

## **Considerations and Lessons Learned from the Use of Gamma Spectroscopy Systems at FUSRAP Project Sites – 17459**

Brian Tucker\*; David Hays\*\*; Michael Winters\*; Doug Black\*

\*Cabrera Services, Inc.

\*\*USACE-Kansas City district

### **ABSTRACT**

This paper will discuss the application of gamma spectroscopy systems for analysis of environmental media samples for parents and daughter progeny of the Natural Uranium and Thorium Decay Series radionuclides. The concentrations and relative ratios of naturally occurring isotopes are used to confirm the presence of FUSRAP-related wastes and for understanding the associated potential dose and health risks. Focus will be given to lessons learned from the set-up and operation of on-site gamma spectroscopy laboratories used to provide stakeholders with defensible and timely results to support FUSRAP site investigations and remediation including adaptive survey design implementation, physical remediation and waste transport, and confirmatory final status survey activities.

This paper focuses on the potential impacts to gamma spectroscopy measurement accuracy from the following:

- Bias caused by sample matrix effects as well as use of different library reference values between performance evaluation study providers and testing laboratories;
- Use of calibration sources with limited energy ranges, and;
- Selection of available alternative isotopic decay reference data for the input of line energies and their intensities.

These lessons learned include discussing how inter-laboratory measurement differences from blind performance evaluation samples are affected by the manufacturer's measurement method, including equilibrium assumptions; the isotope decay information source library used by the study provider and participating laboratories; and calibration source geometry/density variances from actual sample conditions. Any of these factors can significantly impact the accuracy of reported results data. Deepening the reader's understanding of the various factors that impact the accuracy, interpretation, and overall usability of gamma spectroscopy results data from laboratories will help ensure defensible decision-making and cost-effective mission implementation within FUSRAP and similar remediation programs.

### **INTRODUCTION**

The use of high-resolution gamma spectroscopy (GS) in field laboratory configurations are common-place at sites undergoing investigation and remediation within the U.S. Army Corps of Engineers (USACE) Formerly Utilized Sites

Remediation Program (FUSRAP). When isotope(s) of interest are readily measurable or, at least, scalable from some other readily measurable radionuclide, and when proper consideration is given to the intended use of analysis results by stakeholders based upon established data/measurement quality objectives (DQO/MQO), GS can be a sensitive, accurate, precise, and responsive tool to support implementation of a cost-effective FUSRAP site remediation program. However, consideration must be given to the intended use of analysis results by stakeholders and the associated DQO/MQO.

For a result produced in a GS laboratory to be usable, it must first and foremost be accurate. Nuclear Regulatory Commission (NRC) publication (NUREG) 1576, also known as the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP) defines "accuracy" as "the closeness of a measured result to the true value of the quantity being measured" [1]. For the typical FUSRAP site sample analyzed in an on-site laboratory, the true value or concentration in each sample is essentially unknown but, at the same time, critical to remedial decision-making. For a GS laboratory operating under an DOD ELAP accredited program, accuracy should be expected to be a critical MQO assessed on a batch-basis by measuring the known activity concentration of a similar matrix laboratory control sample (LCS) provided by a standard supplier. Project lessons learned identified a key technical challenge that required assessment to better understand and reduce GS measurement results bias, and therefore produce the most accurate results practicable in a field laboratory environment where active site working conditions (e.g., greater potential for temperature fluctuations, vibrations due to construction activities and higher dust levels) can impact GS system performance. Our findings identified this challenge to be the importance of customization of gamma spectroscopy energy libraries, including use of the most up-to-date energy line and energy line intensity library values.

## **DISCUSSION**

### **Resolving Sample Matrix Effects to Reduce Bias**

The soil matrix and the associated source-to-detector geometry and self-attenuation characteristics can have a dramatic impact on the measurement of low-energy photons common to FUSRAP-related sites. More recent technical solutions include use of point source multi-energy calibrations, characterized detectors, and modeled sample geometries to account for matrix and sample size variability. For laboratories where modeled approaches to variations in source geometry and sample density are not well understood and/or readily acceptable to project stakeholders, it becomes more critical to normalize sample media to the extent practicable with both the calibration standards used to establish energy and efficiency curves and with LCS used to verify accuracy in individual analysis batches. For field soil samples, normalization is accomplished by careful drying to remove moisture followed by: passing through a sieve to remove rocks and debris; grinding in a mill to homogenize the sample and eliminate "hot spots"; and finally,

sealing the sample in a suitable analysis container to prevent off-gassing and allow more accurate assessments of radon/thoron daughter progeny.

If the calibration standard is consistent in density and configuration with a dried and prepared soil sample, matrix effects from interactions with photons of varying energies should be accounted for without contributing appreciable bias. When a sample's density and homogeneity are consistent with the calibration standard, accuracy is significantly improved. Note however that if there is discrepancy between the true sample density and the density in the calibration standard, there will remain some limited inherent bias affecting sample accuracy.

### **GS Library Reference Values**

GS systems rely upon a library of reference values when the software evaluates a spectrum of count data and determines the identity and activity concentration of any gamma-emitting nuclides within the calibration's operating energy range. These reference values include the isotope name, radioactive half-life, gamma-emission energies, intensities (i.e., percent yield per decay), and each value's associated uncertainty. The set-up of the laboratory is best addressed via close coordination between the Laboratory Director and a project's Health Physics staff (e.g., Project Certified Health Physicist). Once established, the HP/Lab Team should review the reference value source(s) periodically to ensure their library contains the most accurate, precise, and current reference values available. When reference values are not periodically updated, discrepancies between calibration laboratories, analysis laboratories, and performance testing sample providers can manifest. The authors currently recommend use of the latest isotope decay informational database offered and routinely updated by the National Nuclear Data Center ([http://www.nndc.bnl.gov/nudat2/indx\\_dec.jsp](http://www.nndc.bnl.gov/nudat2/indx_dec.jsp)) at the Brookhaven National Laboratory (BNL) [2] to ensure currency and continuity between providers.

### **Calibration Source Considerations**

GS calibration sources typically contain many isotopes with emission energies spanning the spectrum from approximately 40 keV to several MeV. Quantification of sample results will usually only occur from the assessment of emissions from within the calibrated energy range. Therefore, the ability to select from a wider library of emission energies during evaluation of samples is a key benefit from selecting higher and lower energy ranges in calibration sources. For typical FUSRAP applications, the desired range would be from around 60 keV to 2024 keV.

Americium-241 (Am-241) has a key energy at 59.54 keV and is commonly used to better define intrinsic efficiency at energies below 75 keV. In one unique situation, acquiring Am-241 and possessing it onsite was constrained. Cadmium-109 (Cd-109) became the new low-end isotope in the calibration source with an emission at 88.04 keV. This limitation potentially reduced Th-234 results accuracy by excluding the 63.29 keV photon from the isotope's library and essentially limiting Th-234 to the 92.5 keV doublet peak. Reliance on fewer energy lines when determining the weighted mean concentration for an isotope increases the likelihood of having bias

in one direction, as opposed to using many lines that collectively have positive and negative biases that tend to cancel out. Optimally, Am-241 would be used to represent the low-end of any calibration source used for the typical FUSRAP site application.

### **Key Isotope Quantification Differences between Performance Evaluation (PE) Study Providers, LCS Providers, and Operating Field GS Laboratories**

As noted in earlier discussions, inconsistency with application of library reference values and limitations in the energy spectrum used for quantification may yield vastly different final reported activity concentrations. For background, the activity concentration of a given radionuclide in a typical soil sample, in becquerels per kilogram (Bq/kg), may be expressed by the following simplified equation:

$$A_i = C / (E * T * Y * W) \text{ (Eq. 1)}$$

Where  $A_i$  = activity concentration of isotope  $i$  in Bq/kg

$C$  = net integrated counts in the specified energy region-of-interest

$E$  = intrinsic efficiency at a given energy (photons measured/emitted)

$T$  = sample count time in seconds

$Y$  = Photon Intensity (emissions at a given energy per isotope decay)

$W$  = weight of the sample in kilograms

In the simplest case where one energy line is used, the activity may be calculated using Equation 1 and the yield of that line. If more than one energy line is used, a weighted mean is calculated using each identified emission energy from a specific isotope's library. More weighting is given to those emission energies with the highest yields as their individual line activity values will have more counts per channel and, therefore, be more precise.

#### *Case Study 1. Activity Concentration Differences for Actinium-228 (Ac-228) between the PE Sample Provider and Participating Laboratories for the MRAD-016 (2012) Sample*

The author notes the following differences among the reported values:

FUSRAP Laboratory (Lab): 41.1 Bq/kg

Participating Laboratories Mean: 46.8 Bq/kg

Activity Values Assigned by the PE Sample Provider: 58.1 Bq/kg (Mean); 44.4 – 73.4 Bq/kg (Warning); 37.4 – 80.7 Bq/kg (Control)

The Lab reported value falls within PE acceptance ranges but appears biased low with a relative percent difference (RPD) of 34% and 13.0%, respectively, when compared to the PE provider's value and the participating laboratories mean value. At the time, the PE provider relied primarily on the most abundant Ac-228 gamma emission (911.2 keV with a 29% intensity). In contrast, the Lab utilized the calculated weighted mean from 17 unique photon emissions associated with the decay of Ac-228 to quantitate a result for the PE sample. As previously discussed,

over reliance on a single emission energy enhances the impact of bias in results from the GS efficiency calibration.

The original Lab library for Ac-228 includes the 911.2 keV emission with a 29% intensity. After review of more current analysis library source references, the intensity was updated to the NNDC value of 25.8%. The relative difference between the old yield of 29% and new yield of 25.8% is 11.0%. Thus, if one were only using this line to quantitate Ac-228, the activity would increase by 11%. Since the Lab used 17 lines the weighted effect from this change would have been approximately  $0.3 \times 11\%$ , or an increase of 3.3%. An increase of 3.3% in the Lab value would have reduced the RPD between its value and participant mean value to less than 10%. The lesson learned is that differences in analysis library parameter values may be driving bias between individual laboratories, as well as between an individual laboratory and the PE sample provider.

The author spoke to the PE sample provider about inconsistency between their assigned values and participant lab mean values. A cause was not identified but, the PE sample provider now relies less on internally measured values and more on the overall dataset of reported results from the participant labs when assigning the final study assigned value and associated acceptance limits.

*Case Study 2. Activity Concentration Differences for Thorium-234 (Th-234) between the PE Sample Provider and Participating Laboratories for the MRAD-016 (2012) Sample*

The author notes the following differences among the reported values:

FUSRAP Laboratory (Lab): 68.3 Bq/kg

Participating Laboratories Mean: 79.9 Bq/kg

Activity Value Assigned by the PE Sample Provider: 74.0 Bq/kg (Mean); 42.7 – 120 Bq/kg (Warning); 23.4 – 139 Bq/kg (Control)

Th-234 has two key photon emissions with measurable intensities (greater than one percent) under normal field GS operating conditions: 63.29 keV (4.8%) and a doublet peak centered at 92.5 keV with an intensity of 5.58%. The calculated double library values are based on the 92.38 keV (2.81%) and the 92.80 keV (2.77%) emissions. At the time of the 2012 PE sample analysis, the Lab used these two lines and their associated intensities.

More recently, there have been several changes. First, referencing the earlier discussion of isotope limitations in the calibration source, the Lab limited the calibration range bottom limit to 88.04 keV, corresponding to Cd-109 and thus discontinued use of the 63.29 keV line for quantitating the activity of Th-234 in samples. Second, in accordance with BNL library updates, the Lab changed the 92.5 keV doublet yield value from 5.58% to 4.23%. Third, for those laboratories able to use the 63.29 keV peak, that peak has also seen changes in its yield value, as earlier libraries list a yield of 4.8% and the most current BNL value is 3.7%.

The relative percent changes in the intensities of the 92.5 keV doublet and the 63.29 keV are decreases of 32% and 30%, respectively which cause the same percentage increases in the activity concentrations calculated for each of the two lines. The Lab PE sample result is 14.5% lower than the participant study mean. It appears likely that a significant fraction of the difference between the study mean and a given lab result is due to participants using different yield values for the 63.29 keV and 92.5 keV lines.

## **CONCLUSIONS**

Gamma spectroscopy energy libraries must be customized for users' applications to optimize measurement accuracy. This paper shows that use of different reference libraries may contribute to differences in measurement values between field laboratories, PE sample providers and LCS sample providers, especially in the case of low activity samples containing isotopes with low yield energy lines. Such differences are caused by differences in energy line and percent yield values between different libraries.

The author recommends standardization of energy line and energy line yield values to improve measurement accuracy and inter-laboratory result comparability.

## **REFERENCES**

1. U.S. EPA, DoD, U.S. DOE, and U.S. NRC; Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP), NUREG-1576, July 2004
2. Brookhaven National Laboratory National Nuclear Data Center, Nuclear Structure and Decay Data.