LOW-LEVEL WASTE DRUM ASSAY INTERCOMPARISON STUDY

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ABSTRACT

Nuclear waste assay is an integral element of programs such as safeguards, waste management, and waste disposal. The majority of nuclear waste is packaged in drums and analyzed by various nondestructive assay (NDA) techniques to identify and quantify the radioactive content. Due to various regulations and the public interest in nuclear issues, the analytical results are required to be of high quality and supported by a rigorous Quality Assurance (QA) program. A valuable QA tool is an intercomparison program in which a known sample is analyzed by a number of different facilities. While transuranic waste (TRU) certified NDA teams are evaluated through the Performance Demonstration Program (PDP), low-level waste (LLW) assay specialists have not been afforded a similar opportunity. NDA specialists from throughout the DOE complex were invited to participate in this voluntary drum assay intercomparison study that was organized and facilitated by the Solid Waste Operations and the Safeguards Science and Technology groups at the Los Alamos National Laboratory and by Eberline Services. Each participating NDA team performed six replicate blind measurements of two 55-gallon drums with relatively low-density matrices (a 19.1 kg shredded paper matrix and a 54.4 kg mixed metal, rubber, paper and plastic matrix). This paper presents the results from this study, with an emphasis on discussing the lessons learned as well as desirable follow up programs for the future. The results will discuss the accuracy and precision of the replicate measurements for each NDA team as well as any issues that arose during the effort.

INTRODUCTION

A blind intercomparison measurement program can be a valuable quality assurance (QA) tool for a variety of reasons. Performing well on such 'blind' assay attempts will enable customers and clients to have confidence in the quality of the measurement data provided to them. Furthermore, programs such as this can also identify deficiencies or limitations with measurement systems, and provide an opportunity to correct problems in processes. While TRU certified NDA teams are evaluated through the Waste Isolation Pilot Plant (WIPP) PDP, LLW assay specialists have not been afforded a similar opportunity. The idea for this program originated at the Public and Environmental Radiation Protection (PERP) [a subcommittee of the National Institute of Standards and Technology (NIST)] Workshop on Drum Assay Intercomparison Programs held October 30-31, 2000 at the annual CIRMS (Council on Ionizing Radiation Measurements and Standards) Conference. The Solid Waste Operations (SWO) group and the Safeguards Science and Technology (NIS-5) group at Los Alamos National Laboratory took up the challenge of organizing and facilitating the program. In February 2002, NDA specialists from throughout the DOE complex were invited to participate in this controlled low-level waste drum assay intercomparison study.

Each measurement system was evaluated on the accuracy and precision of their six replicate measurements on two relatively simple 55-gal drums: 1) a low-density drum filled with 19.1 kg (42 lbs) of shredded paper, and another drum filled with 54.4 kg (120 lbs) of a mixed matrix. Accuracy is determined as the mean value of the six replicate measurements compared to the actual value, while precision is the percent relative standard deviation of those six results. It was decided to keep the assay challenges to a minimum in this inaugural effort in order to encourage participation. Therefore, the drum matrices used in the study are relatively low-density and had very uniform matrix distributions as well. Furthermore, the sources are very common gamma-ray emitters (a Co-60, a Cs-137, and a Eu-152 source) with easily detectable quantities and no gamma-ray interferences under normal circumstances.

The sources have a low self-absorption quality so that additional corrections were not needed for gamma-ray attenuation losses in the source itself. All sources also met the following general conditions: 1) they have a primary gamma-ray emission with a minimum yield of 10% per decay and an energy between 100 keV and 2000 keV, and 2) a minimum activity of 2.0 microcuries. The position of the source(s) within each drum was determined randomly. The source(s) could be placed in any of three tubes at any vertical position (up to a maximum of 30 inches from the bottom). The sources were configured in the tubes in a manner that prevented them from shifting during normal assay activities and the stresses encountered during shipment. All sources were chosen and loaded by an unbiased individual from the LANL Safeguards Science and Technology Group (NIS-5). Therefore, the measurements were 'blind' to all participants.

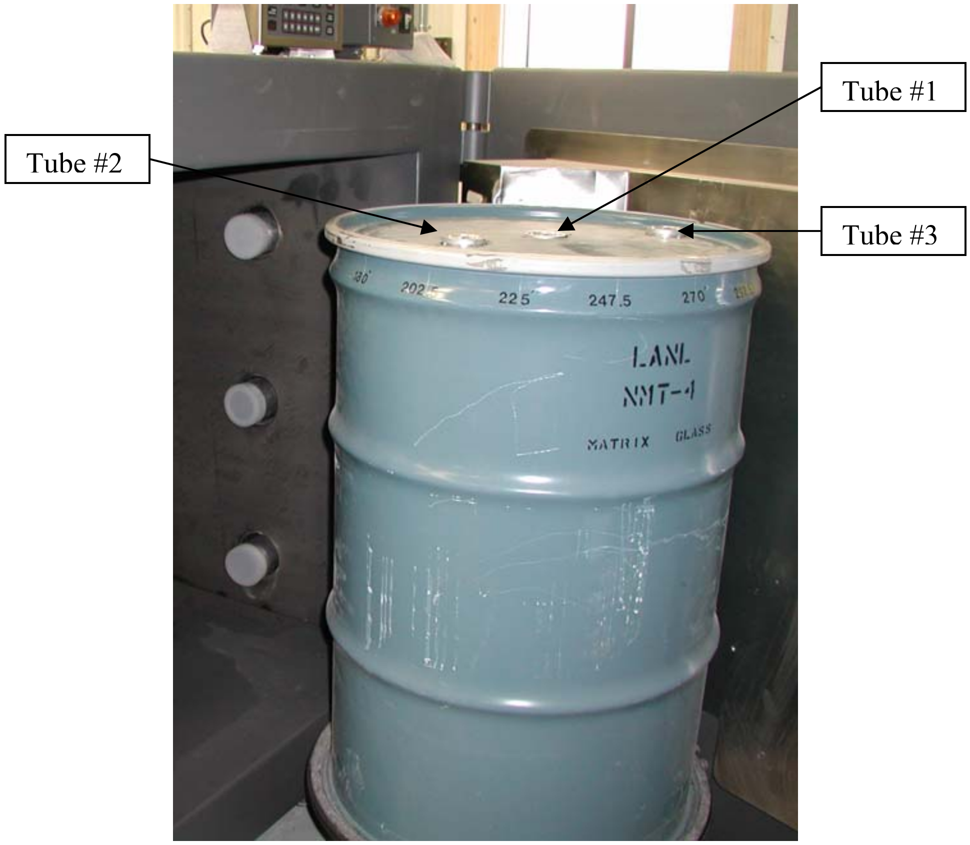
Both drums are PDP-like drums that were initially designed for use in the DOE Performance Demonstration Project program. The drum labeled as the 'Zero Matrix' drum was re-configured to contain uniformly distributed shredded paper. The drum labeled as 'Mixed Metals' has a combination of both metals and low-density materials (including steel, aluminum, copper, rubber, plastic, and paper). The mixed metals drum does <u>not</u> contain any highly dense metals like lead or tungsten.

The following is a thorough physical description of each of the drums (figure 1):

a. Height of outer metal drum: 88.27 cm (34.75 inches).
b. Diameter of outer metal drum: 56.83 cm (22.375 inches).
c. Wall thickness of outer drum: 0.159 cm (0.0625 inches) of carbon steel.
d. There is an inner drum liner of 0.23 cm (0.09 inches) of polyethylene. The inner diameter of the poly liner is 54.6 cm (21.5 inches). The height of the poly liner is 76.84 cm (30.25 inches). All matrix materials and sources reside inside the confines of the poly liner.
e. Each vertical tube is actually a combination of a solid outer tube of 0.159 cm (0.0625 inches) of aluminum and a slotted inner tube of aluminum of the same

thickness. The slotted inner tube is removable while the solid outer tube is fixed in place. The diameter of the solid outer tube is 5.40 cm (2.125 inches) and the slotted inner tube is 5.08 cm (2.0 inches).

Figure 1: Photograph of a PDP-like drum and tube configuration



Further dimensions and descriptions of these drums are available on the Internet at **http://www.wipp.carlsbad.nm.us/library/caolib.htm**. Go to "National Transuranic Program Requirements Documents" and choose the document titled "Performance Demonstration Program Plan for Nondestructive Assay of Drummed Wastes for the TRU Waste Characterization Program". Proceed to Appendix D, "Specifications for Radioactive PDP Standards".

GROUND RULES

The following set of ground rules were established and implemented in order to facilitate an honest, organized, and efficient process:

1. By participating in this program you have obligated your facility to absorb all cost related to assaying the drums and producing your final results for this study.

Shipping will be done by LANL shipping personnel in coordination with your facility's shipping personnel without revealing to you the source identification or quantity.

2. Each of the two drums should arrive with an intact wire tamper indicating device (TID) across the three vertical tubes extruding from the top of the drums. Check the TID by removing the two nuts holding the protective bar and assuring that the TID is intact. Please notify Jozef Kuzminski (505) 667-9062, immediately if the TID has been removed or shows signs of tampering. Participants are not allowed to open the drums or remove the metal tubes during this exercise.
3. You may keep the drums in your possession for a one-week period of time, after which they must be sent back to LANL. We will forward them on to the next participating facility. You must make a formal request to Jozef Kuzminski for exceptions to this ground rule. If the assay system(s) to be used in this study are not functioning properly prior to your scheduled week, please contact us so we can re-schedule your effort at a later date.
4. You may measure the drums as many times as you would like in as many configurations as you choose. However, your final results for each drum must be based upon six consecutive assays performed in the same manner as your 'normal' assay measurements. A full description of your assay process should accompany the reported results as well as a description of the assay system that was used. Report your activity results in the units that they are normally reported in – but make sure the units are specifically indicated (Ci, Bq, etc). There is no need to report concentration or nuclide mass unless that also happens to be part of your normal reporting process.
5. Final results should be sent within 14 calendar days after your measurements have been completed. Reports can be sent either by regular postal mail or electronically to Jozef Kuzminski at one of the following addresses:
    a. P.O. Box 1663, MS E540, Los Alamos, NM 87545
    b. **josephk@lanl.gov**
6. Each participating facility will receive notification of how their results fared compared to the known values once results from all participants have been received. All participants will also receive a copy of the final paper on the program once it has been completed.

RESULTS

Table I presents the official comparisons for each of the participating systems in the study. All systems correctly identified the source nuclides in each case, which was expected. The average percent bias among all systems for Co-60, Cs-137, and Eu-152 are as follows: -6.38%, +21.50%, and +19.29%. The results fell within ±30% from the known value in 16 of the 18 individual results.

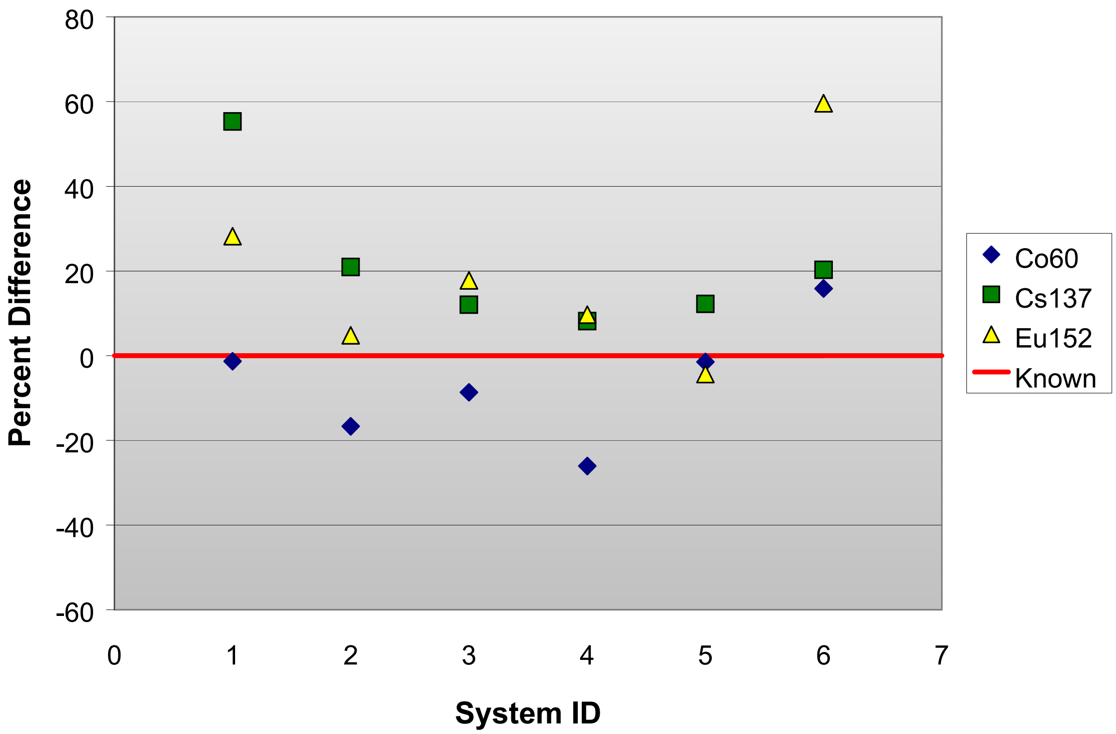
Table I. Comparisons (% Bias and Precision) to Known Values

			Mixed Matrix		Paper Matrix
System ID	**Description**	**Analytical Technique**	**Co-60**	**Cs-137**	**Eu-152**
1	Portable HPGe	Modeling Routine	**-1.32% (1.30%)**	**55.31% (1.60%)**	**28.20% (3.39%)**
2	Portable HPGe	Modeling Routine	**-16.66% (7.34%)**	**20.94% (4.94%)**	**4.79% (3.33)**
3	Q2 System: 3 HPGe Array	Efficiency Calibration	**-8.65% (1.11%)**	**12.05% (0.76%)**	**17.77% (0.62%)**
4	Portable HPGe	Custom Calculation	**-26.06% (NA)**	**8.17% (NA)**	**9.69% (NA)**
5	Portable HPGe	Modeling Routine	**-1.46% (1.24%)**	**12.23% (1.95%)**	**-4.34% (0.92%)**
6	Portable HPGe	Modeling Routine	**15.85% (NA)**	**20.28% (NA)**	**59.60% (NA)**

Note: The value in parentheses is the calculated precision of the replicate measurements.

All individual results are also presented graphically in Figure 1. In general, the results for Co-60 are lower than the known value, and the results for Cs-137 and Eu-152 are higher than the known value (with one exception to each case). These trends are almost certainly related to the position of the sources in the drums. The Co-60 was placed at the top of the second tube – a position in which fewer than normal gamma rays will tend to reach the detector(s). Meanwhile, both the Cs-137 in the mixed matrix drum and the Eu-152 in the paper matrix drum were placed very close to the center of the outer tube. This is a position in which you would expect the detector(s) to encounter a greater than normal quantity of gamma rays. Although the mixed matrix drum has a higher effective density than the paper matrix drum (0.307 g/cm^3 vs. 0.105 g/cm^3), the overall errors were not statistically higher for the results on that drum compared to the lower-density paper drum.

Figure 2: Results Compared to Known Values



DISCUSSION

At this point we are not prepared to comment about the relative accuracy of traditional calibrated systems compared to modeling based analytical techniques. There are too few data points to make a conclusive statement of any kind. Thus far, the most accurate results were obtained with a commercial modeling technique (System #5), but the one calibrated system (System #3) had the next best overall accuracy. This is somewhat interesting given that System #3 is designed more for heightened sensitivity at the cost of accuracy. The three HPGe detectors in System #3 are placed just 8.9 cm (3.5 inches) from the rotating drum, yet the accuracy of those results are above average.

As far as the quality of the results in general, they appear to be quite good. Accuracy requirements for 55-gal drums can vary from site-to-site and project-to-project, but usually any result that falls within ± 30% from the known value is considered reasonably good. Thus far, all but two of the 18 individual assay results fell within this ± 30% benchmark. However, neither the matrix correction effects nor the gamma ray energies were particularly challenging to deal with in this first round, so one would hope that most of the results are pretty accurate. On the other hand, these PDP drums have additional discrete layers of attenuators to deal with that normal 55-gallon drums do not: two aluminum tubes (one slotted) and a poly liner. This could add a small additional bias if not correctly accounted for.

The precision of repeated measurements is relatively low for most systems, as would be expected given the significant gamma ray signals produced by these nuclides. Although

System #2 performed a nearly identical set of measurements as System #5, it's precision is somewhat worse (3 to 7% vs. 1 to 2%). No plausible reason has been identified to explain this discrepancy.

It is also interesting to make note of the wide variety of assay techniques used by the different participants. Some made use of commercially available modeling software (e.g., SNAP, ISOCS, and ISO-Plus), while another used a traditionally calibrated measurement system (System #3) and yet another team performed a custom set of calculations to account for geometry and attenuation correction factors (System #4). Regardless, all the participants produced good-to-excellent results.

Lessons Learned

The most difficult part of conducting this study involved the shipment of the drums to a site, and then back to LANL. In part, this was due to the generally heightened security concern that exists throughout the post-September 11 DOE complex. Although we intentionally chose sources that could be shipped as limited quantity, we consistently had problems with sites' Shipping/Receiving (S/R) personnel who refused to handle the drums without knowing precisely what radionuclides and quantities were present. Furthermore, the sites were also hesitant to ship the drums back to LANL unless they had detailed information about the contents. Attempts to persuade S/R personnel that we were shipping only "DOT limited quantity" of radioactive material were met with persistent rebuttals, even though local RCTs could barely detect any radiation outside the drums. Therefore, we usually had to work separately with the S/R personnel so the drums would be accepted on site. Since we were conducting a 'blind' intercomparison study, the S/R personnel had to agree to keep this information confidential. As far as we know, all assay teams were kept 'blind' during the study.

Because of these difficulties, the time necessary for shipment of drums from one site to another was much longer that originally estimated. In addition, around September 11, 2002, new shipping procedures were issued for use during times of heightened security (such as code orange). This practically stopped all shipment for a brief time until the security condition at DOE sites was changed to a reduced level. As a result, the LLW Intercomparison Project is not yet complete at the time of writing this paper (mid-January, 2003). Three additional NDA teams are still waiting for the drums to be assayed.

We also noticed that some NDA teams forgot to supply important information in their reports: the date of assay, experimental set-up, type of detector system used, type of software used in data analysis, and precision results. We propose that for future studies a template of the Summary Table be sent with the Ground Rules to facilitate a uniform set of results from all participants.

Future Plans

We believe this study was worthwhile and accomplished its' stated purpose. Certainly the participants were enthusiastic about the exercise, and most voiced strong support for a continued program. It is generally believed that the benefits of a 'one-time-only' intercomparison program are limited. We would like to make this effort an annual event and would like to identify an entity that is willing to sponsor it and cover the costs. Although there are no specific regulatory drivers that require such an effort, we believe it can be readily justified as a best management practice. The feedback provided to participants should be for their use only, and poor results should never cause an NDA team to be chastised, blackballed, or eliminated.

There are also a number of technical challenges that would be interesting to incorporate in future studies. Some that could be considered are as follows:

1. Drums with more dense matrices and/or highly non-homogenous matrices.
2. Drums that have an unknown fill height (less than 100%).
3. Radionuclides with gamma lines that interfere with one another or with background gammas.
4. Radionuclides with low energy gamma-rays and intrinsically higher attenuation uncertainties.
5. Using a large number of radionuclides, some of which are not very commonly observed.
6. Lower activity sources that challenge the detection limits of common systems.
7. Special nuclear material that exhibits self-absorption characteristics (e.g., U or Pu lumps).

REFERENCES

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