A FIELD STUDY DESIGNED TO SELECT THE IN-SITU INSTRUMENT MOST USEFUL FOR ESTIMATING URANIUM CONTAMINATION IN SOIL

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ABSTRACT

A requirement for any remedial action involving a site contaminated with radionuclides from the natural series is a field instrument capable of quickly estimating land areas which exceed cleanup guidelines. Much work has been done on uranium mill tailings sites in which a major contaminant was Ra-226. Considering the high energy gamma radiation emitted by Ra-226 daughter products, it is not surprising that detector configurations using a NaI scintillometer operating in a gross count mode have proven quite useful.

Remedial actions are now underway on sites primarily contaminated with uranium not in secular equilibrium with its daughter products (limited Ra-226). In this situation, the choice of the proper field instrument becomes less clear. This paper presents the results of a field study designed to select the instrument most appropriate for detecting uranium in excess of remedial guidelines. The instruments tested were a shielded pancake geiger-mueller detector, a Field Instrument for Detecting Low Energy Radiation (FIDLER) operated in the gross count mode and the single channel mode, and an unshielded 2 X 2-inch NaI scintillometer. It is concluded that the unshielded 2 X 2-inch NaI scintillometer provides the most accurate and precise means of estimating uranium concentrations in soil. In addition, its use minimizes the probability of excavating soil having natural background radionuclide concentrations.

INTRODUCTION

Cleanup of sites contaminated with radionuclides requires an instrument capable of estimating in-situ soil concentrations of radionuclides. On sites contaminated with Ra-226 and daughter products, gamma scintillometers operating in the gross count mode have proven to be reliable for characterizing areas of contamination and guiding excavation. This is because Ra-226 and daughter products have a high emission rate of energetic gamma photons. More recently, however, remedial actions have begun on sites contaminated with uranium not in secular equilibrium with daughter products. In this situation, the choice of the proper field instrument becomes less clear due to a low yield per disintegration of low energy gamma photons and a significantly higher beta emission rate from Pa-234 and Th-234.

The Weldon Spring Remedial Action Project (WSSRAP) involves cleanup of the Weldon Spring Chemical Plant which refined uranium metal from yellow cake (uranium ore concentrate) from 1957 to 1966. As a result of uranium refining operations other land areas in the vicinity of the Weldon Spring Chemical Plant became contaminated with uranium. In conjunction with radiological characterization efforts on these vicinity properties a study was conducted on a seven acre property to select the instrument and method of measurement most capable of estimating in-situ uranium levels. The uranium is not in secular equilibrium with daughter products because the uranium had been extracted from the natural ore prior to shipment to the Weldon Spring facility.

There were two objectives of the study: (1) to select the field instrument most capable of determining the presence or absence of uranium concentrations in excess of remedial guidelines; and (2) once the "best" instrument was selected, to determine the appropriate instrument count rate cutoff value to be used to guide the necessary remedial action. The instrument selection criteria were based on correlation of instrument response to U-238 concentrations in soil and the ability of the instrument to differentiate between a count rate indicative of contamination in excess of remedial guidelines and the natural background count rate. The natural background count rate is due in part to naturally occurring concentrations of U-238 and daughter products, Th-232 and daughter products, and K-40. The instruments used in the field study were a shielded pancake geiger-mueller detector, a Field Instrument for Detecting Low Energy Radiation (FIDLER), and an unshielded 2 X 2-inch NaI scintillometer.

The remedial guideline for the property on which the study was conducted is tentatively set at 2.2 Bq/g U-238. Each instrument, however, must be capable of detecting lower concentrations of uranium since the property may be decontaminated to 0.56 Bq/g U-238 wherever reasonable and cost effective as part of the effort to maintain exposures "as low as reasonably achievable".

The shielded pancake geiger-mueller counter was included in the study because of the abundant beta decays of

Th-234 and Pa-234 relative to the infrequent gamma intensities of Th-234 and Pa-234. Refer to Table I for a listing of the characteristic radiations from these radionuclides. The shielding on the geiger-mueller detector consisted of a half inch of lead on three sides of the detector which reduces the background count rate.

The FIDLER consists of a 0.63-inch thick by five-inch diameter NaI scintillation detector with a 0.01-inch thick beryllium entrance window. It was selected for this study due to its sensitivity to gamma rays from approximately 40 keV to 400 keV and its insensitivity to higher energy cosmic rays and photoelectric events from Ra-226 daughters, K-40 and Th-232 daughters. The FIDLER was operated in both the gross count mode with a threshold of 60 keV and also as a single channel analyzer for energies between approximately 60 keV and 100 keV.

The 2 X 2-inch NaI scintillometer was selected for the study because of its high sensitivity to gamma radiation. No lead shielding was used to collimate the detector field of view because this hinders ease of operation.

METHOD

Instrument count rate measurements were taken with each detector at contact with the soil surface. A *six-inch* deep soil sample was also collected at each measurement location for subsequent U-238 analysis using a high purity germanium detector. The count rate of each instrument could then be correlated to U-238 concentration in soil.

Soil samples were collected with a hand operated three-inch diameter hollow stem bucket auger. The diameter of the soil samples was not completely representative of the field of view of each detector. Since the field of view was different among detector types some error was introduced into this study due to inhomogeneity in the distribution of contamination. This error is considered negligible, however, with respect to uncertainties in the correlation between instrument count rate and U-238 concentration.

Considering the short travel distance of beta radiation and low energy gamma radiation, depth of soil sample collection becomes an important factor. A sample depth of two to three inches should yield a better correlation between instrument count rate and uranium concentration due to low detection probabilities of beta or low energy gamma

TABLE I

Decay Characteristics of Radionuclides
in the Natural Uranium Series With Other Daughters Removed

		ENERGY, MeV		
NUCLIDE	HALF-LIFE	ALPHA	BETA	GAMMA (photons/decay)
U-238	4.51X109y	4.18		
Th-234	24.10d		0.193,0.103	0.092 (0.04) 0.063 (0.03)
Pa-234M	1.175M		2.31	1.0 (0.015), 0.76 (0.0063)
Pa-234	6.66h		0.5	
U-234	2.48X105y	4.763		

radiations which originate deeper in the soil. A sample depth of six inches was selected, however, because remedial standards are based on radionuclide concentrations averaged over six-inch depth increments (40 CFR 192).

Soil samples were homogenized by mixing but were not dried and were sealed in 500 ml cans. Samples were analyzed for U-238 by detection of the 63 keV and 92 keV gamma rays of Th-234 on a high purity germanium detector. A sample analysis time of 15 minutes yielded a lower limit of detection of 0.07 to 0.11 Bq/g U-238.

This study was designed to be representative of actual field conditions during remedial action. For this reason, rigid experimental controls were not considered desirable. This is because field measurements are meant to guide and plan a remedial action during which all variables cannot be tightly controlled.

RESULTS

Results of the measurements are split into two categories: measurements taken at locations having natural background concentrations of radionuclides, and measurements taken where U-238 concentrations were above background. The average, standard deviation, coefficient of variation, and number of measurements for all in-situ measurements taken at soil sample locations having natural background radionuclide concentrations are shown in Table II. Soil sample analyses showing a positive identification of U-238 and the corresponding in-situ measurements are shown on Figs. 1 through 4.

All soil sample results having U-238 concentrations less than the detection limit for U-238 and having Ra-226 and Th-232 concentrations less than 0.07 Bq/g were assumed to approximate natural background soil radionuclide concentrations. Reference to Table II shows that the geiger-mueller detector has the highest coefficient of variation among measurements taken where the soil radionuclide concentrations were at background levels while the coefficient of variation for the other three data sets are approximately the same. The higher coefficient of variation for the geiger-mueller detector may be explained by counting statistics due to low count rate. Longer count times would probably improve performance, however, longer count times lead to increased time and expense for field measurements.

Figures 1 through 4 represent simple linear correlations between each instrument count rate and U-238 concentration. Some soil samples had U-238 concentrations much greater than 2.2 Bq/g, but these results were excluded to reduce potential bias in the correlations. Also shown on the Figures are the 95 percent confidence limits on the predictor.

DISCUSSION

By experimental design this study was conducted to be completely representative of actual remedial action field measurements, and a large variability in correlation between instrument count rate and soil U-238 concentration was expected. Thus, the objective of the study was to select the instrument capable of providing a count rate cutoff value having a high degree of confidence of detecting soil containing a U-238 concentration of 0.56 Bq/g or greater. Two criteria must be satisfied when selecting the appropriate type of in-situ measurement used to guide remedial action: (1) there must be a correlation between instrument count rate and the parameter being measured; and (2) the instrument must be capable of differentiating between the natural background count rate and a count rate indicative of contamination exceeding cleanup criteria.

An evaluation of Figs. 1, 2, and 3 indicates that for U-238 concentrations less than approximately 0.56 Bq/g the instrument count rates seem random. The same is true for the 2 X 2-inch NaI scintillometer for U-238 concentrations less than approximately 0.37 Bq/g (Fig. 4). This suggests that for in-situ measurement concentrations lower than approximately 0.37 to 0.56 Bq/g, U-238 cannot be reliably quantified using these field techniques. To test this hypothesis, the FIDLER and geiger-mueller detector data were grouped by U-238 concentrations less than 0.56 Bq/g. The data for the NaI scintillometer were grouped by U-238 concentration less than 0.37 Bq/g. Linear regressions were performed for each data group. A T-test was performed to evaluate whether the slope of the correlation line was equal to zero. Calculated T-values and probability levels are listed in Table III for each instrument. In all cases it can be concluded with 95 percent confidence that there is no reason to suspect that the slope of the correlation line is different than zero. A slope of zero indicates that there is no correlation between instrument count rate and U-238 concentration. These results show that the FIDLER and the geiger counter cannot quantify less than 0.56 Bq/g while the 2 X 2-inch NaI scintillometer cannot quantify less than 0.37 Bq/g.

One further test was conducted with the 2 X 2-inch NaI scintillometer to verify the 0.37 Bq/g quantification level. In this test only the data pairs having U-238 concentrations less than 0.56 Bq/g were used in a linear correlation. When the hypothesis that the slope of the correlation line was equal to zero was tested, a T-value of 2.5 was calculated. In this case, at the 95 percent level of confidence with 13 degrees of freedom, the hypothesis can be rejected. The conclusion drawn is that the 2 X 2-inch NaI scintillometer does have the ability to quantify concentrations below 0.56 Bq/g.

The above discussion indicates that U-238 can be quantified by in-situ measurements as evidenced by the correlations shown in Figures 1 through 4. There also is a large degree of scatter in each data set. Due to the uncertainty in

TABLE II

In-Situ Measurement Results for Soil Containing Background Radionuclide Concentrations

INSTRUMENT	AVERAGE COUNT RATE(cpm)	STANDARD DEVIATION	COEFFICIENT OF VARIATION	NUMBER OF MEASUREMENTS
Geiger-Mueller Detector	65	9.9	0.152	19
FIDLER Gross Count Mode	10775	720	0.067	38
FIDLER Single Channel Mode	7137	565	0.079	41
2 X 2-Inch NaI Scintillometer	12812	845	0.066	41

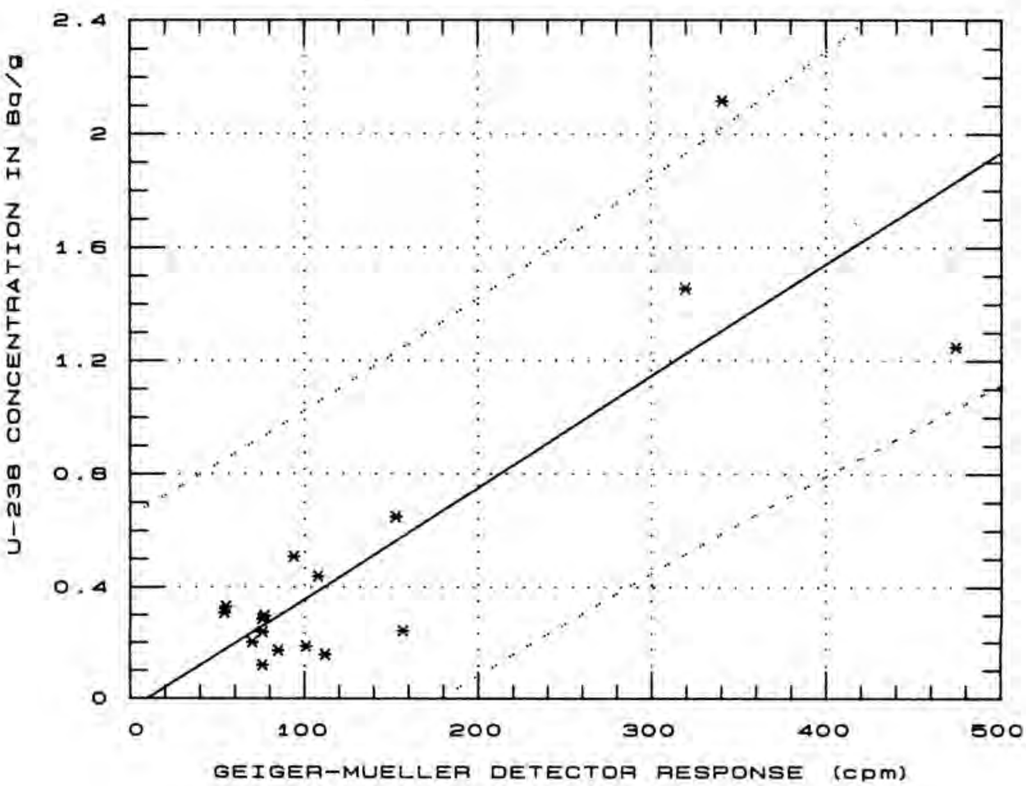

Fig. 1. U-238 Concentration vs Geiger-Muller Detector

TABLE III

Calculated T-Values and Probability Levels

INSTRUMENT	T-VALUE	PROBABILITY LEVEL
2X2 NaI Scintillometer	0.04	0.97
Geiger Counter	-0.09	0.93
FIDLER Gross Count Mode	-0.58	0.57
FIDLER Single Channel Mode	1.22	0.24

the correlation, a count rate cutoff value selected for determining whether the uranium concentration exceeds the cleanup criteria should have a safety factor to reduce the chance of not excavating soil exceeding criteria. If one desires a safety factor that gives 95 percent confidence that no soil exceeds 0.56 Bq/g, reference to Figs. 1 through 4 indicate that cutoff values of approximately zero, 7000, 9000, and 13500 counts per minute should be used for the geiger-mueller detector, the FIDLER operated in the single channel mode, the FIDLER operated in the gross count mode, and the 2 X 2-inch NaI scintillometer, respectively. Obviously, with a cutoff value of zero, the geiger-mueller counter should be excluded from further consideration.

Typically, for an instrument response to be measurable above the background count rate a minimum detectable activity is set at two standard deviations above background. Reference to Table II shows that count rates of approximately 8300, 12200, and 14500 represent the average count rate plus two standard deviations for the FIDLER operated in the single channel mode, the FIDLER operated in the gross count mode, and the 2 X 2-inch

NaI scintillometer, respectively. By comparision of these values to the previously established count rate cutoff values, it is clear that detecting 0.56 Bq/g U-238 with in-situ measurements is difficult. The selection of the appropriate instrument used for field measurements should be based on having 95 percent confidence that material exceeding 0.56 Bq/g U-238 will be removed while excavating the least

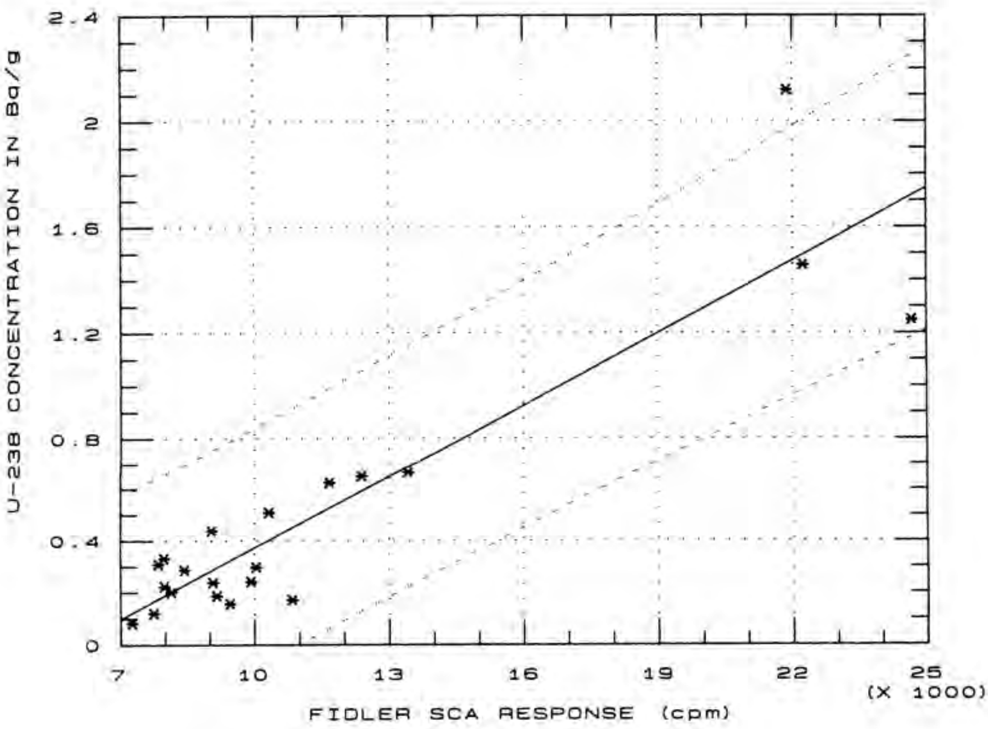

Fig. 2. U-238 Concentration vs. Fidler SCA Mode.

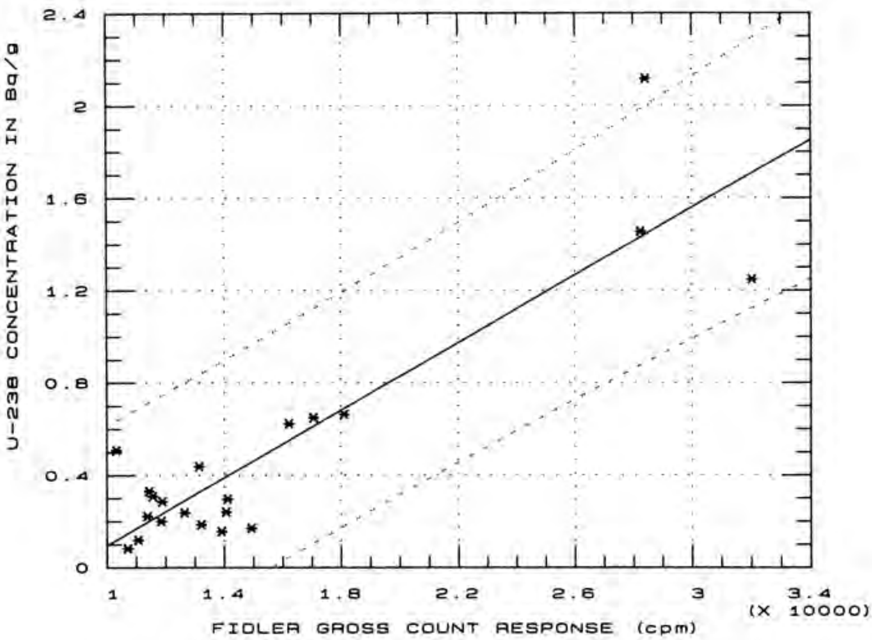

Fig. 3. U-238 Concentration vs. Fidler Gross Count Mode.

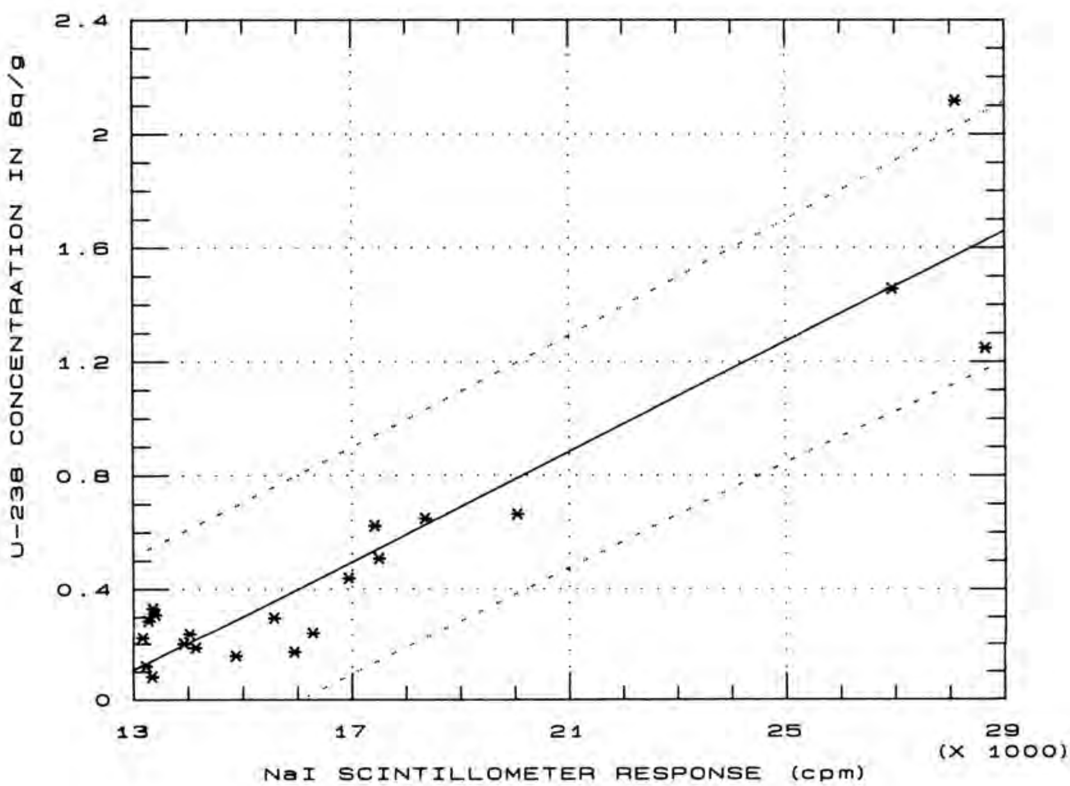

Fig. 4. U-238 Concentration vs NaI Scintillometer.

amount of material having background concentrations of U-238.

By using count rate cutoff values of 7000, 9000, and 13500 counts per minute as determined previously, the probability that soil containing background concentrations of radionuclides will be removed can be calculated by transforming the natural background count rate distribution for each instrument to the standard normal distribution. This is done by subtracting the mean background count rate from the cutoff count rate and dividing by the background standard deviation. When this is done, it is estimated that soil having natural background U-238 concentrations will be excavated 59 percent of the time if the FIDLER is used in the single channel mode, 99 percent of the time if the FIDLER is used in the gross count mode, and 21 percent of the time if the 2 X 2-inch NaI scintillometer is used.

SUMMARY

Detection of land areas contaminated with uranium not in secular equilibrium with daughter products by in-situ measurements is difficult due to the emission of predominantly low energy radiations. During the radiological characterization of a WSSRAP vicinity property a study was conducted to select the field instrument most effective for guiding remedial action. Accurate quantification of uranium concentrations in soil was not the primary objective, rather the ability to have 95 percent confidence of detecting uranium concentrations exceeding cleanup guidelines while excavating a minimum volume of uncontaminated soil was sought.

The appropriate field instrument was selected by correlating soil U-238 concentration with the count rate response of a geiger-mueller detector, a FIDLER, and a 2 X 2-inch NaI scintillometer and by examining the distribution of the count rate of each instrument when measuring natural background concentrations of radionuclides. From these evaluations it is clear that the 2 X 2-inch NaI scintillometer provides the best capability of detecting U-238 contamination exceeding 0.56 Bq/g with the least probability of excavating uncontaminated soil. The quantification limit for the 2 X 2-inch NaI scintillometer is approximately 0.37 Bq/g U-238.