ASSAYS AND SCREENING OF ALPHA CONTAMINATED SOILS USING LOW-RESOLUTION ALPHA SPECTROSCOPY OF THICK SOIL SAMPLES

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Assays and Screening of Alpha Contaminated Soils Using Low-Resolution Alpha Spectroscopy of Thick Soil Samples

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ABSTRACT

A new approach to estimating concentrations of alpha-emitting contaminants (e.g. U, Th, Ra, Pu, Am) in soil samples has been evaluated. The Victoreen Alpha Activity Monitor has been designed to empirically assay soil samples using low-resolution alpha spectroscopy of thick soil samples. Pre-processing of the soil samples is minimal, involving only drying the soils and milling them to reduce inhomogeneities.

Several laboratory tests of the instrument have been conducted. It has been shown that the instrument obeys simple counting statistics with measurement reproducibility improving with the inverse square root of the counting time. Using acquisition times of 1 to 24 hours the instrument generated alpha assays of eighteen reference and field soil samples which were in good agreement with radiochemical analyses. It was observed that the alpha activity due to a 10pCi/g $^{239}$Pu spike added to a clean soil could be very readily distinguished from the 14pCi/g background activity of the soil. In a mock field screening test of 100 Pu-contaminated soils, the instrument demonstrated screening success rates ranging from 70% (at a screening level of 5pCi/g) to greater than 90% (at a screening level of 30pCi/g).

INTRODUCTION

Remediation of radiologically contaminated sites often requires characterization of alpha-emitting isotopes (e.g. U, Th, Ra, Pu, Am) in soils. The two conventional methods for obtaining this information, soil sampling/radiochemistry/alpha spectroscopy [1-3] and in-situ gamma spectroscopy [4], are limited in their abilities to generate data quickly and inexpensively. Soil sampling is labor intensive and radiochemistry is expensive, time-consuming, and generates new waste streams. Gamma spectroscopy requires sophisticated instrumentation, specially trained personnel, provides little information about the depth profile of contaminants, and cannot detect pure alpha emitters ($^{232}$Th, $^{238}$U) or weak gamma emitters ($^{239}$Pu, $^{241}$Pu). Hence there is a need for new instrumentation which can provide on-site same-day assays of alpha activity in soils accurately and inexpensively. This paper describes the results of evaluations and tests of a field-portable low-resolution alpha spectroscopy system which may meet these needs.

The instrument that has been evaluated is the Model 758A1W Alpha Activity Monitor manufactured by Victoreen Inc. (Cleveland, OH). It is an adaptation of the Model 758 AlphaCAM™ Continuous Air Monitor which was designed to determine alpha activity in air via deposition on filter papers. The unit consists of a small sample chamber/Si detector module, multichannel analyzer, control/readout module, and mechanical pump. A PC is required for logging and analyzing data. A program called SANDAL is provided for data acquisition, plotting of alpha
spectra, and analysis.

The conventional approach to performing alpha spectroscopy of environmental samples involves (1) isolating the contaminant(s) of interest, (2) depositing the isolated component in the form of a very thin uniform film on a clean substrate, and (3) characterizing the alpha activity of the film using a vacuum chamber and high-resolution silicon or Frisch grid detector. A thin film format is necessary because alpha particles are easily stopped or slowed down by most materials, and in this process of slowing down their initially discrete energies are degraded and smeared out. The advantage of this approach is that it can distinguish between many different alpha-emitting isotopes closely spaced in energy. A secondary advantage is that detection levels as low as 0.01 pCi/g can be achieved provided that chemical recovery yields are high, sample areas are large, and detection times are long.

The characterization approach being examined here relies on low-resolution alpha spectroscopy of thick nominally unprocessed soil samples. Lucas [5,6] has shown that the alpha spectrum for a radionuclide uniformly distributed in a thick medium will consist of a sharp high-energy edge, corresponding to unscattered alpha particles, plus a broad low-energy tail due to the many particles which have lost varying amounts of material in the medium. The SANDAL analysis algorithm has been designed to seek sharp edges in the acquired spectrum and then fit trapezoidal tails to the low-energy side of the peak. The algorithm does not require any empirical calibration but does require that the operator input a guess of the identity of the isotope(s) in the sample. A typical measurement would proceed as follows:

1) Distribute soil sample in shallow 2" diameter planchet;
2) Load planchet into sample chamber;
3) Pump out chamber (~ 1 minute);
4) Acquire data (10 minutes - 24 hours);
5) Transfer spectrum to PC and store on disk;
6) Plot spectrum, fit it, and generate assay estimate(s).

Some of the advantages of this measurement approach include low capital equipment costs, small equipment footprint, minimal personnel training, same-day analysis, isotope identification capability, no generation of additional waste streams, and (some) ability to discriminate against natural background activity.

One of the potential applications for this measurement technique is in providing rapid soil assays with moderate to high accuracy in a field or on-site laboratory setting. Here "rapid" refers to same-day or next-day analysis, which is sufficient for a great many site characterization scenarios and an order of magnitude faster than typical turn-around times for radiochemical analyses. Using reference soils, spiked soils, and contaminated soils collected from several DOE sites, we have compared the results of radiochemical analysis of these soils with activity estimates generated by the Victoreen instrument. The results will be discussed below.

A second potential application is in quick screening of soil samples to separate areas of 'high' and 'low' contamination. This is frequently the first step in a remediation effort because different degrees of contamination require different levels of personnel protection, planning, instrumentation, and may dictate altogether different remediation technologies. We have conducted a mock screening test in the laboratory to determine how well the instrument can distinguish between 'hot' and 'cold' soil samples at a particular screening level using 10-minute acquisition times.

BASIC SYSTEM PERFORMANCE

A simple procedure was developed to reduce the aliquot-to-aliquot inhomogeneity of soil samples collected in the field. First the soils were dried at 100°C for one hour, then they were milled in a 1 gallon jar mill with burndum cylinders for 3 hours. Finally the soils were sieved at 1mm to remove rocks and pebbles. This processing had the desired effect of reducing the aliquot-to-aliquot variability of the gross alpha emission rate from ~30-50% to ~5-10%. All field samples discussed in this paper were processed in this manner.
A typical alpha spectrum for a highly contaminated soil sample collected from the Nevada Test Site is shown in Fig. 1. Radiochemical analysis at ORNL [7,8] of an aliquot of this sample yielded assays of 757 pCi/g $^{239}$Pu and 167 pCi/g $^{241}$Am. The spectrum exhibits the features expected for a thick source: two edges at approximately 5.2 MeV and 5.5 MeV corresponding to unscattered alpha particles emitted from $^{239}$Pu and $^{241}$Am, plus low-energy tails due to alpha particles which have lost varying amounts of energy in the soil matrix. The detailed shape of the tail depends on how the contaminant is distributed within the soil and the energy-loss characteristics of the medium. The estimated assays generated by the Victoreen algorithm for this sample were 725 pCi/g $^{239}$Pu and 144 pCi/g $^{241}$Am.

The situation is more complicated for soils with low levels of contamination. In this case the natural background alpha activity of the soil matrix, which normally includes $^{238}$U, $^{232}$Th, and as many as fourteen alpha-emitting daughter isotopes, must be taken into account. An example of an alpha spectrum of an uncontaminated thick soil sample (collected in Oak Ridge) is shown in Fig. 2(a). Note that there are no clearly defined sharp spectral edges. The counts recorded in this spectrum are likely due to the natural combination of $^{238}$U (4.2 MeV), $^{234}$U (4.7 MeV), $^{230}$Th (4.7 MeV), $^{226}$Ra (4.8 MeV), $^{222}$Rn (5.5 MeV), $^{218}$Po (6.0 MeV), $^{218}$At (6.7 MeV), $^{210}$Pb (3.7 MeV), $^{210}$Po (5.3 MeV), $^{227}$Th (4.0 MeV), $^{228}$Th (5.4 MeV), $^{224}$Ra (5.7 MeV), $^{220}$Rn (6.3 MeV), $^{216}$Po (6.7 MeV), and $^{212}$Bi (6.1 MeV). Using a subset of ten of these isotopes spanning the energy range as a 'guess', the Victoreen instrument estimated the total alpha activity of this soil sample to be 14 $\pm 0.4$ pCi/g. This estimate did not depend strongly on which subset of isotopes were chosen.

An experiment was then performed to determine how well a single low-level contaminant could be distinguished from the natural background activity. A solution of $^{239}$Pu was added to a weighed quantity of the same 'clean' soil. Based on the known solution concentration and small measurement uncertainties the known plutonium activity of the spiked soil was 10 $\pm 0.5$ pCi/g. The alpha spectrum for this spiked soil is shown in Fig. 2(b). A sharp edge at 5.1 MeV, corresponding to the unscattered $^{239}$Pu energy, can be seen above the background noise. The Victoreen estimate for the $^{239}$Pu component of the alpha activity is 10.2 $\pm 0.2$ pCi/g, in excellent agreement with the known activity. Clearly it is possible to measure single contaminant concentrations down to the same order of magnitude as background activity levels. Multiple low-level contaminants obviously present a more challenging problem and are beyond the scope of this paper.

A set of measurements were conducted to establish that the reproducibility of assays generated by the instrument was governed by simple counting statistics. A reference soil spiked with 100 pCi/g $^{239}$Pu was prepared. One aliquot of this soil was loaded into the instrument, and many alpha spectra were acquired for this aliquot with acquisition times ranging from 2 minutes to 3 hours. Five different spectra were acquired for each acquisition time. For each acquired spectrum the instrument generated an estimated $^{239}$Pu assay. The mean and standard deviation of the mean of the assay estimates were then calculated for each acquisition time. The experimental results are plotted in Fig. 3. Also shown is the counting statistics prediction for the standard deviation of the mean based on $\{N_x(t)\}^{1/2}$ where $N_x$ is the number of alpha particles emitted from the soil surface during the acquisition time t. $N_x$ was calculated from the alpha emission rate averaged over one week. It was observed that the improvement in measurement reproducibility with increasing acquisition time could be accounted for very well with simple counting statistics.

**ACCURACY OF SOIL ASSAYS**

In order to characterize the accuracy of the alpha assay of the Victoreen system a wide range of soils were analyzed. These soils included:

- National Reference Material Disequilibrium Set [9]: $^{238}$U and $^{234}$U mixed with a soil diluent; very fine and homogeneous soil; well characterized
- National Reference Material Equilibrium Set [9]: uranium and thorium ores mixed with a soil diluent; very fine and homogeneous soil; well characterized
Spiked Soils: NIST-traceable solutions of $^{239}$Pu mixed with clean dry Oak Ridge clay-based soils, then dried and milled.

Fernald soils: "calibration bed" soils collected during participation in the Uranium in Soils Integrated Demonstration at the Fernald Environmental Management Project, 7/94

NTS soils: soils collected during field tests at the Nevada Test Site Area 11B [10]

Alpha spectra were acquired on the Victoreen instrument for each of these eighteen soils. Acquisition times ranged from 1 to 24 hours depending on the activity of the soils. For soils with activities above 50 pCi/g, the spectra were fitted only with the peaks corresponding to the known contaminants. For lower activity soils, the spectra were fitted with the known contaminant peaks plus a set of U/Th/daughter peaks to represent the natural background activity of the soil matrix.

The results are summarized in Table 1. The agreement between the standard radiochemical analyses and the Victoreen assays are generally very good, and the bias of the Victoreen assays relative to the radiochemical results appear to be randomly distributed around zero. We believe the observed discrepancies between the Victoreen and radiochemical assays are due to residual aliquot-to-aliquot inhomogeneities, which can never be completely eliminated. The cases of highest bias correspond to the lowest activity soils, Fernald C0 and NTS2-CP. These are 'nominally uncontaminated' or 'background' soils. In this regime the radiochemical approach has an inherent advantage in chemically separating the isotopes. The Victoreen analysis relies on numerical separation of several closely spaced alpha peaks with similar intensities, which results in higher statistical uncertainties.

FIELD SCREENING TEST

A mock field screening test was set up using a set of 100 soil samples from the Rocky Flats Plant (Grand Junction, CO) contaminated with $^{239}$Pu and $^{241}$Am. The soils had been radiochemically analyzed at Rocky Flats and this data was used as a reference. The radiochemical data indicated that the contaminant activity in the soil set ranged from 1.1 to 465 pCi/g with an average of 65 pCi/g and a standard deviation of 143 pCi/g. Each soil sample was characterized only once with the Victoreen instrument using a 10 minute acquisition time. The isotope set that was used to fit the spectra included $^{239}$Pu, $^{241}$Am, and eight U/Th/daughter isotopes to represent the background soil activity. A measurement was scored as a successful screen if either (a) the Victoreen-based assay and the radiochemical assay were both above the chosen screening level or (b) the Victoreen-based assay and the radiochemical assay were both below the chosen screening level. If either (a) the Victoreen estimate was above the screening level while the radiochemical assay was below the screening level (i.e. Type I false positive error) or (b) the Victoreen estimate was below the screening level while the radiochemical assay was above the screening level (i.e. Type II false negative error), the measurement was scored as an unsuccessful screen. A summary of the results for a range of arbitrarily chosen screening levels is plotted in Fig. 4. It was observed that at the most stringent screening level of 5 pCi/g the instrument was successful 70% of the time. At a more realistic screening level of 30 pCi/g the success rate increased to greater than 90%. It should be noted that the detailed shape of the trend will depend on the characteristics of the soil set used for the test.

CONCLUSIONS

A new approach to screening alpha-contaminated soils, low-resolution alpha spectroscopy of thick soil samples, has been evaluated. Two different modes of operation have been investigated: detailed assays of alpha-emitting components and quick field screening of samples. In a test of assay accuracy eighteen reference and field samples were characterized by the instrument. The Victoreen estimates agreed favorably with radiochemical analyses for all but the two lowest activity soils. A mock field screening test was set up using 100 Pu-contaminated soils from the Rocky Flats Plant. Using 10 minute counts the instrument demonstrated a screening success rate ranging from 70% (at a screening level of 5 pCi/g) to greater than 90% (at a screening level of 30 pCi/g).

This instrument presents the potential advantages of same-day or next-day alpha assays, quick
field screening of soil samples, identification of unknown alpha contaminants, minimal operator training, low capital costs, small equipment footprint, no generation of new waste streams, and some discrimination capability against background alpha activity.

The instrument may be powered by a 12V car battery, monitored with a laptop PC, and deployed in a truck or van. Field tests in Oak Ridge of the instrument in this configuration are planned for early 1995.

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REFERENCES

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<th>Soil</th>
<th>Description</th>
<th>Contaminants</th>
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<th>Victoreen assay pCi/g</th>
<th>Victoreen % bias</th>
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<td>NRM1</td>
<td>National Reference Material</td>
<td>$^{238}\text{U}$, $^{234}\text{U}$, $^{226}\text{Ra}$</td>
<td>32 ± 1</td>
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<td>88 ± 2</td>
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Nevada Test Site Soil
725pCi/g Pu239 + 144pCi/g Am241

Figure 1 Alpha spectrum of highly contaminated Nevada Test Site soil.

Low-Level Contamination

Figure 2 Alpha spectra of (a) uncontaminated Oak Ridge soil and (b) same soil spiked with Pu239.
Overall Screening Success Rate

Figure 4: Measured success rate of screening tests on Rocky Flats soils at different screening levels.

Figure 3: Improvement in measurement reproducibility with increasing acquisition time.