calibration standards used	
Efficiency modelling and software	
Other	

12 APPENDIX C – List of Participants

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Nucleco S.p.A.
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Rome
Italy

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