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APPLICATION OF *IN-SITU* GAMMA SPECTROMETRY IN THE REMEDIATION OF RADIOACTIVELY CONTAMINATED SOIL

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ABSTRACT

The Fernald Environmental Management Project (FEMP) is a U.S. Department of Energy site that is undergoing total remediation and closure. Most of the remediation effort entails massive excavation of soil for disposal, both offsite and onsite, at an engineered disposal facility. *In-situ* gamma spectrometry is routinely used to support soil excavation operations to accurately and quickly identify soil areas as being above or below regulatory remediation criteria.

Two different *in-situ* gamma spectrometry systems are used. The first is a sodium iodide (NaI) detector mounted either on a tractor or a jogging stroller, depending on the terrain to be measured. The NaI system allows the collection of a gamma energy spectrum which can be analyzed to identify and quantify radioactive isotopes which are present within the detector's viewing area. Each energy spectrum is tagged by location coordinates provided by an on-board global positioning system (GPS) to precisely locate elevated contamination areas. The second is a tripod-mounted, high purity germanium detector (HPGe) gamma spectrometry system that is functionally similar to the NaI system. The principal advantage of the HPGe is its superior resolution, which allows much more accurate identification and quantification of radionuclide contaminants in soils.

In order to effectively utilize the data quality objective process with these systems, three quality assurance (QA) elements had to be performed. First, method validation studies demonstrated comparability with conventional radiochemistry methods and established performance-based acceptance criteria for key quality control parameters at various data quality levels. The method validation studies for the HPGe system stressed accuracy and comparability, while method validation studies for the NaI systems stressed quantifying measurement uncertainty and detection limits. Second, a "User's Manual" was developed that specifies measurement approaches, provides data interpretation guidelines, and discusses operational and environmental factors that could adversely affect *in-situ* gamma spectrometry measurements. This manual is primarily designed for environmental scientists responsible for remediating soils rather than for analytical chemists who perform the measurements. Third, an *in-situ* gamma spectrometry QA program was implemented to address programmatic QA elements, to ensure legal defensibility of the data, and to specify quality control (QC) criteria, their frequency of measurement, their acceptance limits and whether or not they are to be control charted.

INTRODUCTION

The FEMP is a U.S. Department of Energy site that is undergoing total remediation and closure. Most of the remediation effort entails massive excavation of soil for disposal, both offsite and onsite at an engineered disposal facility. *In-situ* gamma spectrometry is routinely used in support of soil excavation operations to accurately and quickly identify soil areas as being above or below regulatory remediation criteria. Two different *in-situ* gamma spectrometry systems are used. The first is a sodium iodide (NaI) detector system, while the second is a high-purity germanium (HPGe) detector system. The former system is mounted on either a tractor (RTRAK) or a jogging stroller (RSS), depending on the terrain, while the latter system is tripod-mounted.

Both RSS and RTRAK have a measurement system consisting of a 4x4x16 inch NaI detector and associated electronics to provide high-speed pulse height analysis. This system allows the collection of a gamma ray energy spectrum, which can be analyzed to identify and quantify radioactive isotopes that may be present within the detector's viewing area. The RTRAK and RSS are each equipped with a GPS operated in a real-time differential mode to provide location coordinates. Each energy spectrum is tagged with the location coordinates provided by the GPS. All energy and location data are stored on magnetic media by an on-board computer system. This information is used to accurately locate and subsequently map radiological data within the measurement area.

On the RTRAK, the detector is positioned on the tractor horizontal to the ground and perpendicular to the direction of travel at a height of approximately 31 cm above the ground. The detector on the RSS is mounted horizontal to the ground and parallel to the direction of travel at a height of approximately 31 cm. The normal operation of the RTRAK and RSS consists of moving the systems over the measurement area at a predetermined speed. Spectra are continuously collected at regular intervals, typically a few seconds. The viewing area size is a function of the tractor speed, the acquisition time, and the detector's geometrical configuration. For example, for the 4x4x16 inch detector at the 31 cm height, the viewing area is 8.8. m^2 for a single measurement when the system is moving at one mile per hour, with a 4-second data acquisition time (typical operating parameters).

The HPGe detectors are mounted on tripods at heights ranging from 15 cm to 1.0 m above the ground surface. The detectors are connected to 8192 channel multi-channel analyzers which allow the collection of a high resolution gamma ray spectrum. The superior resolution of HPGe detectors relative to NaI detectors allow it to accurately quantify a wide variety of isotopes with minimal interferences. Data acquisition times typically are 15 minutes. The HPGe field of view ranges from over 100 m^2 at a 1.0 m detector height to 3.1 m^2 at a 15 cm detector height.

METHOD VALIDATION STUDIES

The method validation study for HPGe entails determining the similarity between data generated by HPGe measurements and data generated by laboratory analysis of physical samples. It also delineates acceptance criteria for key QC elements and data quality elements. Three radiological contaminants of concern were measured by HPGe and laboratory methods: total uranium, thorium-232 and radium-226. Method validation studies for NaI systems stressed quantifying measurement uncertainty and detection limits. Such assessments were performed as a function of vehicle speed and data acquisition time in order to determine preferred operating parameters.

HPGe Comparability Studies

One part of the method validation study for HPGe entailed assessing the comparability between HPGe measurements and laboratory data. To accomplish this, a series of physical samples were collected from different areas of widely varying concentrations of contaminants. In each area, samples were collected in a "bullseye" pattern to mimic the averaging done by the field HPGe detector. That is, the area from which physical samples were taken can be envisioned as a circle, with the HPGe detector located above the center. The HPGe detector records gamma ray photons from every point within the circle; however, it records more gamma rays from soil closer to the detector than from soil further from the detector.

For comparison with HPGe measurements, a weighted average (weighted based upon gamma photon fluence contributions) of all laboratory data for a given area was calculated. Figures 1 and 2 show plots of HPGe measurements vs weighted average laboratory data for total uranium and thorium-232. High correlation coefficients (R^2 value), line slopes near one, and line intercepts close to 0.0 demonstrate comparability of data. The width of the error bars for laboratory data in Figures 1 and 2 primarily reflect the degree of heterogeneity among samples in a given area rather than laboratory precision.

NaI Method Validation

A major portion of the method validation studies for NaI systems addressed the total system measurement uncertainty for moving systems. Data were acquired experimentally via repeated measurement profiles, which involved moving the RTRAK or RSS back and forth along a given track for 20 iterations. Each track was divided into segments and the mean and standard deviation of the measurements in each segment was determined. Table 1 shows the results of the precision studies for one area with the RTRAK moving at a speed for 0.5 mph, with a 2-second data acquisition time. Such precision studies were carried out in different areas, using a combination of different speeds and data acquisition times in each area. The results of these studies demonstrated that:

1. The uranium-238 measurements display low degrees of precision. This limits the usability of the data for low-concentration measurements. The low degree of precision (high uncertainty) occurs because of the low photon yield at the energy of interest, the high spectrum background, and interferences from thorium-232 and radium-226 daughter gamma rays.

2. The thorium-232 measurements display the highest degree of precision of the three radionuclides of interest. The high degree of precision (small uncertainty) occurs because of a relatively high photon yield at the energy of interest, the low spectrum background, and because of only limited interference from a low intensity radium-226 peak.

3. The radium-226 measurements display a degree of precision similar to that of uranium or between that of the other two radionuclides of interest. This is in part because both the photon yield and the detection efficiency at the energy of interest fall between those of the thorium and uranium.

Knowledge of the overall precision from studies such as the one outlined above was a key factor in ascertaining *a priori* minimum detectable concentrations, determining error rates, and setting trigger levels.

USER'S MANUAL

Early in the remediation process at the FEMP, it became clear that a critical need existed to bridge the gap between primarily analytical information contained in method validation studies and programmatic remediation design documents. The User's Manual bridges that gap by providing user guidelines, data interpretation guidelines, and measurement strategies and approaches; by discussing operational and technical factors that could adversely affect data; and by delineating strengths and limitations of *in-situ* gamma spectrometry. While the document is beneficial to anyone involved with any aspect of *in-situ* gamma spectrometry, it is primarily aimed toward FEMP project personnel who:

- plan soil remediation projects;
- collect *in-situ* gamma spectrometry data for soil remediation projects;
- interpret *in-situ* gamma spectrometry data for soil remediation projects;
- integrate *in-situ* gamma spectrometry data with other data sets or into engineering designs; and

- make decisions based upon *in-situ* gamma spectrometry data.

The User's Manual has four sections: 1) Investigation Approaches; 2) Measurement Approaches; 3) Data Interpretation Guidelines; and 4) Technical Issues. Section 1 deals with broader-scale issues such as how *in-situ* gamma spectrometry is used in pre-design investigations and in soil excavation operations. Section 2 deals with smaller-scale issues such as how *in-situ* gamma spectrometry is used to detect, confirm, and identify hot spots. Section 3 addresses such issues as climatic/weather effects upon in-situ gamma measurements, topographic effects, total activity data interpretation, and mapping conventions. Section 4 addresses technical issues such as data review checklists, minimum detectable concentrations, positioning and surveying, and the effects of radon-222 on radium-226 measurements.

QUALITY CONTROL/QUALITY ASSURANCE

All in-situ gamma spectrometry operations, whether method validation studies or field measurements in support of remediation operations, are governed by a comprehensive QA/QC program. The QA program contains all of the same quality elements as a traditional environmental laboratory QA program. It has ten criteria: 1) QA program; 2) personnel training/qualification; 3) quality improvement; 4) documents and records; 5) work processes; 6) method design, 7) procurement/control of materials and services; 8) facilities and equipment/calibration and maintenance; 9) management assessment; and 10) external assessments and audits.

Of particular interest is the QC program, which is centered around performance-based measurements. In this regard, acceptance criteria of key quality control elements are specified, while the mechanism of how such measurements are obtained are not specified in either the QA plan or QC plans. Table 2 contains such criteria for two data quality levels called Analytical Support Levels (ASLs) at the FEMP. ASL B corresponds generally to the US EPA "screening data" category, while ASL D corresponds to the US EPA's "definitive data" category.

Information from the method validation studies, the User's Manual, and the QA/QC plans are incorporated into Project Specific Plans (PSPs) and project Data Quality Objectives (DQOs) to support specific remediation activities. In-situ gamma spectrometry data are validated to ensure that they satisfy the requirements and needs specified by the PSPs and DQOs.

SUMMARY

Routine utilization of *in-situ* gamma spectrometry in remediation at Fernald rests upon three programmatic elements. method validation studies carried out to delineate key measurement quality control elements such as comparability, representativeness, accuracy, uncertainty, and detection limits; a User's Manual which specifies to environmental engineers and scientists how *in-situ* gamma spectrometry should be used in remediation operations; and a comprehensive QA program to ensure that *in-situ* gamma spectrometry data are of sufficient quality for their intended usage and are legally defensible.

TABLE 1
RTRAK PRECISION STUDIES AT 0.5 MPH WITH A 2.0 SECOND DATA ACQUISITION TIME

		Uranium-238 (pCi/g)			Thorium-232 (pCi/g)			Radium-226 (pCi/g)		
Segment	No. Measurements	Mean	Std Dev	%Std Dev	Mean	Std Dev	%Std Dev	Mean	Std Dev	%Std Dev
01	129	12.4	9.3	75	0.75	0.26	35	0.72	0.50	70
02	217	14.1	9.1	65	0.77	0.32	42	0.79	0.51	64
03	206	15.6	9.0	58	0.75	0.27	36	0.82	0.47	57
004	205	15.2	8.3	55	0.80	0.31	39	0.76	0.53	70
05	216	16.8	8.7	52	0.73	0.29	40	0.82	0.54	66
06	225	14.5	9.4	65	0.76	0.29	38	0.76	0.52	68
07	200	16.5	9.6	58	0.78	0.31	40	0.80	0.54	68
ROAD	120	12.2	7.3	60	0.48	0.29	60	0.59	0.45	76
08	231	17.0	9.2	54	0.75	0.34	45	0.82	0.59	72
09	232	18.0	9.3	51	0.75	0.32	43	0.87	0.51	58
10	240	17.2	9.8	57	0.73	0.31	42	0.77	0.48	63
11	193	15.2	8.6	56	0.75	0.28	37	0.76	0.50	65
Averages		15.7	9.1	59	0.76	0.30	40	0.79	0.52	66
Minimum		12.4	8.3	51	0.48	0.26	35	0.72	0.45	57
Maximum		18.0	9.8	75	0.80	0.34	60	0.87	0.59	76

TABLE 2
TABULATION OF QUALITY CONTROL CRITERIA AND REQUIREMENTS

RTRAK and RSS NaI Detector QC Criteria and Requirements

QC Element	Nuclide	Gamma Energy	QC Criteria	Frequency	Control Chart
Energy Calibration	Tl-208 Pb-212	2614.5 keV 238.6 keV	Channel 447±2 Channel 40±2	Days used, prior to and following use	No
Detector Counting Efficiency Check	Tl-208	2614.5 keV	Predetermined check source value (decay corrected) $\bar{x} \pm 3$ sigma	Days used, prior to and following use	Yes

HPGe Detector QC Criteria and Requirements

QC Element	Nuclide	Gamma Energy	QC Criteria	Frequency	Control Chart
Energy Calibration	Am-241 Cs-137 Co-60	59.5 keV 661.6 keV 1332.5 keV	Channel 158±1 Channel 1763±2 Channel 3553±3	Days used, prior to and following use	No
Detector Resolution	Co-60	1332.5	Measured mean value $\bar{x} \pm 3$ sigma	Days used, prior to and following use	Yes
Detector Counting Efficiency Check	Co-60	1332.5	pre-determined check source value (decay corrected) $\bar{x} \pm 3$ sigma	Days used, prior to and following use	Yes

HPGe Field Measurements QC Criteria and Requirements

QC Element	Gamma Energy Nuclide or Basis	QC Acceptance Criteria	Frequency	Control Chart
Field Measurement Interference	1460.8 keV	keV = 1460.8 FWHM ≤ 3.0 keV or Channel = 3895.0 FWHM ≤ 8 Channels	Each time measurements are made	No
Field Control Station	 Total U Th-232 Ra-226 K-40	ASL -D measured value ±3 sigma measured value ±3 sigma measured value±3 sigma measured value±3 sigma	On each day measurements are made	Yes
Field Control Station	Temperature Humidity Soil Moisture	No Criteria	Each day measurements are made	No

QC Element	Gamma Energy Nuclide or Basis	QC Acceptance Criteria	Frequency	Control Chart
Minimum Detectable Concentration	Free Release Levels for Nuclides of Concern	For ASL-D 95% UCL[1] <FRLs For ASL-B 90% UCL[1] <FRLs	Quarterly	No
Measurement Accuracy	Compared to weighted average of physical samples	ASL-D - weighted average of physical sample ±20% ASL-B - weighted average of physical sample ±35%	Annually	No
Measurement Bias	Compared to weighted average of physical samples	Bias acceptable unless it produces errors resulting in accuracy being exceeded	Annually	No
Precision of Duplicates	At least one per every 20 HPGe measurements.	measured value >(5 x MDC) then RPD ≤±20% measured value <(5 x MDC) then measurement difference ≤ ±MDC	At least one per every 20 HPGe measurements.	No
Detector Counting Efficiency Determination	Determination of conversion (efficiency) factors.	initial conversion factor ±10% for each gamma energy[2]	Annually	No

Note 1. Upper confidence level (UCL) for MDC.

Note 2. Nuclide and Gamma energies measured:

Cs-137	32.2
Eu-152	39.5
Am-241	59.5
Eu-152	121.8
Eu-152	244.7
Eu-152	344.3
Eu-152	411.1
Eu-152	444.0
Cs-137	661.6
Eu-152	778.9
Eu-152	964.0
Co-60	1173.7
Co-60	1332.5
Eu-152	1408.0

Figure 1
Correlation Between HPGe and Laboratory
Data for Total Uranium at a 31 cm Detector Height

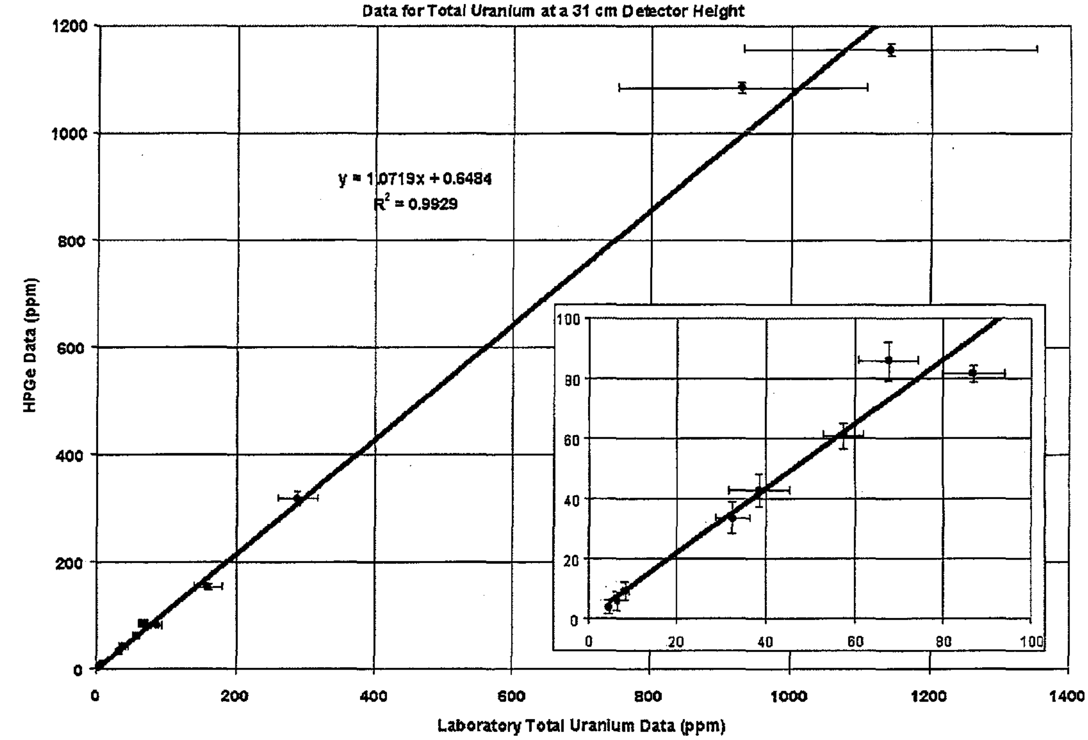

Figure 2
Correlation Between HPGe and Laboratory Data for Th-232
at a 31 cm Detector Height