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SURFACE AND SUBSURFACE CHARACTERIZATION OF
URANIUM CONTAMINATION AT THE
FERNALD ENVIRONMENTAL MANAGEMENT SITE

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EXECUTIVE SUMMARY

The past operations of uranium production and support facilities at several Department of Energy (DOE) sites have occasionally resulted in the local contamination of some surface and subsurface soils, and the three-dimensional distribution of the uranium at these sites must be thoroughly characterized before any effective remedial protocols can be established. To this end, Pacific Northwest Laboratory (PNL) has been tasked by the DOE's Office of Technology Development with adapting, developing, and demonstrating technologies for the measurement of uranium in surface and subsurface soils at the Fernald Uranium in Soils Integrated Demonstration site.

To accomplish the subsurface phase of this program, PNL employed a gamma-ray spectrometer adapted to fit down a 20-cm borehole to measure natural and contaminant uranium concentrations as a function of depth. The surface characterization phase of this program utilized two separate detection systems. The first system measured the gamma-emitting radionuclides over relatively large surface areas (hundreds of square meters), which therefore made it particularly useful in the preliminary screening of the site (i.e., for locating areas of elevated concentrations and providing for near-real-time mapping and generation of surface activity contours). The second surface monitoring system, designed and built by PNL, measured beta radiation emanating from smaller surface areas (0.15 m^2) to determine local surface uranium contamination levels before, during, and after actual remedial efforts.

During August and September of 1992, surveys were conducted at the Fernald's former Drum Baling Area (DBA) and the large field surrounding the Sewage Treatment/Incinerator (STI) site using the three surface and subsurface monitoring technologies. In both areas, soils were found to contain uranium-bearing compounds to various depths as a result of airborne deposition and direct contact with radioactive fluids and/or contaminated scrap items. Uranium contamination was found throughout the DBA (excluding the northernmost region), with activity levels exceeding 3000 pCi/g on the surface and penetrating to depths of 120 cm in at least one area. In the field surrounding the STI site, two broad regions exhibiting elevated uranium concentrations

were identified to the north and west, which evidently correspond to airborne deposition from the incinerator stack and drainage from the sewage treatment area along a natural depression, respectively.

Suggestions are made with regard to the remedial activities necessary in these areas as indicated by the results of this study, and improvements and design modifications to the demonstrated technologies are discussed in some detail.

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INTRODUCTION

The past operations of uranium production and support facilities at several Department of Energy (DOE) sites have occasionally resulted in the local contamination of some surface and subsurface soils. Such contamination commonly occurs within waste burial sites, cribs, pond bottom sediments, and areas surrounding waste tanks or uranium scrap, ore, tailing, and slag heaps. The thorough clean-up of these sites is a major public concern and a high priority for the DOE, but before any effective remedial protocols can be established, the three-dimensional distributions of the contaminants must be adequately characterized. Unfortunately, traditional means of obtaining soil activities (e.g., grab sampling followed by laboratory analyses) are cumbersome, expensive, time-consuming, and often non-representative when very large areas are under consideration. Hence, new technologies must be developed, or existing ones improved, to allow for the cheaper, better, faster, and safer characterization of contaminant concentrations at these critical sites.

The DOE's Fernald Environmental Management Project (FEMP) near Cincinnati, Ohio (previously known as the Feed Materials Production Center), is one specific site of concern. This facility, once the primary production site for uranium materials for defense-related projects, began operation in the early 1950s and was mainly responsible for the processing of uranium metal from concentrates. Production ceased in the late 1980s. Approximately 50% of the surface soils in and around the general production areas are known to be contaminated by various uranium-bearing compounds in excess of 35 pCi/g of soil as a result of normal operations and unintentional spills or emissions.

Pacific Northwest Laboratory (PNL) has been tasked by the DOE's Office of Technology Development with adapting, developing, and demonstrating technologies for the measurement of uranium in surface and subsurface soils at the FEMP Uranium in Soils Integrated Demonstration (U-Soils ID) site. To accomplish the subsurface phase of this program, PNL employed a gamma-ray spectrometer adapted to fit down a 20-cm borehole to measure natural and contaminant uranium concentrations as a function of depth. The surface characterization phase of this program utilized two separate detection systems. The first

system measured the gamma-emitting radionuclides over relatively large surface areas (hundreds of square meters), which therefore made it particularly useful in the preliminary screening of the site (i.e., for locating areas of elevated concentrations and providing for near-real-time mapping and generation of surface activity contours). The second surface monitoring system, designed and built by PNL, measured beta radiation emanating from smaller surface areas (0.15 m^2) to determine local surface uranium contamination levels before, during, and after actual remedial efforts.

The primary purpose of this report is to summarize the overall results of PNL's characterization effort at the FEMP site during FY 1992 and to suggest remedial actions based specifically on these results. Furthermore, it is also the intent of this document to outline the strengths and limitations of the aforementioned technologies in the hopes of facilitating future critical evaluations.

TECHNOLOGICAL THEORY AND METHODOLOGY

Intrinsic germanium (IG) diode gamma-ray spectroscopy has many advantages as an analytical tool for surface and subsurface environmental monitoring: its selective high resolution (enabling precise quantitative multi-radioisotopic characterization), its relatively easy transportability, the utilization of a simple and convenient power source (i.e., a small portable generator), and the capability of rapid data reduction to provide essentially real-time results. Limitations associated with this methodology are minor and include the need for liquid nitrogen temperatures for proper operation, -196°C (77K), and a minimum of 10 to 15 minutes for data acquisition at each sample location.

SUBSURFACE GAMMA-RAY DETECTOR

The general concept for subsurface radionuclide detection (Figure 1), as employed previously by PNL for measurements in contaminated soils (e.g., Brodzinski 1981, Brodzinski and Nielson 1980, Nielson et al. 1977), is based on the use of an IG diode shielded with tungsten collimators that limit the crystal's field of view to a narrow stratum, the thickness of which is determined by the choice of low-absorption spacers. The gamma radiation incident upon this detector is converted into electrical pulses with magnitudes that are directly proportional to the energies deposited by the individual gamma-rays (or, occasionally, the coincident combinations thereof). These energies are uniquely representative of specific radionuclides; e.g., 1.001 MeV for $^{234\text{m}}\text{Pa}$ (from the ^{238}U decay chain), 1.461 MeV for naturally occurring ^{40}K , etc. Hence, by examining the complete gamma-ray spectrum obtained from a particular locality one may identify the presence of such radionuclides and, when the system is properly calibrated, their absolute concentrations.

Overall, this detector unit is approximately 2 m long with a diameter of 7.5 cm (15 cm with collimator rings attached), and comprises a basal detector unit and a central cryogenic region capped by a preamplifier module with associated signal and power supply cables. The system is lowered from a

tripod into wells or boreholes, the latter of which are intended to be produced in such a manner that target substrates experience minimal disturbance, thereby precluding the inadvertent redistribution of subsurface activity. At FEMP, such borings were advanced through the use of a truck-mounted drill rig utilizing a 20-cm outside diameter (O.D.) hollow-stem auger, with an auger head of slightly larger diameter (22-cm O.D.) in order to minimize direct contact with the auger string and consequent smearing of the borehole walls. Each borehole was extended to a depth of approximately 3 m and accepted a 20-cm inside diameter Schedule 40 flush-threaded polyvinyl chloride (PVC) casing to maintain the opening and facilitate radiation screening. The diode, collimated to observe a 2.5-cm strata of soil, was lowered in 10-cm increments for the first 30 cm, followed by a measurement every 30 cm. Data collection was controlled by a dedicated electronics package, which included appropriate Nuclear Instrument Modules (NIMs) such as a detector bias supply, signal amplifier, and pulse height analyzer. A laptop-style personal computer, in conjunction with commercial and in-house software packages, was used to control data acquisition and data reduction in the field (e.g., Hensley et al. 1988).

The initial calibration of this system was performed at PNL's Natural Activities Calibration Facility (NACF). This facility consists of an undisturbed field within the Hanford site (containing a series of steel- and PVC-lined boreholes for downwell observation) which has been thoroughly characterized in terms of natural radionuclides (*viz.*, 14.8 pCi/g of ^{40}K , 0.6 pCi/g of ^{238}U , and 0.7 pCi/g of ^{232}Th). To generate calibration conversion factors, the detector response to these isotopes at the NACF was correlated with their known concentrations, and a linear relationship between count rate and soil activity was assumed. Although minor differences in soil composition and moisture content exist between NACF and FEMP, these should be insignificant with regard to gamma attenuation properties and were therefore ignored. Additional checks of the system's calibration and efficiency were performed at FEMP, in order to confirm the stability and proper operation of the detection unit throughout each working day. This consisted of the periodic monitoring of a "constant geometry" ^{232}Th -enriched standard.

SURFACE GAMMA-RAY DETECTOR

The extended surface area gamma monitor is composed of a down-looking IG diode/cryostat configuration and an electronics package equivalent to that described above for the downwell unit. When suspended 1 m above the soil, this uncollimated detector is sensitive to surface and shallow subsurface activity (e.g., to depths approaching 20 cm for uranium, directly below the unit) over many hundreds of square meters, effectively averaging any horizontal heterogeneities that may exist within this field of view. Although in theory this system would monitor complete 2π space (i.e., from horizon to horizon), it has been established empirically (Helfer and Miller 1988) that approximately 80-90% of the total observed fluence originates within a 10-m radius from the detector, for most gamma energies and soil distributions, due to geometry factors and attenuation in the soil and air.

With appropriate calibration, the concentration (in pCi/g of soil) or surface inventory (in pCi/m²) of the radionuclide(s) of concern can be determined for uniformly or exponentially distributed sources, respectively. The calibration procedure entails the empirical determination of the IG diode response to incident gamma radiation (both normal to the crystal face and for angles of incidence up to 90°), and also utilizes pre-tabulated data that represent the angular flux contributions from various potential soil distributions (see Beck et al. 1972). The combination of these data and the detector response parameters led to an energy-specific list of conversion factors for converting total counts per unit time into soil activity for each radionuclide of interest. Due to the request for "mass" activity units in this characterization exercise (as opposed to "spatial"), a uniformly distributed source was assumed at each sample location despite empirical evidence to the contrary (see below). Such an assumption simplifies the situation somewhat in that activity levels within the upper 15 cm were averaged through this depth.

Based on the sensor's effective field of view as discussed above, two sampling grid scenarios were established on 18.3-m centers for the purpose of analyzing the extent of surface coverage and determining appropriate sampling densities for future *in situ* gamma evaluations. A commercial survey system

was used to precisely locate each grid/sample point in the x - y plane (within 1-cm accuracy), while the results from the surface gamma spectrometry were substituted for the z (altitude) coordinate, thereby leading to the generation of a "pseudo-topography" or isopleth map of uranium concentrations within the surveyed area. Daily checks of the system's efficiency and stability were performed by counting a calibrated check source (^{241}Am , ^{137}Cs , and ^{60}Co) throughout the course of each working day, and data were reduced in the field utilizing an in-house, application-specific software package being developed by R.T. Reiman (EG&G, Rocky Flats).

SURFACE BETA DETECTOR

The surface beta-detection field prototype unit, designed and built at PNL, consists of multiple layers of 1 mm^2 plastic scintillating fibers that detect beta particles emitted from the decay of near-surface uranium (i.e., on the order of 3 to 5 mm depth in standard soils), as well as other natural or contaminant radionuclides that lead to the production of beta particles and/or secondary electrons. Specifically, the 2.29-MeV (maximum energy) beta particle from $^{234\text{m}}\text{Pa}$, a daughter of ^{238}U and in secular equilibrium therewith, was measured due to its relatively high energy and abundance.

This sensor is also somewhat sensitive to gamma rays, which may produce interfering signals. PNL's multi-layer design affords some discrimination against gamma emission by observing, and partially cancelling the contribution from, this ubiquitous phenomenon. Such discrimination is possible since a beta particle (or other charged particle) will cause excitations and consequent scintillations along its entire path (see Figure 2), while a gamma ray might interact with only one (indeed, if any) of the individual layers, thereby generating a secondary Compton or photoelectron which may then continue as above. Hence, by establishing an electronic requisite for coincident signals from each layer (i.e., a triple-coincidence with the existing package), one should detect primarily beta-related events and effectively ignore much of the gamma contribution to the overall detector response. Of course, a secondary electron produced in the first layer (and subsequently traversing the second

and third layers) could lead to a gamma-generated triple coincidence, but such "false" events are expected to be relatively rare under most circumstances.

The field detector unit is approximately 1.5 m long, 15 cm wide and 8 cm tall, and incorporates a large (1500 cm²) beta-sensitive region that measures about 1 m long and extends to the full width of the unit. A 10-m power supply/signal transmission cable connects the detector unit to a rack-mounted signal processing module, the outputs from which are fed to a series of NIM scalers. These scaler data indicate the total number of counts in each layer as well as the number of associated interlayer coincidences. The detector was set directly on the ground (grassy areas were cropped before surface characterization), and a number of consecutive counts were obtained at each sample/grid location. Calibration of this system consisted of monitoring a surrogate soil sample (*viz.*, a medium-grained river sand) that was spiked to 35 pCi/g of ²³⁸U, equating its response above background to this activity level, and assuming system linearity over the expected concentration range at the FEMP site. In order to ensure system stability and optimal operation, numerous background and calibration counts were performed throughout each working day.

FIGURE 1. PNL's downwell detector unit, showing details of the layer collimator assembly.

FIGURE 2. Conceptual design and operation of PNL's surface soil beta monitor showing approximate penetration depths of various incident radiations. The "perforated" plots for the photons are intended to convey the potential for no gamma reaction within the scintillator stack. The existing sensor is comprised of three 1-mm layers.

RESULTS AND DISCUSSION

During August and September of 1992, surveys were conducted at the former Drum Baling Area (DBA) and the large field surrounding the Sewage Treatment/Incinerator (STI) site using the three surface and subsurface monitoring technologies. The latter of these sites is located on the eastern boundary of the FEMP facility and was long used for the treatment of process-related waste waters and the burning of contaminated solid wastes. The former DBA is situated in the northeast corner of the original Production Area and served as a staging area for clean and contaminated scrap materials. Both areas exhibit soils that contain uranium-bearing compounds to various depths as a result of airborne deposition and direct contact with radioactive fluids and/or contaminated scrap items. Figures 3 and 4 indicate the general boundaries for these sites, as well as the individual sample points and boreholes utilized in the U-Soils ID.

Tables 1 and 2 display the beta and/or *in situ* gamma results for the former DBA and STI site, respectively. All quoted standard deviations are one sigma (1σ) and were determined from Poisson statistics and error propagation for the beta detector, or from peak shape information with regard to the gamma spectrometry.^(a) One sigma and upper limit values for the downwell gamma results are listed in Table 3. In each case, the resulting errors are acceptable but could be further reduced by increasing the actual counting times, establishing additional means for the minimization of background and interference contributions, and enhancing detector efficiency (all of which are currently under consideration for the next generation of these detection systems).

Most of the boreholes surveyed at the DBA exhibited "exponential-like" activity distributions versus depth (see Figures 5 through 9), with contamination levels in excess of 35 pCi/g approaching depths of 30 cm. Only Wells

(a) For a gaussian-like peak, which is normally encountered in this technique, the 1σ range is approximately 43% of the peak's full width at half maximum value.

C and E (refer to Figure 3 for borehole locations) were observed to have deeper uranium contamination; i.e., near 120 cm and 60 cm, respectively. Consequently, it is recommended that removal activities initially target a minimum of 30 cm of soil (at least along the north-south transect delineated by Wells A through E) and be followed by surface re-characterization in order to identify those regions in which contamination from greater depths yet remains.^(a)

Subsurface uranium levels in the field surrounding the STI site (see Figures 4 and 10-12) are observed to be lower than 35 pCi/g, and are assumed to be of natural origin. An exception exists at Well Z (situated within a topographic depression) where excess contamination may persist to a depth of approximately 10 cm. The observed elevated uranium content is suspected to have originated from the STI site itself, as a result of natural drainage to the west along this depression. Hence, remedial activity would appear to be necessary within the boundaries of this topographic feature, but only to a depth of 15 cm or less. These observations from the downwell gamma survey are reinforced by the results from the surface beta characterization of this area (Figure 13), which also identified a distinct west-trending lobe of activity associated with this same depression.^(b) Uranium contamination increases to the east within this run-off channel (i.e., toward the STI site) and maximum levels are found to exceed 50 pCi/g. In addition to this feature, another large lobe of uranium contamination extends to the north-northeast and likely resulted from airborne deposition of contaminated smoke and ash particles from the incinerator stack (located in the northwest corner of the STI site). Prevailing southwesterly winds would seem to support this hypothesis. Uranium activity in excess of 40 pCi/g was observed in this region.

-
- (a) These and other suggestions are based solely on the observations made during PNL's characterization efforts, with no *a priori* information regarding excavation capabilities, total cost of soil removal/treatment, remedial regulations, etc.
- (b) All contour plots were generated with SURFER (version 4) by Golden Software, Inc. and assumed an inverse distance squared relationship between sample points for the generation of intersample values.

Both beta and *in situ* gamma analyses were performed in the more highly and heterogeneously contaminated DBA. A tighter sampling grid (essentially, two offset 18.3-m grids; see Figure 3) was utilized for this phase of the characterization to evaluate any effect due to changing initial sample density. Figures 14 and 15 summarize the data obtained from the beta detection and *in situ* gamma spectrometry systems, respectively, and are based on the highest available sampling density. Qualitatively, the beta and gamma results correlate quite well in terms of "hotspot" identification despite the extreme differences in the fields of view and observation depths associated with these inherently disparate methodologies. It is evident from these results that, with the exception of an isolated region in the northernmost portion of the area, virtually all of the DBA is contaminated with uranium at levels exceeding 35 pCi/g. In addition, four distinct areas of elevated activity were detected via the gamma and beta measurements; specifically: near the southern boundary of the test area, at the termination of the southern railroad tracks (possibly a convenient dumping station), in the center of the DBA between these tracks and the gravel road, and at the northwestern edge of the site (which borders on the current decontamination area).

The quantitative results generated by these two techniques, however, are rarely found to be in actual agreement. Indeed, such agreement would occur only in the case of a vertically (and horizontally) uniform uranium distribution. Any distribution that decreases rapidly with depth should be manifested by a beta response which somewhat exceeds that of the *in situ* gamma spectrometer "expecting" a uniform distribution, since the beta detection unit interrogates to depths of only 3 to 5 mm while the IG crystal obtains information from depths often exceeding 15 cm and essentially averages the overall gamma activity throughout this depth. Of course, *a priori* knowledge of the vertical distribution at each sample point could be provided as input to the *in situ* gamma data reduction program which would then generate a response in terms of activity per unit surface area,^(a) yet the spatial variability in depth

(a) Activity per unit weight of soil is not a truly representative unit of measurement for a source which is heterogeneously distributed with depth, except perhaps in the limit of an infinitesimally thin stratum at a strictly specified depth.

distri-butions exhibited in this area (see Figures 5-9) precludes this as a cost-effective option (i.e., a borehole at each sample site).

As an exercise, two new gamma contour plots for the DBA were generated following a reduction of the number of actual sample points in order to investigate the effects of sample density on overall data integrity.^(a) Figures 16 and 17 represent the results associated with excluding one (or the other) of the original pair of offset sample grids, thereby roughly halving the total number of data points for each iteration and increasing the intersample distance to a full 18.3 m. As expected, the finer details that were apparent in the initial interpretation (Figure 15) are lost and the hotspot locations are often shifted slightly by a few meters. This, of course, is an artifact of the IG diode's true field of view, which is operationally limited to a 10-m radius (approximately). In a first-order screening of the DBA, either of the "half-sample" iterations would seem to provide enough detail for characterization of the major features and allow the direction of reasonable remediation efforts. An even wider sample grid, however, would probably not have provided sufficient detail for this purpose (and certainly would not have afforded saturation coverage), due to the limited size of the site and its considerable heterogeneity. Although a tighter sampling grid was not investigated here, it is suspected that the additional information gained would not have been worth the consequent expenditures of time and money.

These results indicate that remediation would be best served if the top 15 cm of soil were removed from the DBA, with the exception of the broad lobe in the northern reaches of the area. Such removal should be followed immediately by further *in situ* gamma spectrometry and surface beta monitoring to determine those regions in which deeper activity remains, and then additional

(a) This exercise was not extended to the β results due to the highly collimated nature of this detection system (i.e., no intersample redundancy exists, unlike the partially overlapping areas of observation in the case of the *in situ* gamma methodology), and truly thorough coverage with this unit would require a continuous sweep of the total area of interest.

soil removed as necessary. This effective combination should be repeated as required until remediation is complete.

FIGURE 3. Map view of the DBA indicating borehole and sample point grid locations. Note that two offset grids (each equivalent to that utilized at the STI site) were used here to test the effects of sample density. Flag A1 is located in the southeast corner and flag designations increase alphabetically and numerically to the north and west, respectively.

FIGURE 4. Map view of the large field surrounding the STI site indicating borehole and sample point grid locations. The small blank area west of the STI site represents a wet, marshy region that was not accessible to the surface beta monitor. Flag A1 is located in the southwest corner and flag designations increase alphabetically and numerically to the north and east, respectively.

FIGURE 5. Vertical uranium distribution in Well A of the DBA. The lower activity on the surface may be an artifact of the partial cave-in experienced during the construction of this first well. Solid symbols represent upper limit values and open symbols indicate values with 1σ standard deviations (for clarity, error bars were not included). The dotted line indicates 35 pCi/g.

FIGURE 6. Vertical uranium distribution in Well B of the DBA (see descriptive notes from Figure 5).

FIGURE 7. Vertical uranium distribution in Well C of the DBA (see descriptive notes from Figure 5). Erratic behavior may be an operational artifact (e.g., inadequate ground penetration) or could indicate the presence of a subsurface impermeable layer or shield material (e.g., clay stratum, large rock, etc.).

FIGURE 8. Vertical uranium distribution in Well D of the DBA (see descriptive notes from Figure 5).

FIGURE 9. Vertical uranium distribution in Well E of the DBA (see descriptive notes from Figure 5).

FIGURE 10. Vertical uranium distribution in Well X in the field surrounding the STI site (see descriptive notes from Figure 5).

FIGURE 11. Vertical uranium distribution in Well Y in the field surrounding the STI site (see descriptive notes from Figure 5).

FIGURE 12. Vertical uranium distribution in Well Z in the field surrounding the STI site (see descriptive notes from Figure 5).

FIGURE 13. Contour plot of activities at the STI site based on PNL's beta monitor (contour interval = 5 pCi/g). Hachure lines indicate decreasing uranium levels in the direction of the lines.

FIGURE 14. Contour plot of activities at the DBA based on PNL's beta monitor. Contour intervals (excluding the 100 pCi/g delineation) are 200 pCi/g (≤ 1000 pCi/g) and 500 pCi/g (≥ 1000 pCi/g).

FIGURE 15. Contour plot of activities at the DBA based on the *in situ* gamma spectrometer. Contour interval (excluding the 35 pCi/g delineation) = 50 pCi/g.

FIGURE 16. Same as Figure 15, but based on *in situ* gamma data from rows A, C, E, etc., only.

FIGURE 17. Same as Figure 15, but based on *in situ* gamma data from rows B, D, F, etc., only.

TABLE 1. Data summary for beta and *in situ* gamma results at the DBA based on 500-second and 10-minute counts, respectively. Flag locations are identified in Figure 3.

DBA flag	gamma (pCi/g)	1 sigma	error (%)	gamma (ppm)	beta (pCi/g)	1 sigma	error (%)	beta (ppm)
	U-238	std. dev.		total U	U-238	std. dev.		total U
A1	112	11	10	335	147	10	7	441
C1	210	13	6	629	1019	66	6	3054
C2	566	17	3	1697	2817	182	6	8440
C3	168	8	5	503	877	57	6	2627
E1	161	13	8	482	405	26	6	1214
E2	266	13	5	796	767	50	6	2299
E3	161	8	5	482	396	26	6	1186
E4	27	5	18	82	46	4	8	138
G1	105	6	6	314	315	21	7	943
G2	70	7	10	210	214	14	7	640
G3	119	7	6	356	373	24	7	1118
G4	31	5	16	94	112	8	7	336
I1	91	5	6	272	306	20	7	917
I2	301	9	3	901	1106	71	6	3313
I3	154	8	5	461	350	23	7	1048
I4	63	6	9	189	163	11	7	489
K1	119	7	6	356	391	25	6	1172
K2	413	12	3	1236	539	35	6	1614
K3	217	9	4	649	458	30	6	1372
K4	126	6	5	377	458	30	6	1373
K5	105	6	6	314	417	27	6	1250
M1	129	8	6	388	550	36	6	1646
M2	270	11	4	809	1233	80	6	3693
M3	70	6	9	210	8	2	29	23
M4	196	10	5	587	873	56	6	2616
M5	78	8	10	235	435	28	6	1304
M6	18	5	28	54	32	3	10	96
M7	210	8	4	629	704	46	6	2110
O1	161	8	5	482	214	14	7	643
O2	119	10	8	356	209	14	7	626
O3	76	6	8	228	233	15	7	699
O4	52	6	12	157	178	12	7	535
O5	8	1	16	23	63	5	7	190
O6	23	4	16	69				
O7	168	8	5	503	713	46	6	2136
O8	133	7	5	398	1902	123	6	5698
Q1	15	3	21	44	51	4	8	154
Q2	25	4	14	75	89	6	7	267
Q3	24	4	16	73	87	6	7	259
Q4	5	1	22	15	48	4	8	144
Q5	3	1	31	10	79	6	7	237
Q6	3	1	28	10	38	3	9	115
Q7	18	4	20	54	46	4	8	137
Q8	22	4	16	67	106	7	7	317

TABLE 1. (contd)

DBA flag	gamma (pCi/g)	1 sigma	error (%)	gamma (ppm)	beta (pCi/g)	1 sigma	error (%)	beta (ppm)
	U-238	std. dev.		total U	U-238	std. dev.		total U
B1	210	13	6	629	978	63	6	2929
B2	98	12	12	293	403	28	6	1207
B3	33	8	25	98	108	7	7	325
D1	343	14	4	1027	1440	93	6	4315
D2	601	18	3	1802	3555	229	6	10650
D3	64	8	9	191	154	10	7	462
F1	112	11	10	335	212	14	7	634
F2	133	8	6	398	465	30	6	1394
F3	54	5	10	161	178	12	7	533
H1	175	9	5	524	453	29	6	1359
H2	91	6	7	272	245	16	7	733
H3	91	5	6	272	253	17	7	758
J1	168	8	5	503	1219	79	6	3652
J2	112	8	7	335	575	37	6	1724
J3	138	7	5	413	507	33	6	1518
J4	44	5	12	132	317	21	7	949
L1	308	12	4	922	1942	125	6	5819
L2	105	7	7	314	49	4	8	145
L3	182	7	4	545	1361	88	6	4079
L4	252	10	4	754	2092	135	6	6269
L5	186	9	5	557	1631	105	6	4886
N1	161	8	5	482	502	33	6	1503
N2	182	7	4	545	817	53	6	2448
N3	49	5	11	147	139	9	7	418
N4	5	2	32	15	69	5	7	208
N5	13	4	35	38	153	10	7	458
N6	126	8	6	377	594	38	6	1778
N7	252	10	4	754	1048	68	6	3139
P1	38	5	12	113	103	7	7	308
P2	15	3	22	46	59	4	8	178
P3	15	3	22	44	32	3	9	97
P4	3	1	36	8				
P5	3	1	35	8	6	2	34	19
P6	2	1	48	6	6	2	34	19
P7	17	3	20	50	31	3	10	93

TABLE 2. Data summary for beta results at the field surrounding the STI site based on 500-second counts. Flag locations are identified in Figure 4.

STI flag	beta (pCi/g)	1 sigma	error (%)	beta (ppm)
	U-238	std. dev.		total U
A1	14	1	7	41
A2	16	1	7	48
A3	20	1	7	60
A4	30	2	7	91
A5	17	1	7	52
A6	14	1	7	42
A7	11	1	8	32
A8	18	1	7	55
A9	18	1	7	53
A10	15	1	7	44
A11	10	1	8	30
C1	17	1	7	51
C2	20	1	7	60
C3	18	1	7	55
C4	19	1	7	56
C5	20	1	7	60
C6	15	1	7	45
C7	32	2	7	95
C8	23	2	7	69
C9	19	1	7	56
C10	23	2	7	68
C11	10	1	8	30
E1	39	3	7	117
E2	27	2	7	82
E3	21	1	7	63
E4	28	2	7	83
E5	2	0	13	4
E6	33	2	7	97
E7	16	1	7	49
E8	17	1	7	52
E9	15	1	7	44
E10	24	2	7	73
E11	11	1	8	34
G1	18	1	7	54
G2	35	2	7	106
G3	26	2	7	79
G4	27	2	7	81
I1	24	2	7	71
I2	30	2	7	90
I3	25	2	7	74
I4	29	2	7	87
K1	30	2	7	91
K2	39	3	7	118
K3	33	2	7	100
K4	31	2	7	92
M1	28	2	7	83
M2	39	3	7	116
M3	35	2	7	104
M4	47	3	7	142

TABLE 2. (contd)

STI flag	beta (pCi/g)	1 sigma	error (%)	beta (ppm)
	U-238	std. dev.		total U
Q1	42	3	7	127
Q4	53	4	7	159
Q4	30	2	7	89
S1	19	1	7	58
S2	10	1	8	29
S3	20	1	7	60
S4	19	1	7	56
S5	27	2	7	81
S6	20	1	7	61
S7	38	3	7	113
S8	33	2	7	100
S9	32	2	7	95
S10	30	2	7	91
S11	11	1	8	34
U1	22	2	7	66
U2	22	2	7	66
U3	30	2	7	91
U4	17	1	7	50
U5	27	2	7	81
U6	22	2	7	66
U7	49	3	7	146
U8	31	2	7	94
U9	30	2	7	90
U10	22	2	7	65
U11	18	1	7	55
W1	15	1	7	45
W2	18	1	7	53
W3	33	2	7	98
W4	29	2	7	86
W5	17	1	7	50
W6	22	2	7	65
W7	27	2	7	81
W8	23	2	7	68
W9	23	2	7	68
W10	24	2	7	70
W11	23	2	7	69
Y1	14	1	7	41
Y2	23	2	7	69
Y3	27	2	7	82
Y4	26	2	7	79
Y5	24	2	7	72
Y6	17	1	7	52
Y7	22	2	7	66
Y8	23	2	7	68
Y9	20	1	7	59
Y10	27	2	7	80
Y11	20	1	7	61

TABLE 3. Downwell results from the DBA and STI site in pCi/g of ^{238}U based on 10-minute counts. Parenthetical values represent 1 σ errors and "less than" values indicate upper limits (3 σ). Borehole locations are identified in Figures 3 and 4. The "0 cm" depth represents the situation in which the lower edge of the top collimator is in line with the ground surface.

depth (cm)	Well A	Well B	Well C	Well D	Well E	Well X	Well Y	Well Z
0	57(15)	454(24)	65(10)	86(12)	761(38)	26(8)	<27	53(9)
10	155(27)	73(12)	129(15)	60(11)	548(27)	<29	<26	<30
20	112(22)	<15	48(12)	40(10)	273(23)	22(8)	<26	<25
30	<28	32(8)	86(12)	33(10)	143(14)	<24	<27	<28
41	<41							
51	<38							
61	<39	26(8)	41(10)	<30	35(10)	<27	<28	<29
91	<32	15(8)	42(11)	<29	27(10)	<29	<28	.28
122	<35							
152	<31	<28	<28	<23	<26	<27	<30	<25
183	<34							
213			<25	17(7)	<25	<25	<28	<27
244								
274					<25		<25	<27
290								
295						<29		
305		22(8)		<28				

FUTURE APPLICATIONS

The technology associated with subsurface gamma monitoring is relatively mature at this stage. The overall utility of this system may be enhanced by the incorporation of a larger, more efficient IG crystal that would decrease total counting times and allow faster site coverage (although larger boreholes may be required, in this case). Conversely, the construction of a much smaller diameter IG crystal (e.g., on the order of 2 to 3 cm) may be advantageous in conjunction with the DOE's cone penetrometer tool for observing underground activity without concomitant radioactive soil removal. Such a methodology would allow characterization of the subsurface while minimizing the volume of secondary waste generated during site characterization. Furthermore, the incorporation of downwell neutron activation analysis and/or x-ray fluorescence excitation and detection equipment within this package could lead to an extremely powerful and versatile subsurface screening tool.

State-of-the-art *in situ* gamma spectrometry for surface soil characterization relies on bulky power supplies, cumbersome cable spans, and local computer terminals that are mechanically awkward to maneuver and require a trained spectroscopist at each unit. Such equipment could be streamlined and miniaturized in order to limit the amount of hardware to be moved in the field (and exposed to potentially contaminated soils), while removing the scientist from the time-consuming and redundant acquisition phase. By utilizing a pre-programmed, compact, and light-weight acquisition system designed to be moved and initialized with single-button control, efficient and more cost-effective coverage of larger surface areas would be possible. Data could be downloaded via removable and easily transported memory cards and reduced by a single scientist as the monitoring proceeds. Such a unit could be utilized at all commercial and government facilities that require characterization before and after remediation. This universal, dedicated data collection and processing package would meet all requirements for the *in situ* uranium characterization at Fernald and would be suitable for gamma spectrometric studies of other facilities as well.

Furthermore, by comparing the intensity ratios of high and low energy photons from the ^{238}U decay chain with those ratios expected from an unattenuated source, one may derive a first-order indication of the uranium distribution within the upper 15 cm of soil without the need for well boring.^(a) The photons of choice for this procedure are the 1001-keV and 766-keV gamma rays from $^{234\text{m}}\text{Pa}$ and the 63-keV and 93-keV x-rays from ^{234}Th (both radionuclides originate from the decay of ^{238}U and are in secular equilibrium therewith). Since lower energy photons are more efficiently attenuated than those of higher energy, the actual intensities of the aforementioned gamma- and x-rays may be directly compared and a determination made as to whether a source re-sides on the soil surface or is buried to some degree. The extent of this distribution with depth can be approximated with an accuracy that is suitable for establishing appropriate remedial activities.

PNL is actively investigating means whereby *in situ* gamma technology may be further improved by incorporating such features as the utilization of larger, higher efficiency diodes to reduce counting times and improve minimum detectable activities, the "bundling" of individual detector units into an integral package to produce a super-efficient multi-diode for extremely fast and accurate surface sample characterization, the mounting of such systems on dedicated, direct- or remote-controlled vehicles to enhance their real-time capability and minimize personnel exposure in hazardous areas, and the modification of existing hardware and data collection/reduction software to make it more accessible to the general operator and facilitate cheaper, faster, better, and safer site characterization.

The plastic scintillation field beta-monitor is still in the prototype stage at PNL, and a number of unique design options are currently undergoing critical evaluations. Future efforts will concentrate on those modifications which will be necessary for the reduction of background interferences (pri-

(a) This has been recently established by Mr. Kevin Miller of the Environmental Measurements Laboratory (New York) and is based on *in situ* gamma analyses performed in conjunction with PNL at the Fernald Site (abstract submitted to the 38th Annual Meeting of the Health Physics Society, July 11-15, 1993).

marily due to gamma-rays and cosmic-induced particles), thereby enhancing sensor precision, decreasing total counting times, and improving the minimum detectable beta activity. To this end, spectroscopic investigations involving the existing system are underway and a number of options are being considered for the next generation of the sensor, such as substituting thinner layers to minimize gamma sensitivity and consequent background, defining electronic discriminator settings for individual layers to eliminate gamma-generated Compton and photoelectron pulses, and adding an additional top layer(s) for anti-coincidence discrimination against cosmic background. The future version of this system will rely solely on battery power and be scaled-down so that it may be manipulated easily by a single operator.

CONCLUSIONS

Three original and/or adapted technologies were introduced that, when utilized concurrently or separately, have the capabilities to significantly outperform current methods for determining uranium concentrations in surface and subsurface soils at the levels projected. The singular or combined use of collimated downwell gamma spectrometry, surface gamma spectrometry, and beta scintillation detection can provide for the precise and accurate three-dimensional uranium characterization of virtually any contaminated or potentially contaminated site, barring personnel inaccessibility. Such techniques exhibit vast improvements over, and largely eliminate the need for, the collection of myriad soil samples and subsequent expensive and time-consuming laboratory analyses.

The results of this study have indicated to some degree the extent of remedial activity required at the DBA and STI site. In the former, uranium contamination exists throughout (excluding the northern boundary), with activity levels exceeding 3000 pCi/g on the surface and penetrating to depths of 120 cm in at least one area. In the field surrounding the STI site, two broad regions exhibiting elevated uranium concentrations were identified to the north and west which evidently correspond to airborne deposition from the incinerator stack and drainage from the sewage treatment area along a natural depression, respectively. Uranium contamination may exist to a depth of at least 10 cm in this topographic feature. Suggestions were made as to the amount of soil that might be removed from these areas based on the aforementioned results, but such recommendations should be taken *cum grano salis* without additional input regarding the costs associated with excavation and treatment, regulatory restrictions, soil removal limitations, etc. A more economical scenario could incorporate soil removal from those areas identified above, but to depths of only 5-10 cm (or less), followed by further *in situ* gamma and surface beta analyses. Such a combination would minimize the volume of waste generated, thereby reducing expenditures in terms of excavation, treatment, and shipping, and ensure thorough remediation of these sites to the levels intended.

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