Journal of Radioanalytical and Nuclear Chemistry, Vol. 269, No.2 (2006) 351–360

Radioanalytical emergency response exercise

K. G. W. Inn,[1]* I. Outola,[1] S. Nour,[1] H. Kurosaki,[1] L. Albin,[2] A. Berne[3]

[1] *NIST, 100 Bureau Dr., MS 8462, Gaithersburg, MD, USA*
[2] *Washington State Department of Health, Olympia, WA, USA*
[3] *Environmental Measurements Laboratory, Department of Homeland Security, New York, NY, USA*

(Received April 6, 2006)

The National Institute of Standards and Technology (NIST Radiochemistry Intercomparison Program [NRIP]) and the Environmental Measurements Laboratory (EML Performance Testing [PT] 0904) recently conducted two separate radiological emergency preparedness exercises to evaluate participating radioanalytical laboratories' capability of making measurements under a short time constraint. Results of the exercises demonstrated that radioanalytical laboratories can respond within eight hours to better than a factor of two, laboratories need to do a better job of estimating uncertainties of their measurements, the quality of the laboratory capabilities must be appropriate and demonstrated prior to real emergencies, and exercises will help laboratories and consequence managers be prepared for the real event.

Introduction

Emergency response plans for a radiological incident include protection of human health, rapid and efficient resumption of crucial commerce, remediation for re-occupancy, and environmental stewardship. Radiological emergency response requires timely, reliable and adequately good radioassay measurement information, acquired under an organized quality system, to make appropriate decisions throughout the entire incident response scenario. Part of the assessment of measurement capability and capacity includes the evaluation of radioassay laboratory response preparedness.

Because the national radioassay laboratory emergency response capability/capacity are not well known, the National Institute of Standards and Technology (NIST/Department of Commerce) and the Environmental Measurements Laboratory (EML/Department of Homeland Security) conducted two separate studies in 2004 to evaluate radioassay laboratory response, identify gaps, and start improvements for laboratory preparedness. The results of the studies were to provide decision-makers and the radioassay community with an initial evaluation of the state of the readiness of response of laboratories. The studies evaluated: (a) measurement capability, (b) measurement capacity, (c) effect of counting time, (d) accuracy (traceability), (e) measurement confidence (uncertainties), and (f) operational issues.

NRIP Protocols

A master solution was gravimetrically prepared from NIST Standard Reference Materials. The master solution's massic activity (Bq/g) was verified by confirmation measurements with standard combined uncertainties of less than two percent for each radionuclide in the mixture. Each blank sample was then gravimetrically spiked with the master solution, and verified by wide-window NaI confirmation measurements with counting uncertainties better than one percent. All of these quality check verification measurement sets were also examined for deviations from a normal distribution. Any sample exhibiting a deviation from the population distribution would be set aside as being suspect and not used in the exercise.[1]

The six participating laboratories received five replicate samples and 3 blanks (5 blanks for soil) of up to 4 matrices. Sample matrices for NRIP'04 included spiked soil, water, air filter, synthetic urine, and synthetic feces. Participants were instructed to analyze and report measurements of gamma-ray emitters (^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu), gross alpha, gross beta and radioisotopes (^{90}Sr, ^{230}Th, ^{234,235,238}U, $^{238,239/240}$Pu, and ^{241}Am) within eight hours of sample receipt. The range of test activity was 0.04 Bq/sample to 900 Bq/sample per radionuclide.

Laboratory selection of NRIP'04 test matrices (based on their capabilities), and radioanalytical result sets are depicted in Tables 1 and 2, respectively.

Table 1. Laboratory selection of NRIP'04 test matrices

	Matrix				
Lab	AF	AW	SS	SU	SF
1		X			
2	X	X	X		
3	X	X	X		
4	X		X	X	X
5	X	X			
6	X	X			

AF: Air filter.
AW: Acidified water.
SS: Spiked soil.
SU: Synthetic urine.
SF: Synthetic feces.

* E-mail: kenneth.inn@nist.gov

0236–5731/USD 20.00

Akadémiai Kiadó, Budapest
Springer, Dordrecht

Table 2. Radionalytical result sets reported by the laboratories

Analysis	AF	AW	SS	SU	SF
Gamma	5	5	3	1	1
Gross alpha	1	2	1	0	0
Gross beta	1	2	1	0	0
Radiochemical	0	2	0	0	0

AF: Air filter.
AW: Acidified water.
SS: Spiked soil.
SU: Synthetic urine.
SF: Synthetic feces.

EML PT 0904 Protocol

Seventeen laboratories participated in PT0904. Two sets of air filters were prepared, each set spiked uniformly with a combination of ^{60}Co, ^{137}Cs, and ^{241}Am. Spike set No. 1 contained 350 Bq of ^{137}Cs, 8.5 Bq of ^{60}Co and 0.29 Bq of ^{241}Am, while spike set No. 2 contained 46, 180 and 0.37 Bq of the same respective radionuclides. The activities chosen for each set were based on early phase Response Levels,[2] assuming collection of 1000 liters of air sampled per filter. Each laboratory received one PT air filter from each set and was instructed to report the results based on a 15-minute and a 60-minute count. The results were expected "as soon as possible."

Results

NRIP acceptance criteria

The acceptance criterion used was from ANSI N42.22[3] where the difference between the reported value and the NIST value should be less than three times the combination of the laboratory's and NIST's standard uncertainties (i.e., *traceability limit* at ninety-nine percent confidence). The Figs 1 to 6 to follow depict the NIST value as the horizontal line, the laboratory's value, and the vertical bars represent the traceability limits (three times the combination of the laboratory's and NIST's standard uncertainties). Reported values that are included within the traceability limits are interpreted as a demonstration of traceability, with ninety-nine percent confidence.

For radiobioassay test results (synthetic urine and synthetic fecal materials), the ANSI N13.30[4] acceptance criteria for bias (–25% ≤ bias (%) ≤ +50%), and precision (standard deviation of the bias ≤ 40%, $n \geq 5$) were also used to assess the laboratory's capabilities.

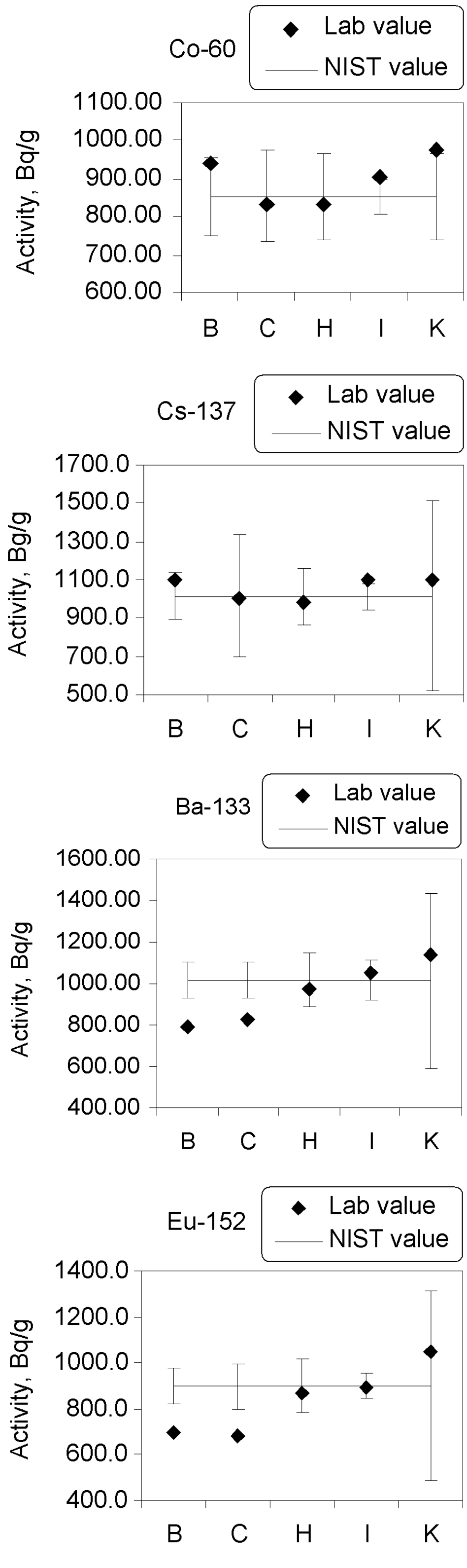

Fig. 1. NRIP air filter gamma-ray results (laboratory value/NIST value; vertical bars are the traceability limits for the measurement)

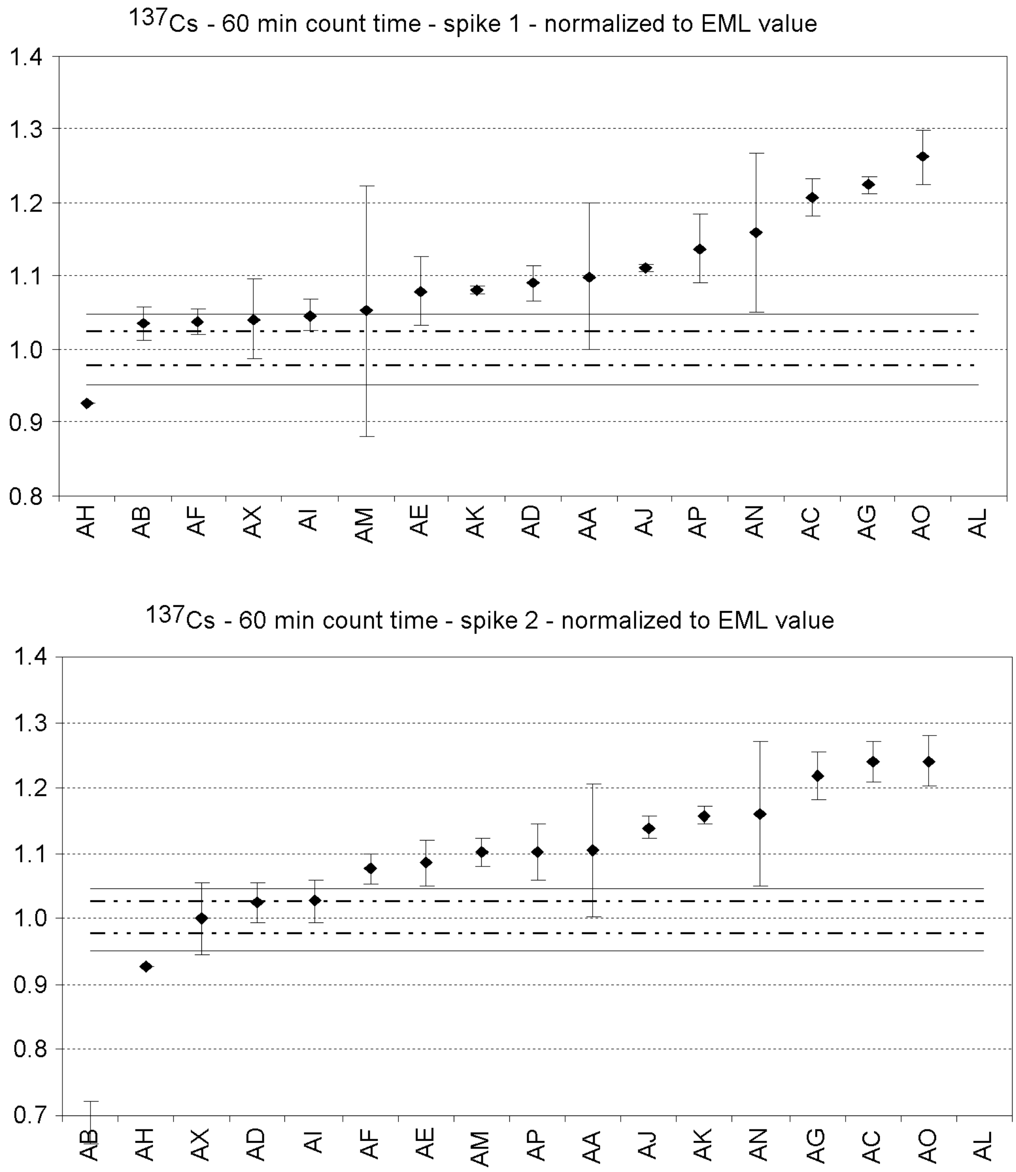

Fig. 2. EML PT0904 ^{137}Cs and ^{60}Co on air filter results (1 hour count; laboratory value/EML value); vertical lines are expanded uncertainty ($k = 2$)

NRIP air filters, gamma-ray emitters

Spiked air filters were analyzed for gamma-ray emitting radionuclides by five laboratories; four laboratories analyzed glass fiber filters, and one laboratory analyzed ashless paper filters. Activities reported for ^{137}Cs and ^{60}Co were within the traceability limits. Some of the reported activities for each of the other radionuclides fell outside of the traceability limits (Fig. 1), perhaps because of summing effects.

NRIP air filter, gross alpha and gross beta analyses

Only one laboratory analyzed the glass fiber filters for gross alpha and beta constituents. After gamma-ray spectroscopy, the air filter samples were returned intact and the samples were acid washed and placed in a sonic water bath. The rinse solution was decanted, and the filters were again treated with acid, placed again in the sonic water bath and the rinse solution decanted. The decanted solutions for each sample were then combined, dried on a planchet under an infrared lamp and flamed prior to gross alpha/beta counting. The gross alpha activity result was found to be within the traceability limit, but the reported gross beta activity exceeded the traceability limit for the test.

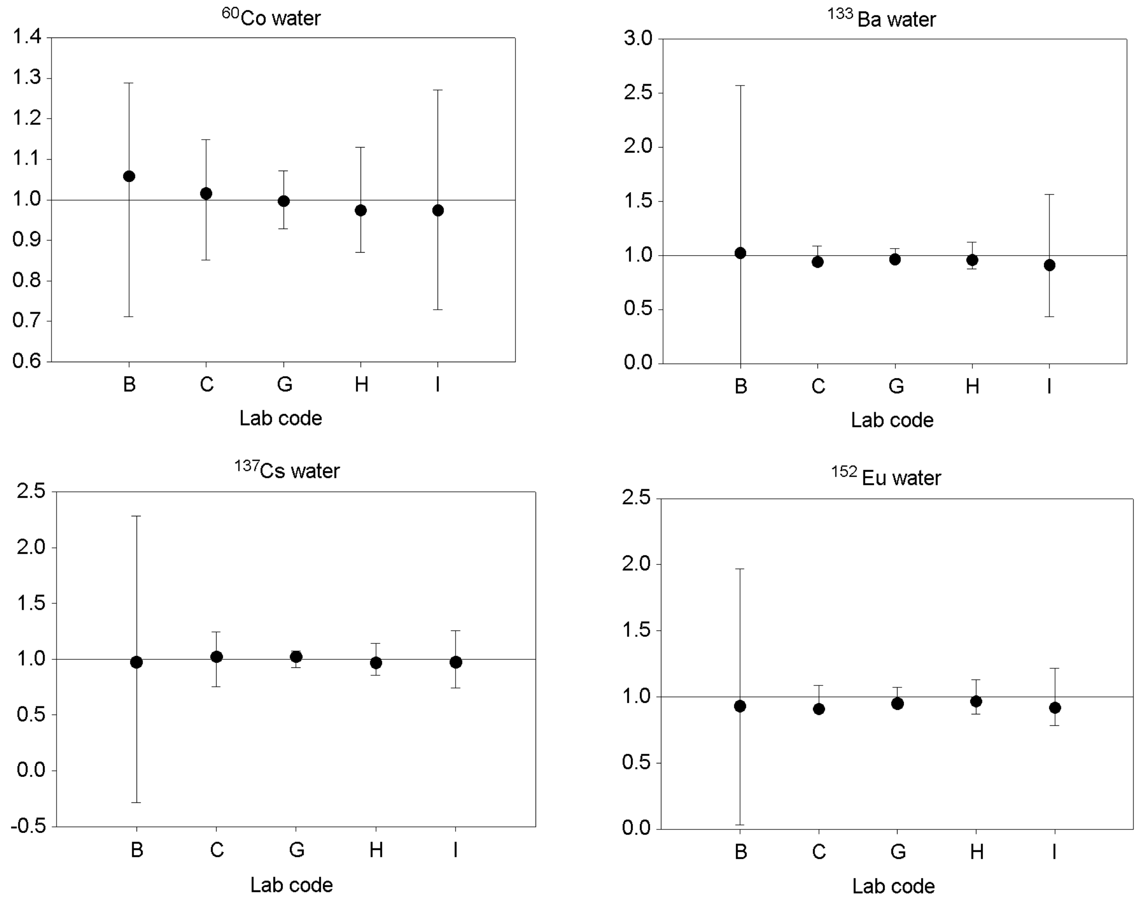

Fig. 3. NRIP acidified water gamma-ray results (laboratory value/NIST value; vertical bars are the traceability limits for the measurement)

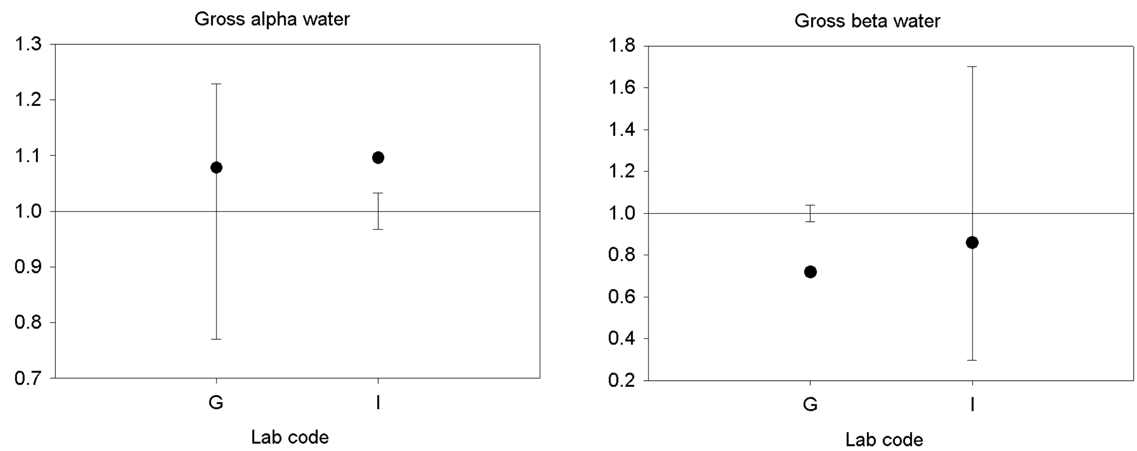

Fig. 4. NRIP water gross alpha and gross beta results (laboratory value/NIST value; vertical bars are the traceability limits for the measurement)

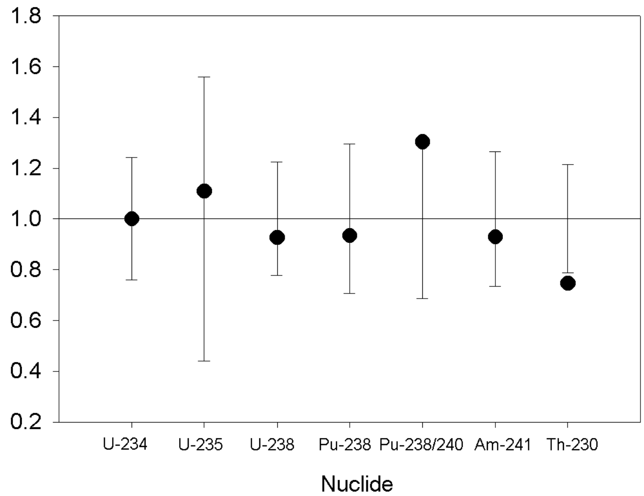

Fig. 5. NRIP radiochemical analysis results for acidified water; only one laboratory (laboratory value/NIST value; vertical bars are the traceability limits for the measurement)

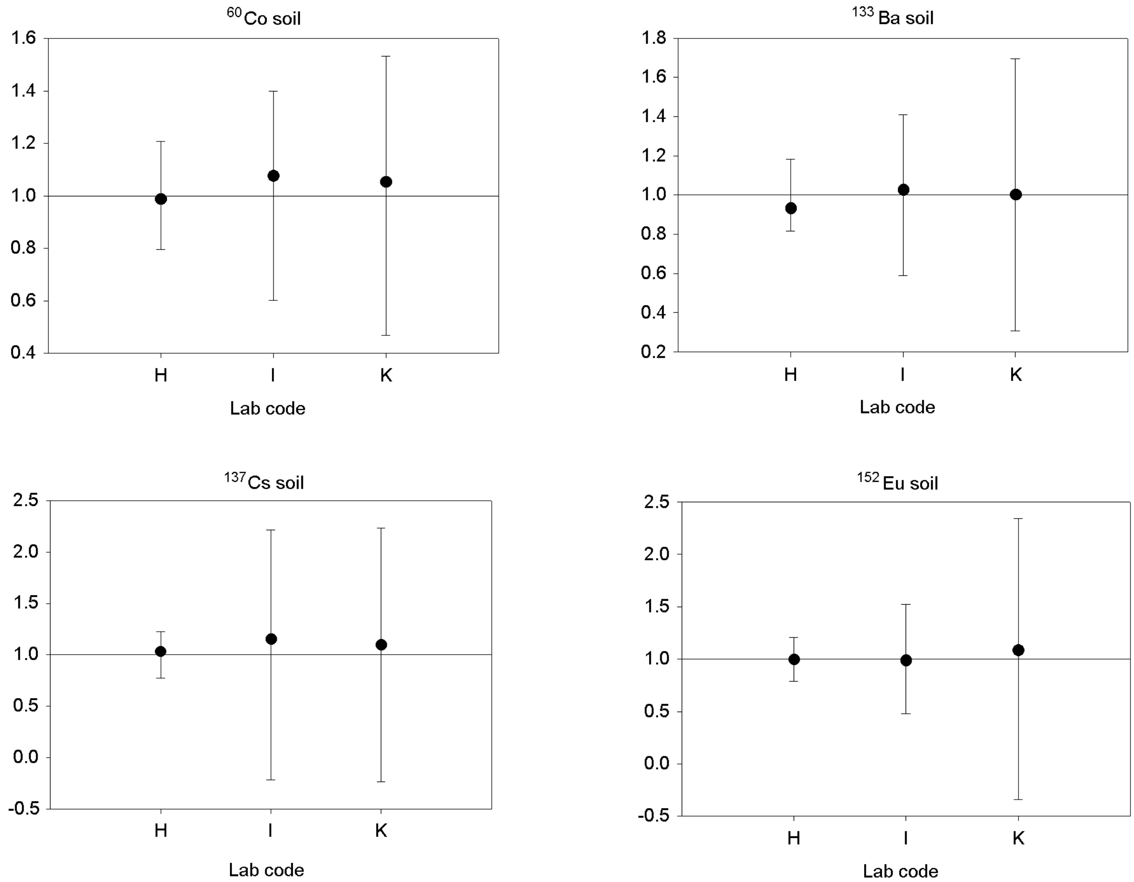

Fig. 6. NRIP soil gamma-ray results (laboratory value/NIST value; vertical bars are the traceability limits for the measurement)

EML air filters

While the results for ^{60}Co and ^{137}Cs were in general acceptable (that means that most laboratories agreed with each other), most labs could not detect ^{241}Am. The combination of low gamma-detection efficiency and a very low Action Guideline for ^{241}Am resulted in levels of activity on the PT filter below a typical minimum detection activity for the counting times required. While most laboratories were fairly competent in analyzing these simple gamma-spectra, two issues became apparent: (1) difficulties in processing the sample quickly (turnaround time ranged from 3.6 to 31.5 hours), and (2) problems with reporting the results in the required format.

Figure 2 summarizes the results for the 60-minute measurements at the highest ^{60}Co and ^{137}Cs activity levels. Contrary to expectations, the results for the 15-minute counting time and the lower activity levels were very similar to the longer counting time at higher activity levels. The dashed and solid lines on the graphs are, respectively, the combined standard uncertainty (u_c) and the expanded uncertainty (U, $k=2$) for each EML reference value and should not be construed as acceptance criteria. The laboratory uncertainty bars (u_c) are based on the reported values.

NRIP acidified water, gamma-ray emitters

Five laboratories analyzed samples for gamma-ray emitters in water. All laboratories reported ^{60}Co ^{133}Ba, ^{137}Cs and ^{152}Eu activities that were within the traceability limits (Fig. 3). There was no apparent unaccounted summing issue among the participating laboratories for the water matrix. However, laboratory B may have overestimated its uncertainty.

NRIP acidified water, gross alpha and gross beta

Two laboratories reported gross alpha and gross beta results (Fig. 4). The difference between the laboratory results and the NIST value may partly be due to differing counting efficiencies because of the mixture of alpha and beta emitting radionuclides in the samples was not the same as that used to calibrate the laboratories' instruments. The results also indicate a need for better estimates of measurement uncertainties.

NRIP acidified water, radiochemical measurements

One laboratory performed radiochemical analyses within the 8 hours timeframe. The results for ^{241}Am, $^{238,239/240}$Pu, ^{234,235,238}U and ^{230}Th are displayed on Fig. 5.

NRIP spiked soil

Three laboratories analyzed synthetic soil for gamma emitting radionuclides. Two laboratories analyzed the soil samples as a solid. The third laboratory leached the soil in nitric acid and counted the solution. Laboratories counted samples on high-purity germanium detectors, and reported activities for ^{60}Co ^{133}Ba, ^{137}Cs and ^{152}Eu. All laboratory results were within the traceability limits (Fig. 6). One laboratory analyzed the sample for gross alpha and gross beta activities. The gross alpha activity reported was within the traceability limit while the gross beta exceeded the traceability limit.

NRIP synthetic feces and urine

Only one laboratory chose to analyze synthetic feces and urine matrices. A comparison of the NIST value and the laboratory's reported value for the four gamma-emitting radionuclides for each matrix is given in Table 3. Sample results were within expected combined laboratory uncertainties of the spiked activity.

Synthetic urine reported values were approximately 60% low for all synthetic urine samples. Reported values exceeded the traceability limit.

NRIP average laboratory results

Although this study was conducted on a limited number of laboratories, is worthwhile to get an overview of the measurement capabilities. The expected result for the average laboratory are reported in Table 4, i.e., the average percent difference from the NIST values as related to matrix and classification of measurements, the range of laboratory mean values, and the average traceability limit. The average percent difference from the NIST values across all matrices and measurement type is about 17% while the average traceability limit is about 32%. As mentioned previously, these results need to be viewed carefully because of the limited number of participants in the study. None the less, these results indicate that the average over many measurements from many laboratories would fall within about thirty-two percent of the expected value ninety-nine percent of the time.

EML PT 0904 average results

The expected results for the average laboratory participating in PT 0904 are reported in Table 5. Irrespective of activity level on the filter, allowed counting time and turnaround time, the average percent difference from the EML value for ^{137}Cs and ^{60}Co were 9 and 0.9%, respectively, and the average standard deviation for the ^{137}Cs and ^{60}Co measurements were 12 and 8%, respectively. Furthermore, the range of reported values for the ^{137}Cs and ^{60}Co measurements were from –31 to 26%. These results are consistent with the overall results from the NRIP study.

Table 3. NRIP urine and fecal gamma-ray results for one laboratory

Nuclide	NIST Massic activity, Bq/g	NIST Value Relative expanded uncertainty, %, k=2	Reported Massic activity, Bq/g	Reported Value Relative expanded uncertainty, %, k=2
Synthetic urine				
^{60}Co	853.2	0.7	330	7.8
^{133}Ba	1014.8	0.52	360	12
^{137}Cs	1013.4	0.68	380	8.9
^{152}Eu	899.6	0.73	380	17
Synthetic feces				
^{60}Co	853.2	0.7	680	22
^{133}Ba	1014.8	0.52	780	15
^{137}Cs	1013.4	0.68	760	11
^{152}Eu	899.6	0.73	740	22

Table 4. NRIP average percent difference, range of results,* and ANSI traceability limit**

Analysis	Air filter	Water	Soil	Urine	Feces
Gamma	–1.3 [–24 to +16] (12)	–2.7 [–9 to +6] (35)	3.6 [–7 to +15] (56)	–61 (11)	–22 (17)
Gross alpha	9.1 (30)	8.8 [+8 to +10] (12)	10 (4)		
Gross beta	22 (21)	–22 [–28 to -14] (44)	–43 (112)		
Radiochemical		–4 [–25 to +11] (31)			

* Range of laboratory mean results as percent are noted in square brackets.
** ANSI traceability limits as percents are noted in parentheses, and are three times the standard combined uncertainties from the laboratory result and the reference value.

Table 5. EML PT0904 average laboratory results

Sample	Bq/filter	Diff., % 15 min count	SD, % 15 min count	Diff., % 60 min count	SD, % 60 min count	Diff., % range
^{137}Cs-1	351.58	9.6	10	10	8.4	–7 to +26
^{137}Cs-2	46.25	7.5	14	8.1	14	–31 to +24
^{60}Co-1	8.544	0.47	8.2	–1.1	10	–22 to +18
^{60}Co-2	181.0	0.31	8.0	1.6	7.5	–7.4 to +18
^{241}Am-1	0.287	79	170	–7.0	140	–280 to +140
^{241}Am-1	0.367	72	82	32	57	–65 to +120

SD: Standard deviation.

The average results from the ^{241}Am measurements were poor because the amounts of activity used in this exercise were near the detection limits for the participating laboratories.

Discussion

The NRIP study revealed that the laboratories could respond with analytical results within eight hours after receiving samples. To respond within this time frame, some laboratories needed to modify their standard operating procedures. While the results would indicate that good technical decisions were made to modify the standard operating procedures, the consequence was to break links to the quality control system that the procedures rely on to demonstrate process control. This exercise, and others to follow, will provide the laboratories with the opportunity to optimize their emergency response procedures, validate them and establish control systems to assure “fit for use” measurement results.

The largest number of measurement results were for gamma-ray emitting radionuclides, presumably because these measurements could be quickly done without much sample preparation and manipulation. The next most reported results were from gross alpha and gross beta measurements. These measurements are an important tool that can be completed fairly rapidly because after some initial sample preparation, no additional time is needed for long chemical separations. Only one laboratory reported radiochemical results within eight hours, and demonstrated that relatively accurate radiochemical analyses can be obtained in the required short time. With radiochemical method streamlining, training and exercising, other laboratories would be able to also do rapid measurements of radiochemically separated radionuclides.

For this limited study, the average measurement (across all matrices and measurement types) would be within about twenty-five to thirty percent of the expected value with 99% confidence. This result needs to be viewed in the context that the range about this value is very wide, depending on the matrix, measurement type and laboratory capability and capacity. Future exercises will be able to confirm this finding, and with experience, it is anticipated that measurement capabilities can improve measurably.

The assessment of the radioassay capabilities is dependent on each laboratory's capability to make real uncertainty statements for its measurements. The study revealed that some of the participating laboratories underestimate their uncertainties while others overestimate their uncertainties. Both groups of laboratories are in need of information, instruction to improve their ability to estimate measurement uncertainties.

All of the lessons learned from these exercises indicates the preparedness exercise (1) needs to be expanded to a larger cohort of laboratories to better assess the capabilities and capacity of the national radioanalytical community, (2) needs to strengthen the laboratory's capabilities to be prepared for an emergency incident, and (3) a quality system needs to be built to improve statistical control over the laboratory's response capability. As these improvements are put in place, it can be expected that emergency radioanalytial response capabilities will be able to provide measurement results with predictable and known accuracy and acceptable uncertainty.

Laboratories participating in the EML PT 0904 exercise reported their results in 3 to 28 hours after the samples were received. Since the average percent difference for ^{137}Cs was the same irrespective of the activity on the filter, turnaround time and the counting time allotted, it appears that there was some systematic influence involved, and type B sources of uncertainty that were not incorporated in the estimate. Because the intervention level and the measurement detection limit for ^{241}Am are nearly the same, better sensitivity for ^{241}Am measurements needs to be developed.

Over the course of the exercises several operational improvements in the conduct of the exercises were revealed. Simply sending samples by priority courier was not sufficient to assure direct transfer of the materials to the laboratory. At least in two cases, samples were sent to a transfer point and then were delayed because higher priority packages needed to be transferred before the samples could resume their trip to the final destinations. Although it is not likely that delays because of weather disturbances can be avoided in the future, the highest cost-effective shipping priority should be selected to minimize delay in transfer.

Due to the time restriction of the exercises, misunderstandings in calculating and reporting results at several laboratories resulted in heightened confusion and increased stress. Before starting a future exercise, a standard reporting format that is understood by all must be established. Furthermore, participant laboratories should be trained to estimate measurement uncertainties. These issues could be addressed by conference call, e-mail transfers, and/or private discussions.

Some laboratories doing gamma-ray measurements were forced to count the samples in non-standardized geometries because the samples they received did not come in the routine standard containers that they receive on a daily basis for their business arrangements. Future shipments of test samples in pre-agreed upon containers would allow the laboratory to set up calibrations for pre-determined geometries prior to receipt of samples. This modification would allow the laboratories to improve measurement accuracy and reduce their measurement uncertainties.

The Certificate reporting the laboratories' inter-comparison results was mailed to the laboratory a month following the exercises. By sending the laboratories their test results much sooner, they will be able to take corrective action in a timelier manner.

This exercise was a learning experience for all involved. Laboratories fully participating in this exercise received more than thirty samples from four matrices to analyze and report within an eight hour period. When possible, laboratories used their standard operating procedures for emergency conditions. Many laboratories found that standard processing and counting procedures needed to be modified to analyze the matrices and sample sizes supplied within the specified time criteria. Laboratories did not solve problems in identical ways. Participants agreed that it would be helpful to share and discuss the merits of the different methodologies so every laboratory's capability could be improved.

Reported activities were evaluated per ANSI N42.22. This method describes an acceptance criterion that is dependent on reported uncertainties. Reported expanded uncertainties ($k=2$) were as follows: 3 to 100% for gamma-ray measurements, 1 to 18% for gross alpha measurements, 3 to 17% for gross beta measurements, and 14 to 21% for radiochemical results. Laboratories did not all calculate the expanded uncertainties in the same way. Some laboratories used only counting errors while others expanded the calculation to include operational factors. This exercise emphasized the importance of understanding uncertainties. Participants are encouraged to re-evaluate uncertainties and use more realistic uncertainty estimates. Better understanding of uncertainty leads to better confidence in the data. This, in turn, allows for better confidence in decisions that are made based on the reported data.

The purpose of NRIP exercises is to help laboratories achieve and maintain good measurement capability and consistency with the national measurement system. In a customary (non-emergency) exercise, the performance of a laboratory would be evaluated and upon successful determination of analytes in a specific matrix, a Certificate of Traceability would be issued. Since NRIP'04 was the first emergency preparedness exercise, no traceability acceptance criteria were applied. Participating laboratories were issued a Report of Traceability by NIST which lists NIST and the laboratory's reported values and expanded uncertainties for each reported radionuclide in each matrix.

Analysis of the operational flaws revealed in the NRIP'04 and PT0904 exercises will be very useful in conducting future emergency preparedness exercises. For first emergency exercises, NRIP'04 and PT0904 were successes. They proved an invaluable tool to the participating laboratories to begin the process of improving their response capabilities to provide measurement results of reliable accuracy.

Future actions

The lessons learned from these exercises have indicated several areas for future development that would help the radioanalytical community to better respond to a radiological emergency, including: (a) NIST should develop mechanisms to encourage capability improvement through technical exchange between laboratories, (b) NIST should develop informational guides so that laboratories can estimate measurement uncertainties in a consistent manner, (c) consequence manager should develop and adopt appropriate and consistent acceptance criteria and measurement quality objectives for emergency response laboratory certification/accreditation, (d) NIST should seek increased participation in its traceable radioassay preparedness exercises, (e) the laboratories should develop improved radiochemical alpha and beta assay capability and capacity, and (f) the Department of Homeland Security should develop guidelines for decision makers to better understand the relevance of measurement uncertainty at various stages of an emergency response.

Conclusions

Future improvement would be realized when decision makers can answer the questions of "how good is good enough?" and "how much confidence is needed to make a sound response decision?" for the radioassay community. Armed with the answers to these questions, the radioassay community could then develop measurement quality objectives[5] and tolerance limits for the individual steps in the measurement process to assure the decision maker of the confidence that can be placed in the measurement information that is to be used for saving lives, restoring commerce, and dealing with long-term remediation issues.

The NRIP and EML radiological emergency exercises have shown, although on a small scale, that the radioassay community can fairly quickly provide gamma-ray and gross alpha/beta screening measurement results for consequence management decision making. It was also demonstrated that radiochemical measurements of relatively high accuracy are possible with an eight hour turnaround.

However, the studies also have shown that there is large variability in the accuracy of the radioassay measurements. This exercise, and others to follow, will provide the laboratories with the opportunity to optimize their emergency response procedures, validate them and establish control systems to assure "fit for use" measurement results. With radiochemical method streamlining, training and exercising, other laboratories would be able to also do rapid measurements of radiochemically separated radionuclides. With experience, it is anticipated that measurement capabilities and the ability to estimate measurement uncertainties can be improved measurably.

All of these lessons indicates the preparedness exercises (1) need to be expanded to a larger cohort of laboratories to better assess the capabilities and capacity of the national radioanalytical community, (2) strengthen the laboratory's capabilities to be prepared for an emergency incident, and (3) a quality system needs to be built to improve statistical control over the laboratory's response capability. As these improvements are put in place, it can be expected that emergency radioanalytial response capabilities will be able to provide measurement results with predictable and known accuracy and acceptable uncertainty.

*

The authors respectfully acknowledge the technical contributions from the participating laboratories in the studies: Mansour AKBARZADEH (DOE/WIPP), Shirley BELL (NC), Roger BREWER (SC), Don BROWN (TX), Donald BUCKLEY (MA), Stan CARRENDER (MO), Roy E. DUNKER (ID), Bernd KAHN (GA), Michael KITTO (NY), Tony HARRISON (CO), Don HENDRIKSE (WI), Richard LARSEN (EML/DHS), Huei MEZNARICH (Fluor Hanford/WSCF), Kirk NEMETH (NJ), Bahman PARSA (NJ), Shiyamalie RUBERU (CA), Marina SILVERSTONE (WA), Jane SMITH (IN), Dominic TO (KS), Jeng-Jong WANG (INER, Taiwan), and Mary WISDOM (EPA/NAREL).

References

1. Z. WU, K. G. W. INN, Z. C. LIN, C. A. MCMAHON, L. R. KARAM, Appl. Radiation Isotopes, 56 (2002) 379.
2. Manual for Protective Action Guides and Protective Actions for Nuclear Incidents, U.S. Environmental Protection Agency, Office of Radiation Programs, Washington, D.C., 1992.
3. American National Standard: Traceability of Radioactive Sources to the National Institute of Standards and Technology (NIST) and Associated Instrument Quality Control; ANSI N42.22-1995, Institute of Electrical and Electronics Engineers, Inc., New York, NY, 1995.
4. American National Standard: Performance Criteria for Radiobioassy, ANSI N13.30-1996, Health Physics Society McLean, VA, 1996.
5. Multi-Agency Radiological Laboratory Analytical Protocols Manual, NTIS PB-2004-105421, Springfield, VA, 2004.