Recovery of Depleted Uranium Fragments from Soil - 8206

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ABSTRACT

A cost-effective method was demonstrated for recovering depleted uranium (DU) fragments from soil. A compacted clean soil pad was prepared adjacent to a pile of soil containing DU fragments. Soil from the contaminated pile was placed on the pad in three-inch lifts using conventional construction equipment. Each lift was scanned with an automatic scanning system consisting of an array of radiation detectors coupled to a detector positioning system. The data were downloaded into ArcGIS for data presentation. Areas of the pad exhibiting scaler counts above the decision level were identified as likely locations of DU fragments. The coordinates of these locations were downloaded into a PDA that was wirelessly connected to the positioning system. The PDA guided technicians to the locations where handheld trowels and shovels were used to remove the fragments. After DU removal, the affected areas were re-scanned and the new data patched into the data base to replace the original data. This new data set along with soil sample results served as final status survey data.

INTRODUCTION

A “proof of concept” study was performed at Sandia National Laboratories to evaluate a method for recovering depleted uranium (DU) fragments from soils. The soils contained DU fragments as a result of impact tests involving DU at the Sandia Long Sled Track test facility. Approximately 5,000 cubic meters of affected soils had been consolidated into three piles several years ago and have been managed as a contaminated soil area since.

Prior to performing the study, laboratory measurements were made using various radiation detectors and DU fragments as radiation sources. Detector intrinsic efficiencies were measured under static conditions with the sources buried beneath various layers of local soil. This data served as a basis for detector selection and for determining the maximum thickness of the soil for each layer.

The method involves scanning a thin layer of soil with an array of radiation detectors whose location is precisely known. The locations of each detector and the associated radiation count rates were automatically recorded. Count rates above the decision level were further investigated by downloading the coordinates of the anomalies into a handheld PDA, navigating back to the locations, and manually using a handheld trowel to remove the DU fragment. After removal of the fragment, only the immediate remediated area was rescanned with the new data replacing the original data.

DETECTORS

DU fragments from another site, ranging from less than 1 gram up to 48.5 grams, were initially used to assess the feasibility of the method. A Ludlum Measurements, Inc Model 44-10 (LMI 44-10), a LMI Model 44-20 and an Alpha Spectra, Inc. Field Instrument for Detecting Low Energy Radiation (FIDLER) detector were evaluated. The LMI instruments are encased in aluminum housings with no entrance window. The LMI 44-10 uses a 5.1-cm by 5.1-cm (2-inch by 2-inch) NaI crystal and the LMI 44-20 uses a 7.6-cm by 7.6-cm (3-inch by 3-inch) NaI crystal. The FIDLER detector consists of a 0.160 cm (0.063 inch) thick Sodium Iodide (NaI) detector and a 0.025 cm (0.010 inch) thick beryllium entrance window.

The operational parameters of the detectors were first established by employing an URSA-II multichannel analyzer (MCA) to look at the energy spectrum of the x-ray and gamma-ray emissions of DU fragments. Figures 1 and 2 show spectra collected using the LMI 44-10 and the FIDLER, respectively. The 63 keV and 92-keV gamma rays from the decay of the uranium-238 progeny are shown. The evidence of detector housing attenuation of the lower energy gamma rays and the relatively high continuum beneath the gamma-ray photopeaks are pronounced for the LMI 44-10 detector relative to that in the FIDLER detector spectrum. These spectra were also used to assure that the meter high voltage and threshold were set to assure detection of the 63 keV gamma ray of interest.

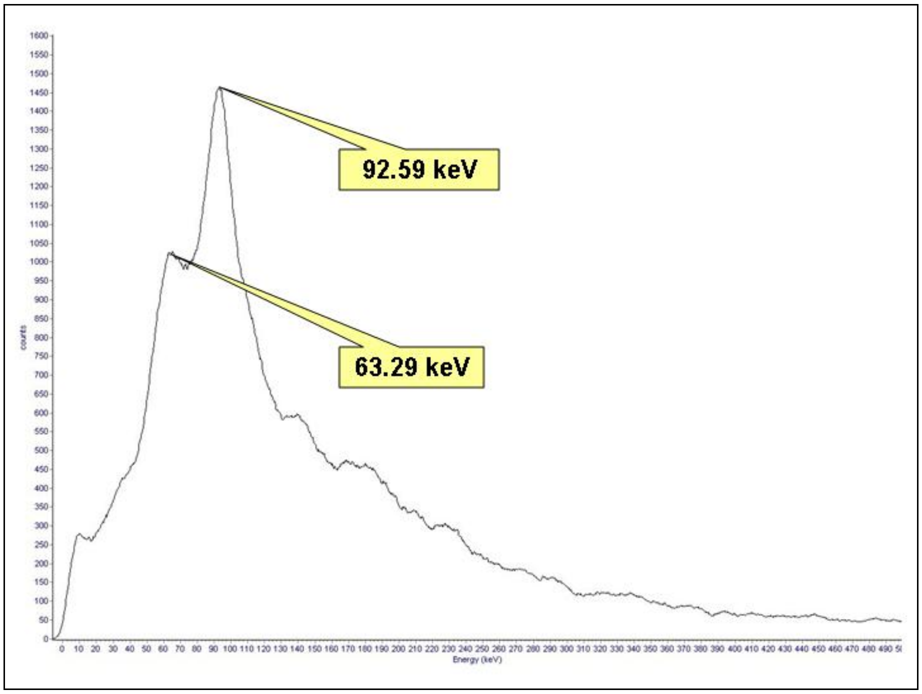

Fig. 1. DU energy spectrum collected using the Ludlum Model 44-10 5.1 cm x 5.1 cm (2-inch x 2-inch) NaI Detector.

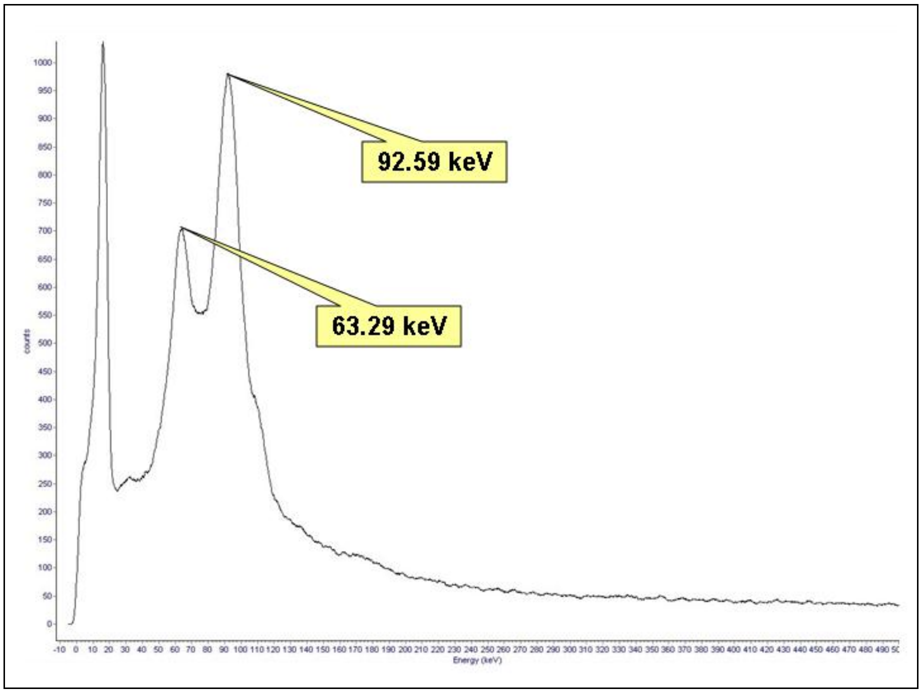

Fig. 2. DU energy spectrum collected using the Alpha Spectra 12.7 cm (5-inch) diameter FIDLER detector.

A Ludlum Model 2360 scaler was used to power each detector type for the intrinsic efficiency measurements. The measurements were made with each DU fragment placed at three depths within uncompacted local soils and the detector located directly above. The Model 2360 threshold was set to include all photons of approximately 20 keV or higher. Table I presents a summary of the results for the three detectors. The net counts per minute per gram of DU is significantly higher for the FIDLER than for the LMI 44-10 and LMI 44-20 detectors. When one considers the relative background count rates shown in Table I, the FIDLER is much superior to the other detectors.

Table I. Detector Performance Characteristics

Detector	Net CPM/gram			Background Count Rate
	Source Depth			
	Surface	2.5 cm	7.6 cm	
LMI 44-10	891	455	227	12,906
LMI 44-20	2923	621	-66	37,075
FIDLER	7,717	2,000	1,292	15,192

The complete set of data for the FIDLER is given in Table II where the background is subtracted from the count rates and the net count rates are normalized by dividing by the mass of the particle. Note the response from the very small particles and the largest fragments differ from the others. The smallest fragments could not be weighed accurately on our scale that reported to

0.1 g. The response to the largest fragment was very low, indicating that the size was large enough for self attenuation of the gamma rays. Therefore the average net counts per minute per gram of DU was calculated without the data from the three smallest fragments and the largest fragment used in the study. The average net counts per minute per gram of DU was measured to be 9329, 2,000, and 1293 when the source was buried beneath 0 cm, 2.5 cm (1 inch), and 7.6 cm (3 inches) of local soils. This corresponds to 466, 100, and 65 counts per 3-second interval. While this study did not evaluate how the physical properties of the fragments affect the normalized net count rate, during fragment removal it was observed that particle shape and perhaps the degree of oxidation affects the gamma emission rates of the particle per unit mass.

Table II. Intrinsic Efficiency of the FIDLER Detector for DU Sources

Fragment Number	Weight (grams)	Height of Detector Above Ground Surface					
		10.2 cm			45 cm		
		Net CPM/gram					
		Source Depth			Source Depth		
		Surface	2.5 cm	7.6 cm	Surface	2.5 cm	7.6 cm
1	0 [a]	2,200	-	-	-11,100	-	-
2	0.4	9,783	2,118	-213	-1,295	-1,150	-1,663
3	1.0	7,620	1,428	411	170	-100	-382
4	2.0	10,510	1,892	1,293	410	-1	-49
5	3.1	8,531	1,897	1,259	489	134	29
6	4.0	9,575	2,111	1,395	477	207	47
7	4.9	10,120	2,328	1,238	564	174	139
8	10.0	9,690	1,796	1,274	453	221	142
9	15.0	7,549	1,974	1,296	386	213	139
10	48.5	1,483	536	523	92	81	45
Average [b]		9,329	2,000	1,292	463	158	75

[a] Weight assumed to be 0.01 grams for calculation purposes.

[b] Average is for fragments between 2.0 and 15.0 grams only.

SCANNING SYSTEM

The ERG Three-Dimensional Indoor Survey System (3-DISS) [1,2] was modified for this project by mounting the instrumentation on a trailer pulled behind a small farm tractor. The system automatically records scaler count data along with associated detector coordinates at user-defined time intervals. The positioning component of the system uses infrared fan lasers with sensors located on the trailer to determine the coordinates of each detector at a rate of up to 20 positions per second. At the end of each counting interval the scaler count and position of each detector is recorded in a simple text file format. The accuracy of the coordinates under field conditions was estimated at 5 cm. An eight-detector array of FIDLER detectors was mounted on the trailer at a height of 15 cm above the ground. The distance between the detectors was 7.6 cm

(3 inches), making the total scanning width approximately 155 cm. Figure 3 shows the system being pulled by the John Deere 4010 tractor.

Fig. 3. Eight detector array and associated electronics pulled by tractor.

The John Deere 4010 tractor has a hydrostatic transmission allowing the trailer to be pulled at a rate of 10 cm per second. Normally a long scaler counting interval is desirable in order to reduce the statistical fluctuations involved in making radiation measurements. However, a long counting interval reduces the sensitivity when making a moving measurement over a fixed point source since it averages the counts of the point source with counts from no source and it also degrades the spatial resolution of the system for locating the fragments. After these considerations, a scaler counting interval of 3-seconds was chosen. This was found to define the location of anomalies to within 30 cm.

The average net result for detectors placed close to the ground surface naturally proved to be far superior for point sources and thus the detectors were placed as close to the ground surface as practical. The 3-DISS trailer was designed for adjustable detector heights. Because of site conditions, a 15-cm height above the soil surface was chosen to protect the detectors from damage due to irregular surfaces.

OPERATIONS

A dirt pad of compacted clean soil was prepared adjacent to the contaminated soil pile. Multiple 7.6 cm (3-inch) lifts of soil, taken from the contaminated pile, were applied to the pad using conventional construction equipment. Each lift was scanned using the modified ERG 3-DISS, remediated, and then rescanned prior to a new lift being applied. A total of eighteen lifts were applied to the pad which averaged approximately 2800 square meters (0.7 acre) in size.

A decision level was established above the normal variation of background radiation levels. For each lift survey data were collected on a laptop and viewed in real time using proprietary ERG software. An example of the initial survey data and post remediation survey data of a complete lift is shown in Figure 4. For further analysis of each lift the data were imported into ESRI ArcGIS, a computer application for data processing and presentation. Count rates above the decision level were further investigated by downloading the coordinates of each anomaly into a handheld PDA and navigating back to the coordinates. A handheld FIDLER detector was then used to further locate the fragment. A hand trowel or shovel was used to remove the DU fragment. After removal of the fragment, only the immediate remediated area was rescanned with the new data replacing the original data. The new data set was used as final status survey data. Approximately 250,000 data records per hectare (2.5 acres) were generated for each 7.6 cm lift.

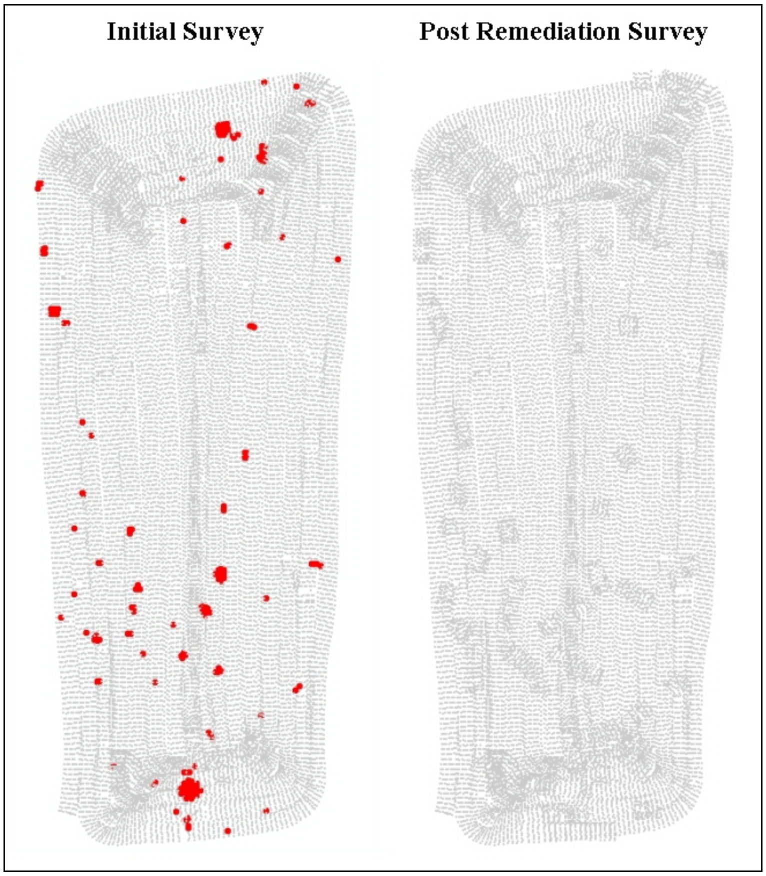

Fig. 4. Comparison between an initial survey and post-remediation survey of the same lift.

In all but a very few cases the anomalies were found to be fragments ranging in size from less than a gram to a few hundred grams and were easily removed with a hand trowel. The few cases where this was not true, areas containing finely divided DU were found to be spread over a few square meters. Remediation of these areas consisted of complete removal of the soils using a flat-bottom hand shovel.

Approximately four 208 liter (55 gallon) drums were filled with DU and DU-contaminated soil from the base pad and 16 layers.

DETECTION CAPABILITIES

A count-rate decision level was established whereby count rates above this level were considered indicators of the possible presence of a radiation source. The decision level needed to provide a reasonable assurance that significant DU sources would be discovered while limiting the number of false positives arising from fluctuations in natural background. If the decision level was established at the mean background plus 3 standard deviations (559 + 132) = 691 counts per counting interval, normal statistics would predict that 0.135 percent of the background counts would exceed the action level. With approximately 70,000 data records per lift, this would result in 95 false positives per lift. Increasing the action level to the mean background plus 4 standard deviations (735) results in only 2 false positives per lift. This level was considered acceptable from a false positives perspective. The text that follows evaluates the acceptability of using this decision level from the perspective of the probability of missing significant DU radiation sources (false negatives).

Detection efficiency experiments were performed on site using DU fragments from the site. Fragments were placed beneath the center of a stationary detector within the array and the count rate measured using a 30-second count. The source was moved outward along the radius in 2.5-cm increments with the count rate measured at each location, out to a total distance of 30.5 cm. The measurements were repeated after burying the sources beneath 2.5 cm, 5.1 cm, and 7.6 cm of soil. Considering that most detectors in the array have another detector nearby that will affect the view of the source, the average efficiency was calculated using the measured efficiencies, the geometry of the array, and the various positions of the source relative to the start of the 3-second counting interval.

The average efficiency for each combination of DU fragment mass and depth was calculated by first generating a “map of efficiencies” for each combination. The map shows the efficiencies of a FIDLER detector when it is located at any of the coordinates on the map and the DU fragment of the specific mass and at the specific depth is at coordinate (0, 0). The horizontal X-axis of the map is the distance along the direction of the movement of the detector during a survey and it spans from negative 30 cm to positive 30 cm, twice the length the detector travels in one 3 second counting interval while surveying. The horizontal Y-axis is the distance perpendicular to that movement and spans from negative 10.2 cm to positive 10.2 cm, the width of the area scanned by the detector. Figure 5 shows a 3-D representation of the map of efficiencies for an 18.1 gram fragment located on the surface.

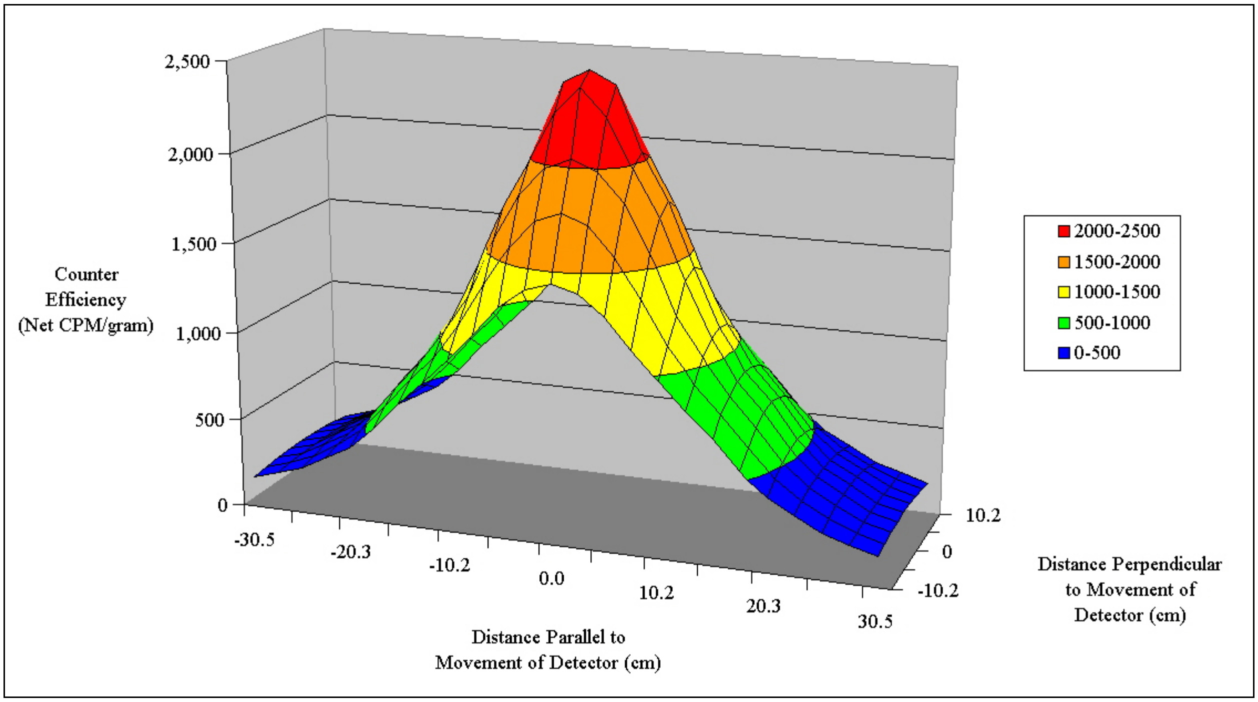

Fig. 5. 3D Representation of the map of efficiencies derived from the measurements with an 18.1 gram DU particle located on the ground surface.

These maps were then used to calculate the average efficiency of the FIDLER detector for each combination of DU fragment mass and depth examined. The average efficiency for a combination of fragment size and depth was calculated by using Equation 1 with the associated map of efficiencies,

$$\text{Average Efficiency} = \frac{\displaystyle\int_{y=-10.2\,\text{cm}}^{y=10.2\,\text{cm}} \int_{a=-30.5\,\text{cm}}^{a=0\,\text{cm}} \int_{x=a}^{x=a+30.5\,\text{cm}} \varepsilon(x,y)\,dx\,da\,dy}{30.5\,\text{cm} \bullet 30.5\,\text{cm} \bullet 20.4\,\text{cm}} \tag{Eq. 1}$$

where x is the distance parallel to the movement of the detector, y is the distance perpendicular to the movement of the detector, a is the x-location of the detector at the beginning of the counting interval, and $\varepsilon(x, y)$ is the efficiency of the detector at x and y on the map. Equation 1 sums and averages all the efficiencies that the detector has over the course of a counting interval and this is done for each possible location of a counting interval in relation to the DU particle located at (0,0). The results of the calculations, their associated maximum efficiencies when the detector was directly over the DU fragment, and the ratio of the two are shown in Table III.

Fragment Mass (grams)	Fragment Depth (cm)	Average Efficiency (Net CPM/g)	Maximum Efficiency (Net CPM/g)	Average/ Maximum
1.2	Surface	4433	7763	0.571
1.2	2.5	768	1112	0.691
1.2	5.1	565	848	0.666
1.2	7.6	399	694	0.575
12.2	Surface	1344	2240	0.600
12.2	2.5	518	842	0.615
12.2	5.1	434	606	0.716
12.2	7.6	293	400	0.733
18.1	Surface	1614	2464	0.655
18.1	2.5	612	896	0.683
18.1	5.1	543	748	0.726
18.1	7.6	319	465	0.686

Table III: Average and maximum counting efficiencies of the FIDLER detector and their ratio for each combination of DU fragment mass and depth examined.

The average efficiencies were used to calculate the maximum concentration of U-238 in a lift for each combination of DU fragment mass and depth examined, after remediation of areas measured to have counts greater than or equal to the decision level. The maximum concentrations were derived numerical by calculating probabilities of false negatives for a range of masses of DU fragments, using normal statistics and the average efficiency, and multiplying those probabilities by their associated concentrations of U-238 in the soil. An associated concentration is the activity of the DU fragment mass divided by the mass of soil scanned in a counting interval. The largest concentration for a combination of DU fragment and depth was used as the maximum concentration for that combination. These values assume the soil in the other layers has no DU, the soil density to be 1.2 g/cm3, and the specific activity of the DU fragments to be 0.4 µCi/g. The maximum concentrations for each combination are shown in Table IV.

Fragment Mass (grams)	Fragment Depth (cm)	Maximum Concentration (pCi/g)
1.2	Surface	33
1.2	2.5	188
1.2	5.1	256
1.2	7.6	362
12.2	Surface	108
12.2	2.5	279
12.2	5.1	333
12.2	7.6	493
18.1	Surface	90
18.1	2.5	236
18.1	5.1	266
18.1	7.6	453

Table IV: Table of maximum concentrations of U-238 in a lift after cleanup, calculated from the average efficiencies listed in Table III.

These maximum concentrations are the worst-case scenarios because if DU were in multiple layers the probability of a false negative would decrease and the resulting concentration would be smaller. For each DU fragment mass examined, all maximum concentrations were less than 500 pCi/g.

Since each DU fragment has an equal probability of being on the surface, 2.5 cm, 5.1 cm, or 7.6 cm below the surface, the four maximum concentrations for each DU fragment mass, one for each depth, can be averaged to yield an average maximum concentration for that mass. For the 1.2 gram fragment, the average maximum concentration was 210 pCi/g. For the 12.2 gram fragment: 303 pCi/g and for the 18.1 gram fragment: 261 pCi/g.

CONFIRMATION SAMPLING AND ANALYSIS

Sandia National Laboratory personnel collected QC verification samples from the remediated pile. After 4 lifts had been applied, nine 30-cm deep composite samples were taken at random locations and analyzed for uranium-238. Uranium-238 concentrations in all samples were less than 5 pCi/g. Thereafter, 43 biased samples were taken from areas within lifts where fragments or contaminated soils had been removed. Only one sample was found to have significant contamination, 158 pCi/g. The average of all samples, excluding the highest value, was 12 pCi/g.

LESSON LEARNED AND CONCLUSIONS

The primary difficulty during this "proof of concept" study was associated with placing the 7.6-cm lift on the pad. The plan was to place a lift of soil on the compacted pad and then remove it prior to placing another lift. Construction personnel suggested that lifts be added to the pad, eliminating a handling step. This approach was accepted but it was soon found that water had to be added and a compaction step added to provide pile stability. In addition, the construction operator had difficulty adding the desired thickness with the available equipment. This led to a very inconsistent average layer thickness of approximately 3 to 4 cm rather than the intended 7.6 cm which greatly reduced the production rate. As the pile grew taller, the area naturally became smaller which also reduced production rate.

This study clearly showed that it is technically possible to use this method to remove DU fragments from soil such that any reasonable cleanup criterion may be met. The number of DU fragments per acre averaged approximately 20 to 25 and therefore the time required to return to each anomaly was not excessive. For this project, the cost to scan and remove the fragments and contaminated soils was approximately $65 US per cubic meter. In a production environment, this cost would be expected to be reduced to less than $40 US per cubic meter. Further improvements in application of the layers so that the detectors could have been placed nearer the ground surface would have also significantly increased the sensitivity of the method. The additional cost for placing the material was not available.

REFERENCES

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