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Evaluation of Measurement Reproducibility Using the Standard-Sites Data, 1994 Fernald Field Characterization Demonstration Project

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**Evaluation of Measurement Reproducibility
Using the Standard-Sites Data,
1994 Fernald Field Characterization Demonstration Project**

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Abstract

The U.S. Department of Energy conducted the 1994 Fernald (Ohio) field characterization demonstration project to evaluate the performance of a group of both industry-standard and proposed alternative technologies in describing the nature and extent of uranium contamination in surficial soils. Detector stability and measurement reproducibility under actual operating conditions encountered in the field is critical to establishing the credibility of the proposed alternative characterization methods. Comparability of measured uranium activities to those reported by conventional, U.S. Environmental Protection Agency (EPA)-certified laboratory methods is also required.

The eleven (11) technologies demonstrated included (1) EPA-standard soil sampling and laboratory mass-spectroscopy analyses, and currently-accepted field-screening techniques using (2) sodium-iodide scintillometers, (3) FIDLER low-energy scintillometers, and (4) a field-portable x-ray fluorescence spectrometer. Proposed advanced characterization techniques included (5) alpha-track detectors, (6) a high-energy beta scintillometer, (7) electret ionization chambers, (8) and (9) a high-resolution gamma-ray spectrometer in two different configurations, (10) a field-adapted laser ablation-inductively coupled plasma-atomic emission spectroscopy (ICP-AES) technique, and (11) a long-range alpha detector. Measurement reproducibility and the accuracy of each method were tested by acquiring numerous replicate measurements of total uranium activity at each of two "standard sites" located within the main field demonstration area. Meteorological variables including temperature, relative humidity, and 24-hour rainfall quantities were also recorded in conjunction with the standard-sites measurements.

Evaluation of the replicate uranium-activity data indicates that the alternative characterization techniques based on detection of alpha-particle radiation are generally unreliable and non-robust under actual field operating conditions; these measurements appear sensitive to changes in soil-moisture content. The alpha-track detectors and the electret ionization chambers, both originally developed as passive radon monitors for indoor use, performed poorly in the field compared to the long-range alpha detector, which was intended as a field instrument. Measurements by the ICP-AES technology were highly variable, and in some instances, notably inaccurate. Poor exper-

imental design of ICP-AES demonstration limits direct comparison of these results to those from the rest of the field demonstration program.

The industry-standard field screening devices, the sodium-iodide scintillometer, the FIDLER detector, and the field-portable x-ray fluorescence device, all provided relatively stable and reproducible measurements of total uranium activity. However, these measurements were somewhat inaccurate: all three field-screening tools reported uranium activities higher than the comparable laboratory analyses. The direction of this error is conservative from an environmental-remediation/regulatory standpoint. However, this conservatism probably would result in increased remediation costs

The high-energy beta scintillometer and the advanced gamma-spectrometry techniques collectively produced the most reproducible and consistently accurate measurements of total uranium activity. The beta scintillometer may be slightly sensitive to changes in soil-moisture content; however all three measurement systems appeared quite robust with respect to the available environmental variables. The measurements obtained by the high-mounted and low-mounted gamma spectrometer configurations vary slightly, but consistently, as would be predicted considering the differences in scale of their respective measurement volumes.

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Evaluation of Measurement Reproducibility Using the Standard-Sites Data, 1994 Fernald Field Characterization Demonstration Program

Introduction

The U.S. Department of Energy (DOE) is investigating advanced methods for characterizing, and ultimately for remediating, the uranium-contaminated soils that occur at a number of DOE facilities throughout the United States as a result of past defense-related operations. The perception is that currently available, commercial characterization technologies are either too costly or insufficiently sensitive for routine use in environmental restoration of these contaminated sites. Consequently, the DOE Office of Technology Development funded development of a number of advanced, alternative characterization technologies for use in field screening activities to identify and describe the extent and nature of uranium contamination.

During the summer of 1994, the DOE conducted a field characterization demonstration program involving six advanced characterization technologies at the Fernald Environmental Management Corporation (FERMCO) site near Fernald, Ohio. The Fernald site is located approximately 20 miles (30 kilometers) northwest of Cincinnati (fig. 1). These advanced technologies are (1) high-resolution gamma-ray spectrometry (two configurations), (2) beta-scintillation counting, (3) field-based laser-ablation inductively coupled-plasma atomic-emission spectrometry, (4) long-range alpha detection, and two variants of passive alpha monitoring: (5) alpha-track detectors and (6) electret ionization chambers. To provide comparable cost and performance information (Douthat and others, 1995), three commercial, industry-standard screening technologies were also demonstrated under the identical field and operating conditions. These

standard characterization technologies are (1) a sodium-iodide gamma-ray scintillometer, (2) a low-energy gamma-ray scintillometer (FIDLER detector), and (3) a field X-ray fluorescence detector. Conventional soil sampling coupled with U.S. Environmental Protection Agency (EPA)-approved laboratory analysis of contained uranium was also conducted as the baseline reference against which to compare the performance of the proposed, advanced-alternative and industry-standard measurement technologies. A complete description of the 1994 Fernald field characterization demonstration program, including relevant planning and operating-instructional documents, functional descriptions of the different measurement devices provided by the technology developers, raw field measurements, calibration procedures and results, and baseline calibrated uranium activities measured by all participating technologies, has been compiled by Rautman and others (1995).

Detector stability is important in an evaluation of proposed alternative characterization technologies, such as were tested during the 1994 field characterization demonstration program at the Fernald site. A credible alternative characterization technology should be able to reproduce measurements of the same physical phenomenon at the same location, and should be able to do so under realistic field operating conditions. This report presents an evaluation of replicate measurements obtained by each of the several industry-standard and proposed alternative field characterization technologies at two "standard sites" located within the field demonstration area. The results of the different detectors have been compared against themselves, against each other, and against the results of EPA-accepted,

Figure 1. Location map showing the U.S. Department of Energy Fernald site in southwestern Ohio and the location of demonstration site for the 1994 Fernald field characterization demonstration program.

conventional chemical analyses of soil samples.

Methodology

Conceptual and Operational Approach

The 1994 Fernald field characterization demonstration program was designed to include repeated measurements by each alternative technology at each of two standard sites to provide a basis for evaluating detector stability and measurement reproducibility. The field demonstration was conducted immediately north of a part of the Fernald plant known as the incinerator area (fig. 1), and these standard sites were located in the field within the actual demonstration measurement grid. The local geology of the soils and the character of the uranium contamination prevailing at the

standard sites is thus essentially identical to that measured as part of the comparative demonstration program (Tidwell, 1994). Similarly, the weather and other environmental conditions present during measurements of the standard sites were virtually identical to those involved in the field survey because the standard sites were remeasured at various times during the survey day. Temperature and humidity readings were obtained at the time of collection of the standard-site measurements; measurements of total precipitation were also obtained for each day of field operations. All standard-sites measurements and supporting information is tabulated in Rautman and others (1995; appendices D and F).

Because of the different logistical arrangements and requirements associated with each alternative characterization technol-

ogy (total time on site, time to make one measurement, etc.), the number of measurements obtained at each of the standard sites varied from 2 to 21. This variability in the underlying measurement set makes strict statistical comparison of the data somewhat difficult. However, there are marked differences among the data for the various alternative technologies that indicate some detectors or techniques are inherently more stable than others.

Two other variances are noteworthy with respect to logistics of the repeated measurements at the standard sites. First, developers of one technology (abbreviated ICP-AES; described in the next section) did not take the requested replicate measurements at the actual standard sites in the field. The purported explanation for this lack of adherence to instructions contained in the project plan was that the ICP-AES method required removing surficial material from the marked standard sites, and thus offered the potential for changing the uranium activity remaining to be measured by subsequent technology demonstrators. The developers of the ICP-AES technology instead recorded replicate measurements of a small quantity of soil removed from three of the manufactured, spiked soil plots (Rautman and others, 1995) developed for calibrating the different measurement devices to a uniform set of known uranium activities. The replicate measurements thus obtained do provide a reasonable basis for evaluating detector repeatability. However, the modified approach defeated the original purpose of the standard-sites measurements, which was to evaluate performance under actual field operating conditions and to allow a comparison of that performance against other measurement techniques.

Second, developers of three other alternative characterization technologies (Beta, GMH, GML; also described in the next section) developed a modified approach for trans-

forming raw measurement values to total uranium activities based upon observed changes in the standard-sites measurements with time. The rationale underlying this modified approach was that the standard sites measurements "should" be effectively constant, all other things being equal. Assuming that the uranium activity in-place does not vary with time, observed changes in the actual measurement values must be reflecting real changes in the physics being captured by the repeated instrument readings (i.e., that all other things were *not* equal). Because the standard-sites measurements for these technologies were observed to decrease and then resume previous magnitudes in temporal association with precipitation events in the field, the developers derived an empirical "soil-moisture correction factor," representing the sum of all environmental effects associated with rainfall, wetting, and subsequent drying of the soil in the field. The Fernald field characterization demonstration data report (Rautman and others, 1995) provides additional detail and a conceptual example of the development of these soil-moisture correction factors.

Because the principal purpose of this evaluation of the standard-sites measurements was to quantify detector reproducibility and to identify causes contributing to such reproducibility or lack thereof, this report uses the "baseline" calibrated data for these three technologies without the application of the soil-moisture correction factors. Both the baseline and the "adjusted" total uranium activities obtained by the Beta, GMH, and GML technologies are included in the data report, appendix G (Rautman and others, 1995).

Brief Description of Alternative Characterization Technologies

The different characterization technologies demonstrated at Fernald during the summer of 1994 can be classified into two types:

industry-standard field screening methods (including conventional, EPA-standard laboratory analysis), and advanced field-measurement technologies (table 1). Additional classifications are possible based upon the physics of the measurement process. Each of these techniques for measuring uranium contamination in soil is described briefly for completeness. For convenience, the different technologies are described in alphabetical

order, according to the abbreviation indicated in table 1. Additional description of the characterization technologies has been presented by Rautman and others (1995), and more detailed descriptions and operational procedures have been provided by the technology developers in the original appendices of the "Field Demonstration Project Plan" (included as appendix A of Rautman and others, 1995).

Table 1: Classification and brief description of alternative characterization technologies demonstrated at the Fernald site

[Sample volumes (in m³) are computed in Appendix A of this report]

Abbreviation	Technology Name	Detection Principle		Sample volume
Industry-Standard Field-Screening Techniques				
FID	FIDLER scintillometer	passive	gamma rays	1.3x10 ⁻³
Lab	Mass spectroscopy	active	ionized elements	2x10 ⁻⁴
NAD	Sodium-iodide scintillometer	passive	gamma rays	2x10 ⁻⁴
XRF	X-ray fluorescence detector	active	photons	5x10 ⁻⁷
Advanced Field-Measurement Technologies				
ATD	Alpha-track detector	passive	alpha particles	2x10 ⁻⁸
Beta	Beta scintillometer	passive	beta particles	2x10 ⁻³
EIC	Electret ionization chamber	passive	alpha particles	9.4x10 ⁻⁸
GMH	High-mount gamma spectrometry	passive	gamma rays	1.6x10 ¹
GML	Low-mount gamma spectrometry	passive	gamma rays	1x10 ⁰
ICP-AES†	Laser-ablation inductively coupled plasma-atomic emission spectrometry	active	visible and ultraviolet light	6.5x10 ⁻⁸
LRAD	Long-range alpha detection	passive	alpha particles	2.5x10 ⁻³

[†]The ICP-AES technology did not make replicate measurements at the two standard sites. A similar style of replicate measurements was obtained using the beds constructed for instrument calibration at the Fernald site. However, direct comparison of results from the ICP-AES technology with those from the other alternative characterization technologies is not possible.

Alpha-Track Detector (ATD)

Alpha-track detectors (ATDs) are passive, single-use devices that were originally developed for the measurement of indoor airborne alpha activity (Alter and Fleisher, 1981), principally that resulting from environmental radon gas. Detection of alpha particles is accomplished by observation of damage tracks left in a proprietary polymer material (Lan-

track[®]). Damage tracks within the plastic detector material are etched chemically after exposure for greater visibility and then counted. The density of tracks (per unit area) is proportional to the monitored alpha activity (Dudney and others, 1994).

Beta Scintillometer (Beta)

The high-energy beta scintillometer (Beta) is an advanced characterization technol-

ogy developed at Pacific Northwest Laboratories (Schilk and Perkins, 1993). The device uses multiple, optically independent layers of a special, scintillating plastic material that is sensitive to the 2.29 MeV (million-electron-volt) beta particles emitted by ^{234m}Pa . The multiple scintillating layers are intended to allow discrimination of these high-energy beta particles from lower-energy background radiation through coincident-counting techniques. Scintillation events detected simultaneously in all three layers of the detector by separate photomultiplier tubes are inferred to represent the desired 2.29 MeV beta particles. Scintillation events recorded in only one or two of the detecting layers are discounted as caused by beta particles with energies too low to represent reliably the inferred decay of uranium daughter products.

Electret Ionization Chamber (EIC)

Electret ionization chambers (EICs) are small, passive electrical devices originally developed for the measurement of indoor alpha activity, again principally from environmental radon gas (Kotrappa and others, 1981, 1988). The device consists of a positively charged piece of Teflon[®]: an "electret." The EIC is activated by screwing the electret into a special, conductive polymer holder, which establishes a static electrical field. Ionizing radiation, including alpha particles, passing through the sensitive volume of the detector creates electron showers that are attracted to the positively charged electret, thus neutralizing that charge in proportion to the radiation load. The charge remaining in the EIC detector after exposure is measured and the measured voltage drop converted to activity.

Because the electret ionization chambers are sensitive to all ionizing radiation, a pair of detectors is utilized in mixed radiation fields. One detector measures the effect of all ambient radiation while the other is shielded

by Tyvek[®] plastic, which absorbs low-energy alpha particles before they reach the detector. Alpha activity is determined as the difference between the two readings (Dudney and others, 1994).

FIDLER Detector (FID)

FIDLER is an acronym for "**F**ield **I**nstrument for **D**etection of **L**ow-**E**nergy **R**adiation." The FIDLER scintillometer (FID) uses a thallium-activated sodium-iodide crystal that converts passing gamma-ray photons to visible light, which is then detected by a photomultiplier tube and associated electronics. The principal difference between the FIDLER scintillometer and the more conventional sodium-iodide scintillometer is that the sodium-iodide crystal primary detector in the FIDLER is only 0.063 inches (1.5 mm) thick (Fermco, 1994). This relatively small detector volume reduces the likelihood that high-energy photons, such as cosmic radiation, will interact with the scintillating material and be detected. The device is principally sensitive to gamma rays in the energy range 10 to 200 keV (thousand electron volts), which includes the primary gamma spectrum of the uranium decay chain.

Gamma-ray Spectrometer (High-Mounted [GMH] and Low-Mounted [GML] Configurations)

The in-situ gamma spectrometry system is based on a standard germanium-crystal gamma-ray sensor, specifically selected for sensitivity to low-energy photons emitted by ^{234}Th (63 keV and 93 keV) as part of the uranium decay chain. The primary indicator of uranium contamination, however, is the 1.0 MeV photon from ^{234m}Pa . Electronics associated with the germanium crystal detect electrical pulses generated within the crystal from excitation of germanium atoms by passing gamma-ray photons. The current associated with these pulses is proportional to the energy

of the triggering radiation, allowing resolution of the spectrum of energies associated with incoming radiation (Schilk and Perkins, 1994).

The gamma spectrometer system was demonstrated at Fernald in two different configurations. In the high-mount configuration (GMH), the detector crystal is mounted approximately one meter above the ground surface on a tripod. The unshielded device detects all incoming radiation from all directions. The effective measurement area of the GMH detector is approximately 300 m², or a circle with a radius of approximately 10 m. Because gamma radiation is penetrating, contaminants can be detected at soil depths of up to 15 or 20 cm immediately beneath the spectrometer. The depth of detection decreases radially outward with increasing distance from the center of the detector.

In the low-mount configuration (GML), the detector crystal is mounted about 30 cm above ground level on a tripod. In this case, however, the field of view of the detector is collimated by the addition of tungsten or lead shields. This collimation effectively reduces the size of the region examined at the Fernald site to approximately 10 m², or a circle with a radius of 3 m (Schilk and Perkins, 1994).

Laser Ablation-Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES)

The laser ablation-inductively coupled plasma-atomic emission spectroscopy characterization technology is a unique, field adaptation of the more classical, inductively coupled plasma (ICP) atomic emission spectroscopy (AES) laboratory method of chemical analysis (Anderson, 1994). Field soils are sampled directly by focusing an intense beam of laser radiation on the soil surface via a fiber-optic cable. This laser beam ablates soil particles, which are then entrained by a stream of argon

gas and transported through a umbilical tube to an ICP burner located in a mobile laboratory trailer where the minute soil particles are vaporized. Light from this plasma source is transmitted to a standard atomic emission spectroscope and wavelengths characteristic of various elements can be detected and quantified. Up to 20 elements currently can be identified simultaneously at the parts-per-million level.

Long-Range Alpha Detector (LRAD)

The long-range alpha detector (LRAD) measures radioactivity by quantifying the ionization of ambient air enclosed within the detector volume caused by passing radiation (Bounds and MacArthur, 1994). Alpha radiation is detected preferentially, because neutron, beta, and gamma radiation deposit less energy per unit distance traveled in air for a given activity level. Because of the short range of alpha particles, many ions are created near the source. These ions can be detected as they induce an electrical current in a grounded, electrostatic detector. The intensity of the current is proportional to the extent of air ionization, and to the causal, dominantly alpha, activity. The LRAD detector demonstrated at Fernald consisted of a shallow box, approximately one meter square, that is placed open end down on the soil to be measured. Alpha particles emitted from approximately the upper 10 to 20 micrometers of the soil surface can produce measurable ionization.

Sodium-Iodide Scintillometer (NAD)

Sodium-iodide scintillometers are the classic radiation detection equipment in use since the 1950s. The scintillometer uses a thallium-activated sodium-iodide crystal that converts adsorbed gamma-ray photons to visible light, which is then detected by a photomultiplier tube (Fermco, 1994). A typical detector crystal measures 2 inches in diameter and 2 inches high. The sodium-iodide detector is

sensitive to gamma radiation more energetic than about 60 keV, which does include photons from sources other than the uranium decay chain. A thin lead shield may be placed around the detector to decrease sensitivity to very high energy cosmic radiation.

X-Ray Fluorescence Unit (XRF)

The X-ray fluorescence unit (XRF) demonstrated at the Fernald site consists of a field portable unit containing three radioactive sources, ^{55}Fe , ^{109}Cd , and ^{241}Am (Fermco, 1994). A sample, which may be solid, liquid, or powdered, is exposed to radiation from one or more of these sources. Atoms within the sample absorb this radiation, and re-emit X-ray photons (i.e., fluoresce) with characteristic energy. These secondary X-ray photons are then absorbed by a mercuric-iodide crystal, lose energy, and produce a current pulse in the crystal that is proportional to the energy given up by the photons. Processing of the current pulse knowing the excitation-source radiation allows determination of both the identity and quantity of various elements present in the sample. The specific commercial XRF unit used at Fernald (Spectrace 9000®) is configured and "tuned" for detection of specific elements of common environmental interest, including uranium and thorium.

Environmental Conditions

Plans for the 1994 summer field characterization demonstration called for a number of environmental variables to be recorded for each standard-site measurement by each alternative characterization technology (Tidwell, 1994). The principal environmental conditions recorded were temperature and relative humidity. Daily rainfall accumulations were also recorded. The timing of these precipitation events during the day is not available.

Figure 2 summarizes available data regarding temporal variations in the major

environmental variables throughout the course of the field characterization demonstration. Dates and times were converted to fractional days measured from an arbitrarily selected reference of midnight, May 1, 1994. These day values are plotted on the x-axis in this and subsequent time-sequence illustrations throughout this report. In addition to the temperature and relative humidity data recorded for the majority of the standard-site measurements, daily rainfall accumulations are shown as bars associated with the relative humidity plot. Measurable rainfall events typically are associated with episodes of higher relative humidity.

Also shown in figure 2 are the times of the various standard-site measurements for each alternative characterization technology. Replicate measurements made over a very short period of time may plot as a "single" symbol at the scale of the illustration. The heavy horizontal bars on the diagram connect the first and last standard-site measurement by each different technology, and are included merely to emphasize the various periods of field activity.

Soil Geochemical Data and Ground Truth

Soil samples taken from the standard sites after completion of the field demonstration program and analyzed in the laboratory using EPA-certified ICP (inductively coupled plasma) mass-spectrometry procedures serve as the ground truth against which to evaluate the various alternative characterization technologies. Although the soil samples were collected following demonstration of the other technologies, there is no evidence from other Fernald remediation activities that uranium contaminant levels change over a time scale of weeks to months.

Surficial soil samples were collected by scraping an area of soil to a depth not exceeding one centimeter in depth. Sample

Figure 2. Temporal variations in temperature, relative humidity, and daily rainfall accumulations during the 1994 summer field demonstration. Periods of field activity for each alternative characterization technology are shown in the center of the diagram. From Rautman and others (1995).

mass was approximately 500 g (Tidwell, 1994). Five such soil samples were collected from each standard site, representing four corners and a central sample. These individual samples were not composited. However, because each sample was of approximately the same size, the results may be averaged and used as a reasonable composite uranium value for the standard sites. The measured soil geochemical analyses obtained at the two standard sites are tabulated in table 2; a statistical

summary of these soil geochemistry values is presented in table 3.

Although there may be questions regarding the "real" accuracy of soil geochemistry measurements, data produced by this method are routinely accepted by regulatory agencies, and thus serve as a reasonable basis for comparison. Collectively, the individual soil samples also contain information regarding the small-scale variability of the uranium contamination in this part of the Fernald site.

Table 2: Laboratory measurements of uranium contamination at the standard sites

[Analyses by ICP mass spectrometry; values are total uranium in picocuries per gram. Data from Rautman and others (1995), Table G-8]

Site 1		Site 2	
Sample No.	Total U	Sample No.	Total U
LAB104	74.82	LAB109	49.84
LAB105	57.30	LAB110	47.99
LAB106	68.18	LAB111	52.23
LAB107	76.19	LAB112	46.98
LAB108	70.70	LAB113	46.93

Table 3: Statistical summary of laboratory uranium analyses for soil samples from standard sites

[All values are total uranium in picocuries per gram, except as noted. Std. Dev., standard deviation; C.V., coefficient of variation, a standardized measure of variability, is defined as the standard deviation divided by the mean, and generally is expressed in percent; N, number of samples]

	Site 1	Site 2
Mean	69	49
Std. Dev.	7.5	2.3
C.V., %	11	4.6
Minimum	57	47
Maximum	76	52
N	5	5

Because of the small size of these individual samples, variability of the magnitude indicated in table 3 appears to represent a more-or-less irreducible uncertainty in the representation of any sizeable physical area by a single small sample. This variability is shown graphically in figure 3. Additional information regarding reproducibility of the soil sampling/laboratory analysis characterization technique has been provided for duplicate field samples by Rautman and others (1995, table 10).

Figure 3. Average uranium activity and small-scale variability of laboratory measurements of uranium contamination at standard sites 1 and 2 (table 2). Error bars represent plus and minus one standard deviation from table 3.

Results

A summary of the standard-site measurements for each alternative characterization technology other than the ICP-AES technology is presented in figure 4(a) for standard site 1 and in figure 4(b) for standard site 2. The data are presented in box-plot form. A box plot consists of a rectangular box, the ends of which indicate the value of the first and third quartile values of the set of measurements. The second quartile, or median value is indicated by a bar within the box itself. Fingers extend above and below the box to represent the tenth and ninetieth percentile values, and they provide a measure of the "tails" of the distribution. Values that are less than the tenth or greater than the ninetieth percentile are shown individually as dots at their appropriate value. The number of individual measurements for each alternative technology is indicated above or below the appropriate box plot. A standard statistical summary of the standard-site measurements is presented in numerical form in table 4.

Figure 4. Comparative box plots showing variability of repeated measurements of uranium activities by alternative characterization technologies at (a) standard site 1 and (b) standard site 2. Horizontal lines represent the mean $\pm$ one standard deviation for the laboratory-determined uranium activity from table 3. Numeric values are number of replicate measurements for each technology.

Table 4: Statistical summary of measured uranium activities at standard sites

[Measurement values are total uranium in picocuries per gram. Std. Dev., standard deviation; C.V., coefficient of variation, a standardized measure of variability, is defined as the standard deviation divided by the mean, and generally is expressed in percent; N, number of samples. ATD, alpha-track detector; Beta, beta scintillometer; EIC, electret ionization chamber; FID, FIDLER detector; GML, low-mount gamma spectrometer; GMH, high-mount gamma spectrometer; LRAD, long-range alpha detector; NAD, sodium-iodide detector; XRF, field X-ray fluorescence unit]

Technology	ATD	Beta	EIC	FID	GMH	GML	LRAD	NAD	XRF
<i>standard site 1</i>									
Mean	73	67	34	91	75	82	46	130	86
Std. Dev.	98	7.4	55	12	19	8.9	33	11	8.1
C.V., %	134	11	162	13	18	11	71	8	9
Minimum	14	56	-4.9	68	42	72	-13	116	75
Maximum	187	82	73	104	86	92	79	155	104
N	3	19	2	21	9	9	10	18	18
<i>standard site 2</i>									
Mean	25	54	15	76	50	48	69	118	64
Std. Dev.	24	12	10	16	6.0	8.0	31	7.1	9.6
C.V., %	97	22	68	17	12	17	45	6	15
Minimum	5	34	8.0	54	42	36	-13	108	39
Maximum	52	86	23	98	56	56	100	132	77
N	3	17	2	20	7	8	9	17	17

Variability of the repeated measurements by the different technologies at the two standard sites is apparent in the figures. Some technologies, such as the beta scintillation counter (Beta), the high-mount and low-mount high-resolution gamma spectrometer (GMH and GML), the sodium-iodide detector (NAD) and the field X-ray fluorescence unit (XRF), generally report a restricted range of values, although a few erratic outliers may be reported. Other technologies, such as the passive alpha-track detector (ATD), the electret ionization chamber (EIC), the FIDLER gamma scintillometer (FID), and the long-range alpha detector (LRAD), report replicate readings that are quite variable. The long-range alpha detector actually reported at least one negative value for uranium activity at standard site 1 [off-scale finger in fig. 4(a)]. In general, the technologies exhibiting the largest spread of measurements are those that took the smallest number of replicate measurements. However, a small number of repeated measurements does not necessarily correspond to a wide spread of values.

Another observation regarding the comparative performance of the several alternative characterization technologies is possible through examination of figures 4(a) and 4(b). Note that some technologies, despite producing a larger or smaller spread of measurements, tend to report one absolute level of uranium activity, whereas other technologies tend to report markedly different levels. For example, at standard site 2 [fig. 4(b)], the beta scintillometer and the high-mount and low-mount gamma spectrometers reported median uranium activities of approximately 50 picocuries per gram, the same value reported by the laboratory analyses of the soil samples. In contrast, the FIDLER scintillometer, the long-range alpha detector, and the field X-ray fluorescence detector reported median activities of about 70 picocuries per gram. The sodium-iodide detector reported the highest median

activity of approximately 115 picocuries per gram, whereas the alpha-track detector and the electret ionization chamber technologies reported the lowest observed values of approximately 20 picocuries per gram. A similar, although not identical, grouping of technologies is identifiable within the data for standard site 1 [fig. 4(a)]. Clearly, the different alternative characterization technologies are not "seeing" the same thing, even though each technology returned to the same physical location to repeat the measurements.

The individual technologies obtained values of the same absolute magnitude with only varying degrees of success. The range of the replicate values and the degree of skewness associated with some of the box plots suggests that external factors may have influenced some measurements but not others. The physics of some measurement techniques, in particular, suggest that readings may be sensitive to external environmental factors, such as temperature, humidity, or soil-moisture content. Each alternative technology is examined individually in the sections that follow. Measurement values are plotted sequentially as a function of time, together with the suspected environmental controls, temperature and humidity, generally recorded within approximately 30 minutes of the uranium measurement.[†] Daily rainfall amounts are also plotted for identification of correlation with anomalous uranium measurements. Note that whenever possible, the variability of uranium activities and of the environmental variables has been plotted at the same scale for ease of comparison. This con-

[†]In some instances, no environmental measurements were collected in direct association with the standard-sites measurements. In these cases, the closest available temperature and humidity data were selected if they were recorded within 4 hours of the uranium measurement. The maximum time difference is indicated on the relevant figures. Data for the longer time-lag pairs (greater than 1 hour) were omitted from correlation analyses whenever possible.

sistency of scale is generally maintained, even if the origin of the displayed uranium-activity axis had to be shifted to accommodate the absolute magnitude of the measurements represented by a particular alternative characterization technology.

The inductively coupled plasma-atomic emission spectroscopy technique is unique in that no measurements were obtained by this alternative characterization technology at the field standard sites. Results of replicate measurements of samples taken from the manufactured calibration beds serve a similar purpose, in terms of evaluating detector stability. However, because the results, strictly speaking, are not directly comparable to the field standard-site measurements made by the other technologies, the ICP-AES data will be discussed separately.

Alpha-Track Detector

The time sequence of uranium activities measured by the passive alpha-track detector and its associated environmental variables is shown in figure 5(a) for standard site 1 and in figure 5(b) for standard site 2. The correlation of measured uranium activity with temperature and humidity is shown explicitly in figures 6(a) and 6(b). Although there are only three measurements for either standard site, which constitute very small samples for statistical purposes, it is not evident that there is any control of measured uranium by these environmental conditions. The coefficients of determination (r^2) for the two cross plots in figure 6 are not significant (less than 0.5). There is no apparent correspondence of uranium activity with daily rainfall accumulations as reflected in the sequence plots of figures 5 (a) and (b).

Figure 5. Sequential plot of alpha-track detector data for (a) standard site 1 and (b) standard site 2, showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Also shown is a boxplot summary of measured uranium activities (see text for definition of box plots). Note that uranium activity scale in (a) is twice the scale used in succeeding, similar figures. Maximum time difference: 30 minutes.

Figure 6. Cross plot of alpha-track detector data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 30 minutes for both (a) and (b).

Consideration of the alpha-track detector technology suggests that no direct correlation of these environmental conditions with measured uranium activity is expected. The alpha-track detector is a passive technology, which involves emplacing the detector in the ground and retrieving it some time later and counting the number of tracks left by alpha particles in the detector medium. Thus, there is no reason to suspect that near instantaneous measurements of temperature or humidity taken at the time of retrieval would affect the reported measurement, particularly for an extended measurement period. Available field notes indicate that the alpha-track detectors were emplaced overnight for an exposure period of slightly less than 24 hours. The measured uranium activity might be a function of some integrated measure of environmental variation over the period of time the detector was left in place. However, it is unclear what "integrated measure" would be appropriate, given the available meteorologic data. Furthermore, it is unclear that the factor-of-four variation observed in reported uranium values collected on the same day for standard site 1

[fig. 5(a)] could be explained by any reasonable integrated environmental variable, which, presumably, would have affected both measurements equally.

Although it is not possible to identify from the available data any reasonable environmental control on the uranium activities measured by the alpha-track detector, the time-sequence plots of figures 5 (a) and (b) suggest that some systematic factor is influencing these measurements. Specifically, the last measurement from each standard site is markedly higher than either of the other two values. There is no remarkable information contained in the field notes associated with this later measurement episode, and the origin of these elevated measurements is unknown.

Beta Scintillometer

The time sequence of baseline uranium activities measured by the beta scintillometer, taken over a one-week primary testing period that was followed by an additional set of readings five days later, is shown in figure 7 (a) for

Figure 7. Sequential plot of beta scintillometer data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Maximum time difference: 2 hours 15 minutes for (a); 45 minutes for (b).

Figure 8. Cross plot of beta scintillometer data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 30 minutes for (a) and 45 minutes for (b).

standard site 1. Figure 7 (b) shows the equivalent graph for standard site 2. Compared to some of the other alternative characterization technology readings, the uranium activities measured by the beta scintillometer are reasonably stable and consistent across the series.

Cross plots of measured uranium activities against the environmental variables, temperature and relative humidity, are presented in figures 8 (a) and (b). There is no obvious correlation of measured uranium activity with these primary environmental variables; r^2 val-

ues are less than 0.1. The time-sequence plot of figure 7(a) (standard site 1) suggests a weak correlation with precipitation, presumably through the influence of soil-moisture content. Initial uranium measurements on days 67 and 68 are approximately 70 to 75 picocuries per gram. Note the cluster of some five readings of approximately 50 to 55 picocuries per gram on day 69, which immediately follow a rainfall event of nearly one-half inch on day 68. A day later, on day 70, the measured activities rebound slightly to approximately 65 picocuries per gram. No definite information is available regarding the exact timing of the rainfall event with respect to the standard-site measurements. However, there is no mention of rain in the field notes, so presumably the precipitation of day 68 occurred during the early morning hours prior to the day's field activities. Measured uranium activities are somewhat reduced on day 76, three days following a heavy rainfall event of nearly three inches.

This pattern of reduced uranium measurements following precipitation events is not repeated particularly well, however, for the readings taken at standard site 2 [fig. 7(b)]. There is a suggestion of reduced uranium activities from the initial two readings taken on day 69, but later measurements taken that same day are approximately equal to those taken prior to the rainfall event of the preceding day. The two lowest measurements do follow (by three days) the heavy rainfall event on day 73, as was the case for readings taken at standard site 1. Expanding the uranium activity scale of the figure aids in identifying these weak trends.

Electret Ionization Chamber

Diagrams showing the time sequence of uranium measurements using the electret ionization chamber detector are shown in figures 9(a) and 9(b). There are only two such measurements, taken nearly seven weeks

apart. Accordingly, the concept of a time series is relatively meaningless. The temporally closest available environmental measurements have also been plotted on the figures; note that for the first measurements on day 31, the environmental variables were recorded nearly four hours before the uranium measurements. Cross plots of uranium activity against temperature and relative humidity readings are presented in figures 10 (a) and (b).

Figures 9 (a) and (b) clearly indicate that the replicate uranium measurements by the electret ionization chamber are markedly different. However, because of the minimal sample size of two readings, it is possible only to speculate on the cause(s) of these differences. Although there is little direct evidence, an obvious feature evident in both parts of figure 9 is that the second, markedly lower uranium measurement was obtained a few days after a major precipitation event. Cumulative rainfall over the preceding week probably was in excess of three inches. Given that the electret ionization chamber is an alpha-particle detector, it is likely that the large increase in soil moisture that would have resulted from heavy rains may have affected the quantity of alpha particles being released from the soil.

Control of the electret ionization chamber readings by moisture-related phenomena is also clearly indicated by the environmental variable cross plots of figures 10, parts (a) and (b). High uranium activities are associated with lower relative humidity readings and lower uranium activities are associated with high humidity readings. The sample size of two, however, virtually assures a strong regression relationship of some type, as two points always define a straight line. The actual validity of this apparent relationship is unclear.

FIDLER Detector

Plots showing the time sequence of uranium activities measured by the FIDLER

Figure 9. Sequential plot of EIC detector data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Note the vertically shifted uranium activity scale for (a). Maximum time difference: 4 hours for both (a) and (b).

Figure 10. Cross plot of EIC detector data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 4 hours for both (a) and (b).

detector at the two standard sites are shown in figures 11(a) and 11(b). The corresponding cross plots with the environmental variables are presented in figures 12 (a) and (b).

The time sequence plots indicate quite a bit of intra-survey variability, although the control for this variability is not immediately obvious. The cross plots of figures 12 (a) and (b) do not indicate meaningful control by

either temperature or humidity. The highest r^2 value associated with figure 12(a) and 12(b), is less than 0.2. The lowest uranium activities measured at standard site 1 correspond to some of the times of highest humidity [fig. 12(a)]. However, high uranium activity levels were also measured at even higher humidities.

The two time-sequence illustrations [figs. 11 (a) and (b)] do exhibit some common

Figure 11. Sequential plot of FIDLER detector data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Note vertically shifted uranium activity scale. Maximum time difference: 15 minutes for both (a) and (b).

Figure 12. Cross plot of FIDLER detector data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 15 minutes for both (a) and (b).

features, particularly if plotted with an expanded uranium activity scale, suggesting a common external influence. Both sets of readings start out high, decline noticeably followed by a relatively sharp rebound during day 40, then decline again, and eventually return to relatively high values (including for standard site 1, the highest observed value [fig. 11(a)]).

These changes in measured activity potentially correspond to changes in soil moisture content, as suggested by the bars indicating daily rainfall totals. The most noticeable decreases in measured uranium values at standard site 1 occur on days 36 and 43, when the Fernald site received measurable rainfall after two to three days of no precipitation. Reduced activities

also occur on day 39, again associated with measurable precipitation. Note, however, that a relatively high measured uranium activity was also measured on day 36. Exact intra-day timing of rainfall with respect to the timing of the standard measurements is not available; however, available information from field notes suggest that the rainfall event of day 36 occurred after completion of the standard site measurements.

High-Mount Gamma Spectrometer

Baseline uranium activities measured by the high-mount gamma spectrometer are presented in figure 13(a) for standard site 1 and in figure 13(b) for standard site 2. The correlation of these measurements with the environmental variables, temperature and relative humidity, is presented in figures 14 (a) and (b).

Figure 13. Sequential plot of high-mount gamma spectrometer data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Maximum time difference: 30 minutes for (a), 15 minutes for (b).

Figure 14. Cross plot of high-mount gamma spectrometer data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 30 minutes for (a) and 15 minutes for (b).

The high-mount gamma data are similar, both in magnitude and in variability, to those obtained by this device in the lower mounting position (GML; see page 19). The time-sequence profiles are relatively flat at both standard sites, with the noticeable exception of the final measurement taken at standard site 1 [fig. 13(a)]. This datum clearly stands out as an outlier on the box plots of both figures 4 and 13(a). This anomalous measured activity corresponds to the highest observed relative humidity reading. However, the time-sequence profile clearly indicates that other high humidity values did not appear to influence the other replicate uranium activity measurements. Barring the formation of condensation in the instrument, or some similar problem uniquely associated with this particular datum, there is no evidence for general control of measured uranium activity by either temperature or humidity. Coefficients of determination (r^2) computed from figures 14 (a) and (b) are all less than 0.3. Interestingly, the nearly synchronous measurement at standard site 2 indicates no similar effect, whatsoever. This observation also suggests that a unique,

instrument-related effect probably is responsible for this one outlier at standard site 1.

Low-Mount Gamma Spectrometer

The sequence of baseline uranium activities measured by the low-mount gamma spectrometer is shown for standard site 1 in figure 15(a) and for site 2 in figure 15(b). The associated environmental-variable cross plots are presented in figure 16, parts (a) and (b).

The uranium activities measured by the low-mount gamma detector are perhaps the least variable of all the measurements obtained by any characterization technology (see also fig 4, table 4). In both figures 15(a) and 15(b), the time-sequence profile is essentially flat. There is virtually no correlation between either temperature or humidity and the measured activity observable in figures 16 (a) or (b); the r^2 values are less than 0.2. These measurements appear relatively insensitive to soil moisture as well, although again, the exact intra-day timing of rainfall and individual replicate measurements is unknown.

Figure 15. Sequential plot of low-mount gamma spectrometer data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Maximum time difference: 15 minutes for both (a) and (b).

Figure 16. Cross plot of low-mount gamma spectrometer data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 15 minutes for both (a) and (b).

Long-Range Alpha Detector

Time-sequence profiles of the measurements obtained by the long-range alpha detector are shown in figures 17 (a) and (b) for standard sites 1 and 2, respectively. The corresponding environmental-variable cross plots are presented in figures 18 (a) and (b). The long-range alpha detector measurements are clearly anomalous, in that the technique produced out-of-range (negative) readings at standard site 1 [fig. 18(a)]. A very similar, large-magnitude decrease in the observed uranium activity was also observed at standard site 2, at nearly coincident times [fig. 18(b)]. This temporal coincidence strongly suggests some form of environmental influence.

The cross plots of measured uranium activity against the environmental variables, as well as the time-sequence profiles, suggest a weak correlation of measured activity with temperature and with relative humidity. Figure 17(a) clearly indicates that the first five, generally increasing uranium values measured over days 38, 39, and 40, coincide with generally decreasing relative humidities. The same pat-

tern can be detected in figure 17(b). This correlation between humidity and measured uranium activity is not well exhibited, however, in the cross plots of figures 18 (a) and 18(b). Instead, there is a suggestion that temperature may be more responsible for the change in measured activity, and that the temperature scale involved in the time-sequence diagrams may be obscuring this relationship in those figures. The r^2 values associated with these correlations of measured uranium activity with temperature are 0.46 and 0.39 for figures 18 (a) and (b). Thus, in any event, the control by temperature is not a strong one.

Although the temporal resolution of the precipitation data is insufficient to make strong statements, the occurrence of the negative measured uranium values with the largest rainfall accumulation on day 43 [fig. 17(a)] suggests that soil moisture may be controlling the erratic uranium activities observed by the long-range alpha detector. Available field notes regarding soil moisture conditions indicate that conditions changed from "moist" to "fairly dry" to "dry" on days 38 to 40, changing to "very damp" on day 43 and returning to

Figure 17. Sequential plot of long-range alpha detector data for (a) standard site 1 (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Note expanded uranium activity scales in both (a) and (b). Maximum time difference: 15 minutes for both (a) and (b).

Figure 18. Cross plot of long-range alpha detector data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 15 minutes for both (a) and (b).

“dry” on day 44. Measurements by the LRAD device may be very sensitive to small changes in water content, as indicated by the rapid rebound of measured uranium values during day 43 for both standard sites [figs. 17 (a) and (b)]. Such sensitivity would be consistent with the physics of alpha-particle emission and travel from soil with time-varying water content. More precise information regarding the

timing of rainfall on days 38 and 39 with respect to the generally increasing measured uranium activity values would be required to confirm this potential relationship. The available data suggest that quantitative measurements of soil moisture content appear to be necessary to provide compensation for variations in this environmental condition.

Sodium Iodide Detector

The time sequences of replicate measurements made by the sodium-iodide detector and their associated environmental variables for the two standard sites are presented in figures 19, parts (a) and (b). Cross-plot diagrams of the correlation between measured uranium activities and both temperature and relative humidity are shown in figure 20 (a) and (b).

The measurements made by the sodium-iodide detector are relatively consistent over time, particularly for standard site 2 [fig. 19(b)]. However, as indicated by the comparative box plots of figures 4 (a) and 4(b), the measured uranium activities are markedly higher than those measured by most of the other technologies. The source of this apparent calibration discrepancy is not immediately obvious.

Figure 19. Sequential plot of sodium-iodide detector data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Note vertically shifted uranium activity scales in both (a) and (b). Maximum time difference: 4 hours for both (a) and (b).

Figure 20. Cross plot of sodium-iodide detector data for (a) standard site 1 and (b) standard site 2 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 30 minutes for (a) and 15 minutes for (b).

Two readings at standard site 1 (fig. 19) on days 40 and 47 are clearly anomalous, as these two measurements fall outside the 90th percentile range of the measurement set. No environmental cause is apparent for these aberrant readings, although relative humidity data appear not to have been recorded coincident with one of these field measurements ("no data" in fig. 19). The cross plots of figures 20(a) and 20(b) do not suggest a strong relationship between measured activity and humidity; in fact, the r^2 values associated with these figures are less than 0.1. These same illustrations do indicate a weak inverse correlation with ambient temperature. The coefficients of determination for this correlation are only 0.37 and 0.62 for standard sites 1 and 2, respectively, indicating that the effect of temperature is quite weak, indeed. Reference to figures 19 (a) and (b) does not indicate any particular correlation of changes in measured uranium activity with daily rainfall occurrence or amounts. Presumably, the sodium-iodide detector is relatively insensitive to changes in soil moisture.

Field X-Ray Fluorescence Detector

The replicate measurements of uranium activity for the two standard sites by the field x-ray fluorescence detector are shown in figure 21 (a) and (b), as well as the associated temporal variations of the environmental variables. Figure 22, parts (a) and (b), presents cross plots of measured uranium activity as a function of temperature and humidity.

The measurements for the two standard sites are relatively consistent, although figure 21(b) indicates that the first and last measurements taken at standard site 2 are relative outliers. There is no obvious correlation with the environmental variables that accounts for these rather marked changes in measured uranium values. A major rainfall event occurred on the first day of the survey (day 36). However, no such precipitation event was associated with the similarly anomalous measurement taken on day 61. Temperature and humidity readings were not acquired following the initial stage of the field XRF survey, which was completed on day 46.

Figure 21. Sequential plot of X-ray fluorescence detector data for (a) standard site 1 and (b) standard site 2 showing uranium activities, measured temperature and humidity, and daily rainfall quantities as a function of time. Note vertically shifted uranium activity scale in (a). Maximum time difference: 1 hour 15 minutes for (a), 1 hour 30 minutes for (b).

Figure 22. Cross plot of X-ray fluorescence detector data for standard site 1 showing measured uranium activities as a function of the temperature and relative humidity prevailing at the time of measurement. Maximum time difference: 45 minutes for both (a) and (b).

The cross-plot diagrams of figures 22 (a) and (b) do not indicate any correlation between measured uranium activity and temperature or relative humidity; r^2 values are less than 0.1. This analysis is also complicated by the lack of environmental data for the measurements taken after day 46.

Laser Ablation-Inductively Coupled Plasma-Atomic Emission Spectroscopy

Replicate measurements were not taken at the field standard sites by the field-adapted laser ablation-inductively coupled plasma-atomic emission spectroscopy technology. Thus, no direct comparison between the accuracy of the ICP-AES technology and that of the other alternative characterization technologies is possible. However, the technology did obtain replicate measurements for several of the manufactured calibration beds used by both this and all other technologies to relate raw measurement values to total uranium activities (Rautman and others, 1995). These replicate data make it possible to compare the results of these replicate measurements to the

laboratory measurements of the calibration beds and to draw some conclusions regarding the stability of this detector technology.

The calibration beds consisted of uranium-spiked soils, homogenized and placed in square pans measuring 1.6 meters on a side to a depth of 22.5 cm (Tidwell, 1994). "True" activities of these calibration beds were determined through replicate soil sampling and laboratory analyses. A statistical summary of these laboratory measurements is presented in table 5.

Figure 23 shows a set of box plots showing the variability of ICP-AES measured uranium activities for three of the calibration beds created for the 1994 Fernald field characterization demonstration program. A statistical summary of the ICP-AES measurements is given in table 6. No environmental variables were measured in association with the ICP-AES determinations.

Several observations can be made from figure 23 and the associated tabular information. First, the ICP-AES measurements gener-

Table 5: Statistical summary of replicate laboratory measurements of soil samples from three calibration beds

[All values are total uranium in picocuries per gram except as noted. Std.Dev., standard deviation; C.V., coefficient of variation, a standardized measure of variability, is defined as the standard deviation divided by the mean, and is generally expressed in percent; N, number of measurements]

	Calibration Bed		
	C0	C35	CP
Mean	5.73	94.57	51.09
Std. Dev.	0.42	6.76	18.54
C.V., %	7	7	36
Minimum	5.26	84.26	26.15
Maximum	6.45	97.33	79.66
N	7	7	7

Table 6: Statistical summary of replicate "standard" measurements by the inductively coupled plasma-atomic emission spectrometry technique

[All values are total uranium in picocuries per gram except as noted. Std.Dev., standard deviation; C.V., coefficient of variation, a standardized measure of variability, is defined as the standard deviation divided by the mean, and is generally expressed in percent; N, number of measurements]

	Sample		
	"C0"	"C35"	"CP"
Mean	12	115	134
Std. Dev.	13	145	20
C.V., %	106	126	15
Minimum	-2.6	58	111
Maximum	44	893	148
N	16	32	3

Figure 23. Box plots showing total uranium activities measured for samples from the three calibration beds by the ICP-AES technology. Numeric values next to box plots indicate number of replicate measurements. Smaller, lighter grey box plots to left of each ICP-AES box plot show replicate laboratory analyses of calibration beds (equivalent to fig. 3). Note expanded uranium activity scale and extreme-valued outlier for sample C35.

ally span a broader range than the equivalent laboratory measurements, especially for the two calibration beds that were subjected to a statistically meaningful number of measurements. In this respect, the technique is less precise. Second, the average value measured by the ICP-AES technique does not accurately or consistently approximate the true activity of the three calibration beds (tables 5, 6). The ICP-AES values overestimate the activity of the C0 bed and underestimate that of the C35 bed. In this latter case, however, the true activity is contained within the interquartile range of the replicate measurements. If the clearly anomalous outlier value of 893.15 pCi/g is omitted from the calculation for sample C35, the mean value drops to 89.9 pCi/g, thus further underestimating the uranium activity of the corresponding calibration bed. Third, the ICP-AES measurements for sample CP appear to be seriously in error. Indeed, the range of values measured for sample CP by the ICP-AES device are completely outside the range of uranium activities obtained through laboratory analyses of this calibration bed.

It is important to note that the ICP-AES measurements represent replicate analyses of a *single soil sample* taken from the relevant calibration bed. If, for some reason, this physical sample was not representative of the overall material in that bed, then the technique might yield precise replicate values that bear little resemblance to the laboratory geochemical analyses. Although the wider range of values reported by the ICP-AES technique compared to the range of values obtained by the laboratory mass spectrometry method generally argues against this explanation, the non-representative-sample effect may have impacted the uranium activity measurements for sample CP. Table 5 indicates that the laboratory values themselves obtained from repeated physical sampling of this calibration bed are considerably more variable than for either of the other calibration plots; thus, the actual activity of this manufactured material is less homogeneous and more uncertain. The box plot for laboratory measurements of calibration bed CP in figure 23 clearly indicates the substantially greater spread of measured values for these replicate samples. In fact, calibration bed "CP" was created somewhat after the fact by diluting material from another calibration bed ("C200") after laboratory measurements of the numerically designated calibration plots indicated that the actual activities exceeded the desired levels (Rochelle Chernikoff, Fermco, written communication, 1995). Evidently, the more ad-hoc "CP" calibration material was not homogenized as thoroughly as the soils composing the calibration beds prepared in advance.

Soil-Moisture Correction Factors

As described on page 3, developers of the beta scintillometer and high-mount and low-mount gamma spectrometer technologies proposed to adjust the measured uranium activities to compensate for changes in soil moisture following rainfall events through

adoption of a soil-moisture correction factor. These soil-moisture correction factors attempt to equalize the temporally varying standard-sites measurement values, and to carry the necessary percentage adjustments to the non-replicated field-survey measurements in proportion to the time spacing of the affected data (A. Schilk, Pacific Northwest Laboratories, personal communication, 1994).

Figures 24 through 26 present time-sequences of the baseline and soil-moisture-adjusted uranium activities for these three technologies. These sequence plots may be compared to figures 7, 13, and 15, respectively. Note that whereas figures 7, 13, and 15 were plotted using a common activity-axis scale, figures 24 through 26 are plotted with scales that capture the full range of variability of these data. Coincident data points (that overlap on the figure) indicate soil-moisture correction factors equal to one.

Figure 24. Comparison of uranium activities for (a) standard site 1 and (b) standard site 2 measured by the beta scintillometer with and without adjustment by the soil-moisture correction factors from Rautman and others (1995), appendix G.

Figure 25. Comparison of uranium activities for (a) standard site 1 and (b) standard site 2 measured by the high-mount gamma spectrometer with and without adjustment by the soil-moisture correction factors from Rautman and others (1995), appendix G.

Figure 26. Comparison of uranium activities for (a) standard site 1 and (b) standard site 2 measured by the low-mount gamma spectrometer (with and without adjustment by the soil-moisture correction factors from Rautman and others (1995), appendix G.

The most obvious difference visible in figures 24 through 26 is that the variations in reported uranium activity as described by the interquartile ranges indicated by the several

boxplots are markedly reduced for the adjusted values (lighter grey, dashed-line square symbols) relative to the baseline values (black, solid-line circle symbols). The overall ranges of the adjusted values (indicated by the boxplot fingers and/or outlier symbols) are generally reduced as well. Even the relatively small degree of variability among the replicate measurements shown in figures 7, 13, and 15 has been significantly damped by this empirically derived adjustment process. Although the soil-moisture correction factors are effectively empirical "fudge-factors" that may encompass more than simply the effect of soil-moisture variations with time, the use of such empirical adjustments based on repeated readings at a reference location is in keeping with standard practices for many types of geophysical surveying. Now-classical reference books on conducting field geophysical surveys (e.g., Dobrin 1960; Lahey, 1961) describe the creation of "drift curves" and time- or distance-distributed corrections for "closure errors" based on this principle.

Discussion

A considerable degree of variability exists in the replicate, standard-site measurements obtained by the several characterization technologies tested at the Fernald site during the 1994 field characterization demonstration program. Although consideration of the physical differences inherent in the demonstrated technologies suggested that variation would be observed, the extent of that variation is somewhat surprising in retrospect.

The graphical summaries of the uranium measurements shown in figures 4 (a) and (b) clearly indicate at least a two-fold subdivision of the alternative technologies. Most of the measurement methods yield reasonably precise (reproducible) estimates of uranium contamination; however, the alpha-track detectors, electret ionization chambers, and long-

range alpha detector clearly lack precision. In the case of the first two of these techniques, it appears that significant problems were encountered in transferring these passive devices from their original, indoor application to the much less-controlled physical environment in the field.

The variability exhibited by the alpha-track detector is particularly confusing, given that field notes indicate that each reported "measured value" is actually the average of four individual ATD readings taken in close proximity to the marked grid location (Dudney and others, 1994). Application of the central-limit theorem from statistical theory through this averaging process would be expected to *reduce* the variability exhibited by the final, reported uranium activity.

One factor influencing the alpha-track detector measurements can be identified from the description of this measurement technology presented by Dudney and others (1994). The ATD records alpha tracks left by passing alpha particles. In and of itself, no discrimination is possible between alpha particles originating directly from decay of ^{238}U and those emanating from other sources, including environmental radon. Dudney and others state that the final measured uranium activity is computed by subtracting the average track density on the side of the ATD plastic detector that was located away from the soil from the average track density on the side closest to the soil (Dudney and others, 1994; see page 59 of Rautman and others, 1995):

The detector face in proximity to the surface will measure alpha emission due to the surface contamination plus environmental radon. The other, or distal, face will measure only alpha emission due to environmental radon.

Thus, it seems likely that small errors in counting the number of tracks present on either side

of the 1-mm-thick detector medium could either accentuate or reduce the necessary difference between the two records of alpha intensity. Unless these errors were systematically biased, the net effect would be to increase the observed level of variability in the final, composite, ATD values.

The measurements of uranium contamination obtained by the electret ionization chambers represent another case of the "measured value" being a function of a difference between two physical readings. Dudney and others (1994) state that "in mixed fields (radon, beta, and/or gamma)," a pair of detectors is used with one shielded by porous Tyvek® material (see p. 59 of Rautman and others, 1995). Uranium activity, as represented by alpha activity (presumably including that originating from decay of radon gas) is determined as the difference in observed voltage drops in the pair of detectors. Any small-scale heterogeneity in the distribution of uranium-bearing material could easily work to accentuate this difference in apparent radiation fields and thus to produce either excessively high or excessively low reported uranium values. Given a sample size of two (representing four individual EIC devices) at each of the two standard sites, there is no statistical mass to compensate for this type of erroneous differential reading.

Following completion of the field characterization demonstration program, C. S. Dudney (Oak Ridge National Laboratory, written communication, 1995) identified other potential factors that may have contributed to the anomalous performance of the ATD and EIC devices compared to the other demonstration technologies. With respect to the alpha-track detectors—as with any radiometric-based measurement technique—the results are sensitive to the counting statistics, which in turn are a function of the length of time over which those counts are acquired. Alpha-track

detectors were emplaced in the field at Fernald for approximately 24 hours. Based on more extensive, unpublished results, Dudney states that the detection limit under these conditions would be approximately 32 ± 6 pCi/g; the laboratory-measured uranium activities at the two standard sites are definitely above this limit (table 2). However, the accuracy of the alpha-track detector measurements are unquestionably limited by the fact that the true uranium activity is only a factor of two above the specified detection limit. For comparison, Dudney also states that ATD exposure times of up to six (6) days (=144 hours) have been required for accurate measurement of low alpha activities in experiments conducted at the Nevada Test Site. Dudney also suggested that condensation of moisture from humid Fernald soils on the Lantrack® strips during the overnight exposure of the alpha-track detectors may have shielded the sensitive detector medium from low-energy alpha particles.

With respect to the apparent failure of the electret ionization chamber technology, Dudney (written communication, 1995) suggested that foreign substances in the field, not normally found in the more conventional indoor applications of these devices, might have come in contact with the exposed, charged electret, thus draining stored electrical charge from these detectors in excess of the amount attributable to ionizing radiation. Contact with blades of grass was suggested as a likely possibility, although the grass at the individual sample locations was "cropped and residual thatch removed" (Tidwell, 1994). Crawling insects emerging from the natural Fernald soils during the roughly 5-hour field-exposure period could also have contributed to this problem in the relatively uncontrolled field environment.

Interestingly, the other clearly anomalous measurement technology, the long-range alpha detector, also measures uranium activity

through detection of this weak, short-range type of radiation (table 1). In this case, the measurement is a single reading of current induced in the detector electronics through the ionization of air by alpha particles. However, information presented in the Results section [figs. 17 (a) and (b)] strongly suggests that soil moisture may play an important role in the observed variability of the standard-site LRAD measurements. Unfortunately, without independent, quantitative data on actual soil moisture levels, it is impossible to compensate these readings for this effect. Presumably, it would be possible to compensate partially for general changes in soil moisture at all sample locations by normalizing those readings to accommodate "drift" in repeated measurements at a single location. This is the approach proposed by the developers of the Beta, GMH, and GML technologies (see discussion beginning on page 27).

A second group of technologies stand out — albeit less obviously — on figures 4 (a) and 4(b) as being somewhat inaccurate compared with the EPA-acceptable laboratory soil geochemistry values. This is true even though the measurements are relatively precise replications. In figure 4, both parts (a) and (b), the FIDLER, sodium-iodide, and field X-ray fluorescence measurements are distinctly higher than the actual geochemical activities. Of the two gamma-sensitive detectors (table 1), the sodium-iodide detector, in particular, reports nearly twice the magnitude of uranium contamination at both standard sites, whereas the FIDLER measurements are approximately 50 percent higher. The origin of these apparent mis-calibrations is unclear, as both devices appeared to be well calibrated based on the data presented in Rautman and others (1995).

As noted in the description of the alternative characterization technologies, the sodium-iodide detector does not discriminate well among gamma radiation from various

sources (see page 6 and following). The FIDLER detector was specifically designed to avoid some of the excess sensitivity of the more massive NAD detector crystal to non-target gamma rays (Fermco, 1994). The difference in measured uranium levels by these two techniques is consistent with decreased sensitivity to high-energy gamma rays by the FIDLER detector. The available information suggests that calibration of these two instruments may not have been conducted under the same gross-background radiation conditions prevailing in the field.

Because the field x-ray fluorescence unit is a chemical, rather than a radiological, measurement technique, it is unclear what factor appears to be systematically influencing these replicate measurements. In each case, the number of replicate measurements is sufficiently large (minimum of 17) that these differences appear not to be simply the result of statistical fluctuations. "Matrix" effects, in which evaluation of the target photon-energy peak(s) is confounded by unrelated secondary peaks from other fluorescing elements or by excess adsorption of target photons by other sample constituents, are well known in laboratory x-ray fluorescence studies. However, the calibration beds for the Fernald field characterization demonstration program were manufactured by spiking local soils with uranium contamination and analyzing the resulting material using mass spectroscopy. Without more complete bulk chemical analyses to provide quantitative information on the presence of interfering elements, the source(s) of this systematic high bias is uncertain.

Of the remaining measurement technologies, differences in the high-mount and low-mount gamma scintillometer readings are consistent with the differences in scale of these measurements. The GMH readings are consistently lower than the GML values, as would be expected as the field of view of the scintillom-

eter increased to include heterogeneities that represent potentially less contaminated material that was excluded from measurement in the lower-positioned, collimated configuration. At both standard sites 1 and 2, the beta scintillometer consistently performed best in replicating the measured soil geochemistry values.

Effect of Sample Volumes on Measurement Variability

The existence of spatial heterogeneity in the distribution of uranium contamination suggests that some variability in activity of replicate measurements may be caused by variability in the quantity of material "observed" or otherwise sampled by the different characterization technologies. The effect of changes in the scale of a measurement ("measurement support," in geostatistical terminology) on the variance of a set of those measurements is relatively well known (Journel and Huijbregts, 1978; Clark, 1979). In contrast to the variance, the mean of additive properties, such as composition, is generally unaffected by changes in measurement support. Information summarized in the section on a "Brief Description of Alternative Characterization Technologies," beginning on page 3, suggests that there are orders-of-magnitude differences in the effective scale of material measured by the different technologies. Thus, it is important to consider the effect of sample volumes on measurement variability in comparing the performance of the different methods.

Figure 27 presents the relationship between the sample volume interrogated by each alternative measurement technique and the observed standard deviation of the replicate uranium measurements from the Fernald standard sites. Note that both axes of the diagram are scaled logarithmically. Details of the volume computations, as well as a table sum-

marizing the estimated total volumes, for each technique are presented in Appendix A. The standard deviations of the replicate measurements are taken from tables 3, 4, and 6.

Figure 27. Relationship between the computed approximate volume sampled by each alternative characterization technology and the standard deviation of the measurements reported by that technology at the two standard sites.

Figure 27 indicates a definite inverse dependence of observed variability of measurement values on the volume of material sampled: the smaller the sample volume, the greater the variation in measured activity. This correlation exists across nearly nine orders of magnitude change in sampling scale, and the relationship holds for both standard sites.

The correlation between sample volume and measurement variability shown on this figure is not particularly strong; the associated coefficient of determination (r^2 value) is less than 0.35. However, examination of figure 27 suggests that the low r^2 is, in part, caused by a few data points that plot substantially off the regression line, particularly those with large standard deviations. These points include the EIC detector (site 1), the alpha-track detector

(site 1) and the LRAD system (both sites 1 and 2). These are the same alternative characterization technologies that displayed anomalously large variability on figures 4 (a) and (b). If these points are excluded from the computation of the coefficient of determination, the resulting r^2 value increases to 0.41. Significantly, all three of these technologies are those based on detection of alpha particles (table 1). Additionally, the markedly larger standard deviations reported for the EIC and alpha-track detectors from standard sites 1 and 2, given the somewhat similar sample volumes, is compatible with the fact that these two technologies compute the "measured" uranium activity from the difference between two individual physical readings.

Conclusions

A principal conclusion of this evaluation of measurement reproducibility using the replicate standard-site data from the Fernald site is that it appears that alternative characterization technologies based on detection of alpha particles are generally unreliable and non-robust under actual field operating conditions. The extreme variability and low accuracy of replicate alpha-particle measurements is attributed to two primary causes. First, the physics involved in detection of alpha particles renders techniques based on this type of radiation extremely sensitive to soil moisture contents. Absent independent soil-moisture data (for example, from calibrated neutron moisture meter readings) by which to adjust the measured alpha values, absorption of the target radiation by rapidly varying soil water contents appears to render these uranium-activity data unreliable. Second, the variability of replicate "measurements" appears to be accentuated through the use of differential physical readings designed to discriminate alpha-particles derived from uranium decay from those related to decay of ^{222}Rn . Small errors in counting alpha-particle tracks, in measuring

voltage drops for paired EIC electrets, or from small spatial heterogeneities in alpha intensities, appear to cause wide swings in the reported uranium activities. Small errors in the individual physical readings can interact to increase the apparent differences, which are then accentuated through operation of the squared term in the computational formula for standard deviations. Some of the unreliability exhibited by the alpha-track detector and the electret ionization chamber technologies may be attributed to poor experimental design. Neither of these technologies obtained sufficient statistical mass (number of measurements) to demonstrate a reliable ability to measure meaningful uranium contamination under field conditions. The potential for additional interference in these alpha-particle measurements by moisture condensing on the sensitive alpha-track-recording polymer or by foreign objects touching the charged electrets was noted following completion of the field demonstration.

The second principal conclusion of this evaluation is that the advanced gamma-ray spectrometry technologies produced the most repeatable and stable measurements. The gamma-ray spectrometer measurements of uranium contamination, in both high-mount and low-mount configurations, appear extremely robust with respect to known environmental conditions measured during the field characterization demonstration program. Infrequently occurring, potentially humidity-related variations have been observed, indicating possible internal condensation or other instrument-related effects. The uranium activities measured by the gamma-ray spectrometer were invariably higher than the activities indicated by soil geochemistry sampling. The GMH values were generally lower and less variable than those reported by the spectrometer in its low-mount (GML) configuration. These two relationships are consistent with the volume-variance relationship anticipated from consideration of the relative volumes of mate-

rial evaluated by these devices. Depending upon the regulatory setting, these volume effects may be either a significant advantage or disadvantage compared with soil geochemistry. It is unclear that the soil sampling techniques employed for the Fernald field characterization demonstration program were completely optimized in terms of regulatory compliance.

A third major conclusion is that the remaining alternative characterization technologies, the beta scintillometer and the field X-ray fluorescence device, provide relatively stable replicate readings and appear to be relatively robust with respect to the measured environmental variables. Of these two technologies, the beta scintillometer provided essentially identical to slightly lower measured uranium activities than the EPA-standard soil geochemistry values. The beta scintillometer may be slightly sensitive to major changes in soil moisture, as indicated by somewhat lower measurements following major rainfall events. Overall, however, the beta scintillometer appeared to be one of the better performers of the alternative characterization technologies examined. The field X-ray fluorescence device provided good repeatability; however, some type of calibration problem produced field measurements that were consistently higher than the corresponding soil geochemistry values. Also, the first and last measurements obtained by the X-ray fluorescence device are clearly anomalous without apparent external cause.

A fourth conclusion is that the industry-standard field measurement techniques involving sodium-iodide-crystal-based gamma scintillometry provided relatively stable measurements of uranium contamination at the standard sites. Time-sequential plots of replicate measurements indicates that the FIDLER detector appears to be responding to some external, but unknown, environmental

factor. The simpler sodium-iodide gamma scintillometer proved somewhat more robust in replicating the same uranium activity levels despite variation in several environmental variables. Both these standard measurement technologies appear to have suffered from poor calibration in moving from the calibration beds to the field standard sites. Both techniques yielded values higher than the soil geochemistry measurements, with the classical sodium-iodide detector yielding the highest average results of any of the characterization technologies. Given the sensitivity of both detectors to gross gamma radiation, it may be that overall background gamma levels at the Fernald site generally are sufficiently high that the initial calibration was inadequate.

Finally with respect to the different characterization technologies demonstrated during the 1994 Fernald field characterization demonstration program, it is possible to state that the replicate determinations of uranium activity for three different soil samples measured as "standards" by the ICP-AES device indicate that this field-adapted laboratory technology yielded particularly erratic, and hence probably unreliable, results. The experimental design associated with demonstration of the ICP-AES technology was not good, and it is not possible to compare performance of the laser-ablation inductively coupled plasma-atomic emission spectroscopy device directly with the other alternative characterization technologies.

With respect to characterization methodology, the information regarding soil-moisture correction factors presented by the developers of the beta scintillometry, the high-, and low-mounted gamma spectrometry systems strongly suggests that future characterization activities should make use of the time-honored geophysical practice of reoccupying a "standard" station *several* times during the course of each day's measurement schedule. It

is desirable to bracket (in time) all potentially meaningful environmental changes (rainfall events, passage of perceptible weather fronts). Development of well-constrained drift curves as a function of time, and the distribution of "closure" errors, probably would reduce the spread of replicate measurements presumed to occur in response to progressively changing environmental conditions. This recommendation should apply to surveys undertaken with any of the radiometric instruments demonstrated during the field characterization demonstration program.

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[†]This report refers to a number of informally prepared descriptions of the several characterization technologies and planned operational practices; collectively these are found in the "Field Demonstration Project Plan." Although the only known formally available reference to these various planning documents is as an appendix to the Fernald field characterization demonstration program data report (Rautman and others, 1995), we have adopted the practice of referring to the appropriate sections of the Project Plan as if they were separately available articles or reports (e.g., Anderson, 1994). This practice facilitates locating the subject matter within the lengthy data report itself and it accentuates attribution of the relevant original work to the many individual principal investigators and project staff.

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Appendix A: Sample Volume Calculations

Sample Volume Calculations

Variability of replicate measurements may be affected by the volume of material sampled, as well as by a number of other factors. As a numerical thought experiment, the volume of soil material sampled by each alternative characterization technology was computed from information about the technology contained in 1994 field characterization demonstration program planning documents (Tidwell, 1994). In some cases, the underlying information regarding depth of penetration for various types of radiation are only approximate. However, the estimates of the order-of-magnitude volume examined by each technology are fairly accurate, particularly because the variations in the volumes among the different techniques is so large. A summary of these volume estimates is presented in table A-1. The details of this calculation for each characterization technology and the information on which the volume estimates are based are given in this Appendix.

Table A-1: Approximate sample volumes of alternative characterization technologies
[Volumes in cubic meters]

Technology	Sample Volume
ATD	2×10^{-8}
Beta	2×10^{-3}
EIC	9.4×10^{-8}
FID	1.3×10^{-3}
GMH	1.6×10^1
GML	1×10^0
ICP-AES	6.5×10^{-8}
Lab	2×10^{-4}
LRAD	2.5×10^{-5}
NAD	2×10^{-4}
XRF	5×10^{-7}

Alpha-track Detector

The Field Demonstration Project Plan (Tidwell, 1994) describes the area sampled by

the alpha-track detector as "a few square centimeters in size." Alpha particles do not penetrate very far in soil, and the radiation reaching the plastic detector material may be presumed to come from the upper 10 to 20 μm of the soil.

If "a few square centimeters" can be estimated to an order of magnitude as about 10 cm^2 ("... a few, that is eight, ..." I Peter 3:20, NRSV), and the depth of penetration is assumed to be the maximum 20 μm , then the physical volume of material contributing to the measurement can be computed as approximately $2 \times 10^{-8} \text{ m}^3$.

Beta Scintillometer

The Field Demonstration Project Plan (Tidwell, 1994) describes the beta scintillometer detector as measuring about 0.2 m^2 . Additional descriptive information on detector construction given in Schilk and Perkins (1994) gives the dimensions of the detector as 30 cm x 60 cm, which actually computes to 0.18 m^2 . Both sources state that beta particles emanating from the top one centimeter of the soil are detectable. Multiplying area by depth of detection thus yields an active sample volume of approximately $2 \times 10^{-3} \text{ m}^3$ for the beta scintillometer.

Electret Ionization Chamber

Dudney and others (1994) describe the area sampled by an electret ionization chamber as 47 cm^2 . Assuming the maximum penetration depth of 20 μm for alpha particles, as was done for the equivalent calculation for the alpha-track detector, the sample volume for an EIC can be computed as about $9.4 \times 10^{-8} \text{ m}^3$.

FIDLER Scintillometer

Description of the FIDLER detector by Fermco (1994) states that the sodium-iodide crystal used in this scintillometer is five inches in diameter. Tidwell (1994) states that gamma

radiation is detected by this device from the "upper few centimeters" of the soil.

By converting the stated five-inch diameter to a radius of 6.35 cm, and assuming that a "few centimeters" is approximately 10 cm, it is possible to estimate the cylindrical volume sampled by the FIDLER scintillometer as about $1.3 \times 10^{-3} \text{ m}^3$.

High-Mount Gamma Spectrometer

The high-mount gamma spectrometer measures the largest volume of all the alternative characterization technologies. Tidwell (1994) describes the device as measuring gamma radiation from an area with a radius of approximately ten meters, with a depth of penetration that decreases from a maximum of 10 to 15 cm directly beneath the detector. Although the "average" depth of penetration for gamma rays is difficult to estimate precisely, a reasonable average depth might be 5 cm across the entire circular area ($=314.16 \text{ m}^2$) surveyed by the spectrometer. The cylindrical volume examined by the high-mount gamma spectrometer is thus approximated as nearly 16 m^3 .

Low-Mount Gamma Spectrometer

Tidwell (1994) states that "the detection area [of the gamma spectrometer], when operating in collimated mode, will be approximately 10 m^2 ." If we assume that the average depth of penetration by gamma rays from this much-reduced area is closer to 10 cm (of the "10 to 15 cm directly under the detector"), then the cylindrical volume sampled by the low-mount gamma detector is on the order of 1 m^3 .

Inductively Coupled Plasma-Atomic Emission Spectroscopy

Tidwell (1994) describes the ICP-AES technique as implemented at the Fernald site as

having a "sampling area of 6.5 cm^2 ," and as "affecting approximately $100 \text{ }\mu\text{m}$." These measurements allow computation of an effective sample volume of about $6.5 \times 10^{-8} \text{ m}^3$.

Soil Sampling and Laboratory Analysis

The soil samples taken for laboratory analysis are described by Tidwell (1994) as having a mass of approximately 500 g. If this sample mass is converted to a volume using the density of quartzose material (2.67 g/cm^3), we obtain a sample volume of $1.87 \times 10^{-4} \text{ m}^3$, or approximately $2 \times 10^{-4} \text{ m}^3$. This volume estimate is probably low for unconsolidated materials; however, the approximation is probably sufficient for purposes of this comparison.

Long-Range Alpha Detector

Bounds and MacArthur (1994) describe the implementation of the LRAD device demonstrated at Fernald as a one-meter-square box. Assuming maximum penetration of $20 \text{ }\mu\text{m}$ for detectable alpha particles, the sampled volume of the LRAD can be calculated as $2 \times 10^{-5} \text{ m}^3$. Bounds and MacArthur report a volume of $3.6 \times 10^{-5} \text{ m}^3$, which implies a slightly greater depth of penetration for alpha particles than that estimated by Tidwell (1994). This difference between the two estimates suggests the general level of uncertainty associated with the volumes presented in this appendix.

Sodium-Iodide Detector

The classic sodium-iodide gamma scintillometer has a "sensitive area of 20 cm^2 ," according to Fermco (1994). Tidwell (1994) describes the sodium-iodide detector as measuring an area that is "tens of square centimeters in size." The depth of sensitivity, which is given as "the upper few centimeters of soil," is taken as 10 cm for consistency with the values used for other gamma-ray detectors. Together, these data indicate that the volume of material

sampled by the sodium-iodide detector is approximately $2 \times 10^{-4} \text{ m}^3$.

X-Ray Fluorescence Detector

No quantitative information is given in the 1994 Fernald Demonstration Program Plan regarding the volume of material sampled by the field X-ray fluorescence detector. Tidwell (1994) refers to it as providing essentially a "point measurement." Laboratory X-ray fluorescence equipment may use samples in a variety of configurations. However, a typical specimen consists of a compressed powder pellet one-inch (2.54 cm) in diameter and about one-quarter to one-half inch (0.6–1.3 cm) thick. If the fluorescing material emits detectable X-rays from the top one millimeter of the sample, the volume examined can be calculated as approximately $5 \times 10^{-7} \text{ m}^3$. If the entire centimeter-scale thickness of the pellet fluoresces, the volume is approximately $5 \times 10^{-6} \text{ m}^3$.

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