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Australasian Gamma-ray Spectrometry Capability Exercise – 2015

Stephen Long

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by

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The author wishes to thank all of the laboratories that participated in the exercise.

EXECUTIVE SUMMARY

International experience with significant radiological incidents has shown that extensive measurement of environmental samples is required (Japanese Ministry of Health, Labour and Welfare, 2015) (IAEA, 1988). Many of these samples would require analysis by gamma-spectrometry. ARPANSA's capacity to analyse samples could be overwhelmed by the response required by a significant radiological incident. Therefore, assistance from other laboratories might be required to respond to a significant radiological incident.

This exercise was conducted to ascertain the capability of Australasian laboratories with respect to the measurement of radionuclides in food by gamma-ray spectrometry. The purpose of the exercise was simply to determine whether the methods currently used by each participating laboratory produced acceptable results when applied to a sample containing radionuclides created in a nuclear reactor.

Nine of the ten Australasian radioanalytical laboratories identified as having gamma-spectrometry capabilities, participated in this exercise. The tenth laboratory had previously declined to participate because they specialise in analyses that do not include the radionuclides used in the exercise.

Participants were asked to analyse 500 ml of dried spinach, containing known amounts of certain radionuclides, obtained from an accredited supplier of Certified Reference Materials and Proficiency Testing products. The sample contained Am-241, Cs-134, Cs-137, Co-60, K-40 and Zn-65.

Many of the participants had difficulty with the analysis of zinc-65 due to the low activity of this radionuclide remaining in the sample. However, the poor results for this radionuclide highlighted the fact that five of the nine participants chose to measure volumes that maximised both measurement uncertainties (or counting times) and the effects of True Coincidence Summing. Nonetheless, this radionuclide was discarded from further consideration.

Forty two of the forty five reported results were within the Performance Acceptance Limits defined by the supplier. However, only five of nine participants were able to provide results which were consistent with the reference value, for all radionuclides.

Cs-137 is a key indicator of radioactive contamination. It is, therefore, concerning that three of the participants reported discrepant results for this radionuclide. These laboratories may require assistance before they could be relied upon to provide assistance for some radiological incidents, particularly those involving the dispersion of radioactive material created in a nuclear reactor.

1. INTRODUCTION

ARPANSA maintains a radioanalytical laboratory, in part, to respond to radiological incidents. Theoretically, this laboratory has the capacity to measure about 100 samples per day by gamma-spectrometry. However, even this capacity could be overwhelmed by the response required by a significant radiological incident. Therefore, assistance from other laboratories might be required to respond to such an incident.

ARPANSA has identified ten radioanalytical laboratories in Australia and New Zealand. This list includes 3 laboratories run by the Australian Nuclear Science and Technology Organisation (ANSTO), ARPANSA's own laboratory, 2 commercial laboratories as well as four other state or national laboratories. However, there is considerable specialisation in the radionuclides analysed by these laboratories. This is because many of these laboratories exist to fulfil specific needs and are not funded or specifically tasked with providing analyses in the event of general radiological incident. Therefore, some laboratories may not be able to provide assistance for some radiological incidents, particularly those involving the dispersion of radioactive material created in a nuclear reactor.

This exercise was conducted to ascertain the capability of participating laboratories with respect to the measurement of radionuclides in food by gamma-ray spectrometry and was a follow-on from a similar exercise conducted in 2013 (Long, 2014). The earlier exercise assessed general capability with gamma-ray spectrometry and six of eight identified laboratories participated.

It should be noted that this was not a Proficiency Test Exercise because participants were not judged as to the acceptability of their procedures. The procedures used by each laboratory are appropriate to their particular objectives. Rather, the purpose of the exercise was simply to determine whether the methods currently used by each participating laboratory produced acceptable results when used for another purpose.

2. METHODOLOGY

Nine of the ten known Australasian radioanalytical laboratories (including ARPANSA) agreed to participate in the exercise. The tenth laboratory had previously declined to participate because they specialise in analyses that do not include the radionuclides used in the exercise.

ARPANSA purchased nine containers of a certified reference material from an accredited supplier of such products. The material was prepared, by the supplier, by adding known amounts of Am-241, Cs-134, Cs-137, Co-60, Mn-54 and Zn-65 to dried, powdered spinach and then thoroughly mixing to ensure homogeneity. As spinach naturally contains K-40, this radionuclide was also included in the list of radionuclides.

The supplier also provided a certificate of analysis stating the certified values of the activity concentration of each radionuclide in the dried material. The certificate of analysis also included Performance Acceptance Limits (PALs) approximating 95% confidence intervals of the performance that an experienced laboratory should achieve.

A container holding 500 ml of the vegetation was sent to each laboratory along with a form for reporting their results. The participants were asked to measure the activity concentration of the radionuclides in the dried material by gamma-spectrometry and to report their results within eight weeks of receiving the sample.

2.1 Analysis of Results

For each radionuclide, ARPANSA calculated the Relative Bias (*R*) of the measured activity concentration (*M*), relative to the certified activity concentration supplied by the manufacturer (*C*):

Equation 1:

$$\mathbf{R = \frac{M-C}{C} \times 100}$$

Due to the uncertainties in both the certified value (u_c) and the measured value (u_m), the Relative Bias for an individual radionuclide may not be statistically significant. Therefore, ARPANSA also calculated the U-test value (*U*):

Equation 2:

$$\mathbf{U = \frac{\sqrt{(M-C)^2}}{\sqrt{u_m^2 + u_c^2}}}$$

The Relative Bias for an individual radionuclide is statistically significant if U > 2.58 and may be statistically significant if U > 1.64.

If a participant under- or over-estimated the uncertainty associated with a measurement, the U-test value will over- or underestimate the statistical significance of the Relative Bias. Therefore, ARPANSA also compared the measured value to the PALs provided by the supplier of the sample.

Each participant was provided with an individual report detailing the Relative Bias, U-test value and comparison with the PALs for each radionuclide. This report also indicated if there appeared to be systematic errors in the laboratory's measured values and indicated the potential sources of these errors, based on the results and responses from the report form.

The laboratories were given a designation based on the date they reported their results: laboratory 1 was the first to report; laboratory 2 was the second to report; and so on.

3. RESULTS AND DISCUSSION

In interpreting the results of the survey, the information contained in Annex A of the report of the previous exercise (Long, 2014) may prove useful.

3.1 Zinc-65

The measurement of Zn-65 in the sample proved challenging to most of the participants. Indeed, only three of the nine participants were able to provide a result that was within the PALs for this radionuclide. The results reported by each participant are shown in Figure 1.

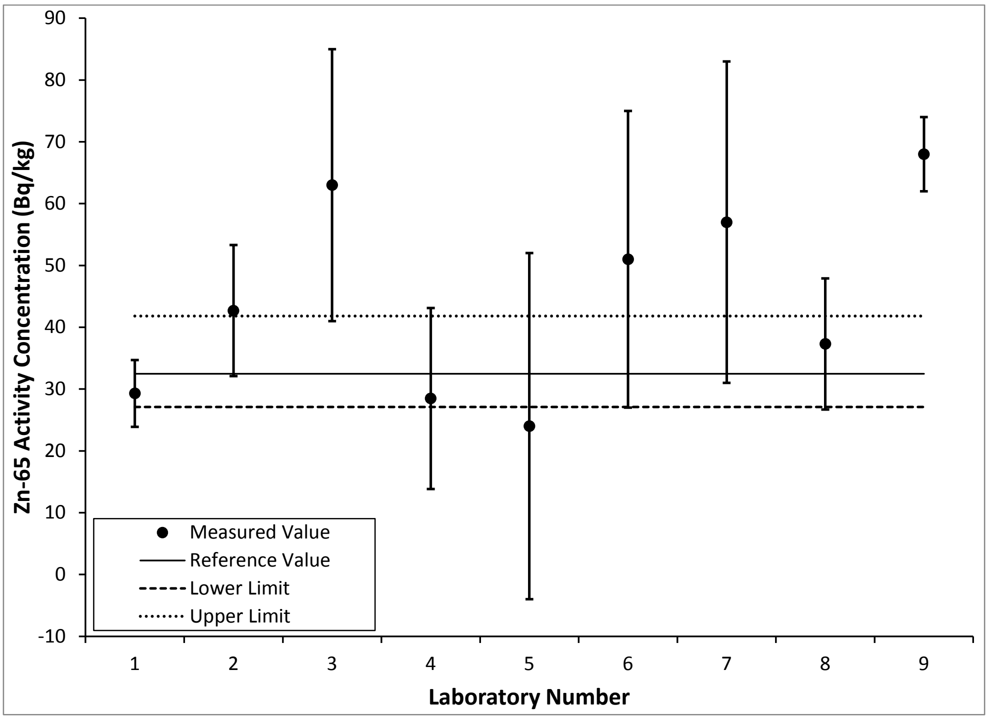

Figure 1: Reported results for zinc-65.
The error bars indicate the 95% confidence interval of the measurement.

The most striking feature of Figure 1 is that the error bars are so large. This is because the activity of Zn-65 in the sample, at the time of measurement (May 2015), was very low. However, the participants were required to report the activity at the reference date of the sample (March 2013). Unlike the other radionuclides in the sample, which have half-lives of at least a few years, Zn-65 has a half-life of only 244 days. Therefore, while the sample had approximately 30 Bq/kg Zn-65 activity concentration in March 2013, only a tenth of this activity remained by May 2015.

The difficulty in measuring such a low activity concentration was further exacerbated by the sample volume chosen for measurement by many of the participants. Table 1 lists the pertinent details of the Zn-65 measurement for each laboratory. While each laboratory was supplied with 500 ml of the sample, six laboratories used less than 50% of the sample for their measurement. This makes a considerable difference to ease of measurement, for example: laboratory 4 was attempting to measure 1.7 Bq of Zn-65, while laboratory 6 was attempting to measure only 0.1 Bq.

Table 1: Details of Zn-65 Measurements

The shaded rows indicate results affected by True Coincidence Summing				
Lab. Number	**Measured Volume (ml)**	**Fraction of Sample**	**Relative Uncertainty**	**Relative Bias in Zn-65 Result**
6	35	7%	9%	57%
7	45	9%	12%	75%
9	45	9%	17%	109%
3	55	11%	26%	94%
5	85	17%	58%	26%
8	200	40%	24%	15%
1	450	90%	23%	10%
2	500	100%	14%	31%
4	500	100%	4%	12%

Using smaller volumes of sample does mean that the counting efficiency is increased. However, this increase is only about a factor of two between the largest and smallest volumes used. Therefore, if laboratory 4 observed 100 counts in the Zn-65 peak for a given counting period, laboratory 6 would only observe about 15 counts in the same period. So, if both laboratories measure the sample over the same period, laboratory 6 would have an uncertainty at least 2.5 times larger than laboratory 4. Alternatively, if laboratory 4 observed 100 counts in the Zn-65 peak over a counting period of 4 hours, laboratory 6 would have to count for approximately 28 hours to observe the same number of counts and have the same counting uncertainty. Table 1 shows the large variation in relative uncertainty due to variations in sample volume and measurement time.

True Coincidence Summing (TCS) effects increase as sample volume decreases due to the increase in detection efficiency. The majority of laboratories that used small sample volumes were, unfortunately, also those that do not correct for TCS effects.

3.2 Performance Relative to Acceptance Limits

As noted in section 2, the certificate of analysis for each sample also included PALs approximating 95% confidence intervals of the performance that an experienced laboratory should achieve. The reported activity concentration for each radionuclide, for each laboratory, was compared to these acceptance limits. Table 2 details the number of laboratories reporting results within these limits. This table also details the fraction of laboratories reporting acceptable results in the 2013 exercise.

Table 2: Laboratories reporting acceptable results

Note that the results for zinc-65 have been discarded from further analysis				
	2015 Exercise		2013 Exercise	
Radionuclide	Number	Fraction	Number	Fraction
Am-241	8	89%	5	83%
Co-60	9	100%	6	100%
Cs-134	8	89%	6	100%
Cs-137	8	89%	6	100%
K-40	9	100%		
Zn-65	3	33%	5	83%

As was the case in the 2013 exercise, most laboratories reported acceptable results for most radionuclides. This indicates that the methods currently used by each participating laboratory produced acceptable results when used for another purpose.

3.3 Performance relative to the certified activity

The comparison with manufacturer-defined acceptance limits indicates that the procedures used by each laboratory are equivalent to those used by their peers world-wide. However, these acceptance limits accommodate common analysis errors, such as the failure to correct for TCS.

Highly capable laboratories should produce a result that is commensurate with the certified value. The usual method to test whether a reported result is not significantly different to a certified value is the U-test (see equation 2). This test compares the measured result with the certified result in a way that accounts for the uncertainty in both values.

The U-test values for each radionuclide and each laboratory are shown in Figure 2. In the figure, the dotted line indicates a U-test value of 1.64. Results with a U-test value below 1.64 are not statistically different from the certified value and are, therefore, consistent. The dashed line in the figure indicates a U-test value of 2.58. Results with a U-test value greater than 2.58 are statistically different from the certified value. That is, those results with a U-test value greater than 2.58 are discrepant and indicate a significant error in the analysis.

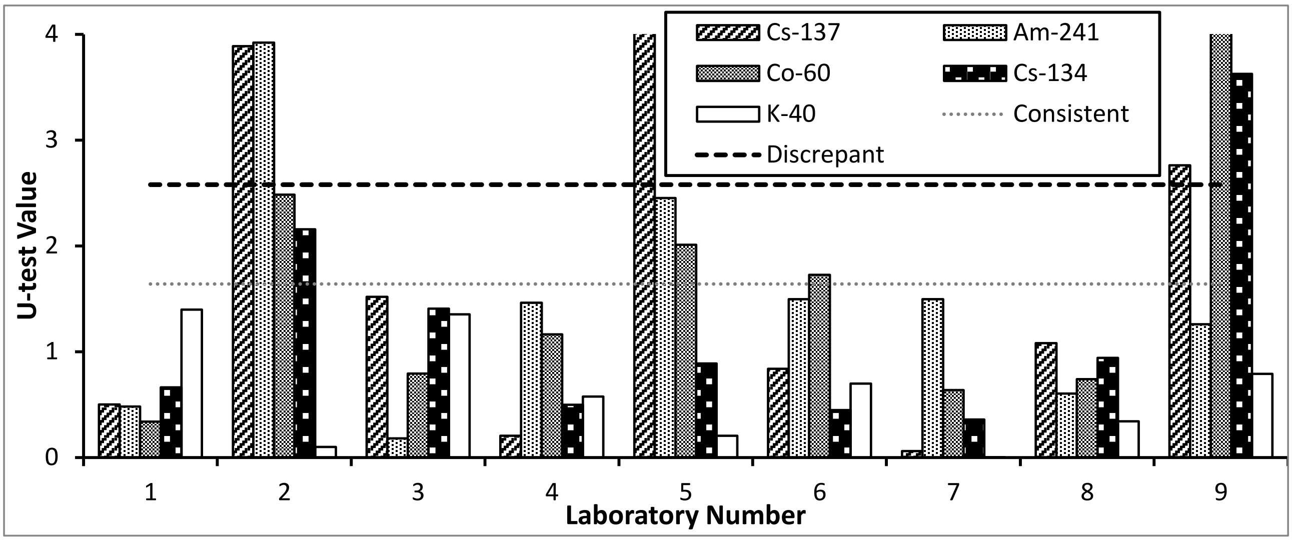

Figure 2: U-test values for each radionuclide and each laboratory.

Figure 2 indicates that laboratories 1, 3, 4, 7 and 8 provided excellent results because all of their reported values were consistent with the reference value.

Figure 2 also shows that laboratories 2, 5 and 9 each produced results with U-test values greater than 2.58. This indicates that significant errors were made by each of these laboratories when analysing the sample. Of particular concern is that all three of these laboratories were not able to provide a result for Cs-137 that was commensurate with the certified value.

Laboratory 6 provided only one result with a U-test value greater than 1.64: that for Co-60 with a U-test value of 1.72. While this indicates that an error in the analysis might have been made, this value could also be due to simple statistical variation.

4. CONCLUSION

This exercise was conducted to ascertain the capability of participating laboratories with respect to the measurement of radionuclides in food by gamma-ray spectrometry. It should be noted the purpose of the exercise was simply to determine whether the methods currently used by each participating laboratory produced acceptable results when applied to a sample containing radionuclides created in a nuclear reactor.

Nine of the ten Australasian radioanalytical laboratories identified as having gamma-spectrometry capabilities participated in this exercise. A container of a Certified Reference Material, purchased from an accredited supplier of Proficiency Testing products, was sent to each participant. The material comprised dried, powdered spinach to which had been added known amounts of Am-241, Cs-134, Cs-137, Co-60, Mn-54 and Zn-65. The laboratories were given a designation based on the date they reported their results: laboratory 1 was the first to report, laboratory 2 was the second to report, and so on.

Many of the participants had difficulty with the analysis of zinc-65 due to the low activity of this radionuclide remaining in the sample. However, the poor results for this radionuclide highlighted the fact that five of the nine participants chose to measure volumes that maximised both measurement uncertainties (or counting times) and the effects of True Coincidence Summing. Nonetheless, this radionuclide was discarded from further consideration.

In almost all cases, the participants reported results that would be considered acceptable by the supplier of the samples. That is, 42 of the 45 reported results that were within the PALs defined by the supplier.

However, the PALs tolerate common errors made by laboratories, such as the failure to correct for TCS effects. Therefore, the reported results were also compared with the reference value given by the supplier. This analysis showed that only five of the nine laboratories were able to provide results that were consistent with the reference value, for all radionuclides.

Cs-137 is a key indicator of radioactive contamination. It is, therefore, concerning that three of the participants reported discrepant results for this radionuclide. Therefore, these laboratories may require assistance before they could be relied upon to provide assistance for some radiological incidents, particularly those involving the dispersion of radioactive material created in a nuclear reactor.

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