Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

Title: In-situ method development to identify radiological contamination in soils

Many areas across the United States (Savannah River Site, Hanford Reservation) and the world (Chernobyl, Fukushima) are contaminated with radionuclides. One of the most common contaminants is ^{137}Cs, a medium-lived byproduct of nuclear fission and the focus of this work. Cleaning up these areas requires the contaminated soil to be identified and then removed and properly disposed. Rapid measurements at the site could eliminate the need for 90% of laboratory analyses, which would reduce waste and decrease cost. Increasing the number of sites analyzed per dollar spent could more precisely define a contamination area.

A shielded, commercial-off-the-shelf detector system was used to detect ^{137}Cs near the surface (top 6 inches) of contaminated soil. The measurements were coupled with a nuclear physics model to create a report of the possible contamination levels within the measured volume.

The system was tested at several locations at the Savannah River Site. At locations with known ^{137}Cs contamination, it could be detected within 60 seconds. At locations where ^{137}Cs contamination was not expected to exceed the global background, none was detected even after a one-hour measurement. In the final method, each measurement was made for 10 minutes.

The method developed from this project is appropriate for use by minimally trained personnel at sites with contamination levels greater than cleanup standards. It is not appropriate for general environmental monitoring where contamination levels are no higher than the global background.

Awards and Recognition

None.

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publicly published in its current form.

SRNL Legal Signature

______________________________ ______________

Signature **Date**

LDRD-2018-00125
LDRD Report
SRNL-MS-2018-00169

Title: In-situ method development to identify radiological contamination in soils

Project Team: T. S. Whiteside (Primary), K. M. Fenker, A. D. Brand, T. Aucott

Thrust Area: ES / NS

Project Start Date: October 1, 2017
Project End Date: September 30, 2018

Rapid in-situ analysis of soil contamination could eliminate the need for 90% of laboratory analyses, reduce waste, and decrease cost. Increasing the number of sites analyzed per dollar spent would allow for more precisely defined contamination areas. The proposed work fits in well with DOE-EM and DOE-NNSA missions to monitor the environment during clean-up or post-detonation analysis. It fits with SRNL business-unit program plans for (SRNL-ES) advancing techniques for characterizing contaminants in-situ and (SRNL-NS) advancing sampling and analytical capabilities for detection.

This project has developed an in-situ method to characterize radiological contamination in soils using commercial-off-the-shelf detectors coupled with nuclear physics models so that it is inexpensive, fast, and can be done by minimally trained personnel.

FY2018 Objectives

- Develop a method to measure any type of soil (clay, sand, loam) for contamination of ^{137}Cs, in the field, at accuracies equivalent to those obtained by laboratory measurements.
- Determine if different soil composition and soil moisture content significantly impact radiation measurements.
- Determine quality of portable USB-based instrumentation
- Determine if the depth of contamination can be calculated from measured radiation energies

Introduction

Currently, when sampling soils for radiological contamination, technicians dig holes, remove samples, transport the samples to a laboratory, analyze the samples in the laboratory, and then return the sample to the site or dispose of the sample. Rapid in-situ analysis could eliminate the need for 90% of laboratory analyses, reduce waste, and decrease cost. Increasing the number of sites analyzed per dollar spent would allow for more precisely defined contamination areas. This work builds on a method developed for the Savannah River Nuclear Solutions' Environmental Compliance group to determine ^{137}Cs in fauna and technology developed for DOE's Office of Nuclear Incident Response. The ^{137}Cs method previously developed is as accurate as the laboratory measurements and provides results in minimal time. We extended this method to other media (soils) and were able to report possible contamination levels within the measured volume.

The method developed can be used to rapidly delineate the footprint of a contamination area and report the possible contamination in a specific volume of soil. In addition, this work has examined the performance of different pieces of hardware used during the measurement process which has led to a better understanding of when to select a specific instrument. Furthermore, this work has explored the possibilities of measuring contamination that is buried and using the spectra to report the depth.

Approach

To develop a rapid, portable method for measuring ^{137}Cs in soils we started with the method developed for measuring ^{137}Cs in animals hunted at the Savannah River Site. We determined the impact different soil types (sandy, clayey, loamy) and compositions (wet, dry, porous, solid) would have on the measurements by creating several models using nuclear physics code. To determine the minimum count time, we considered the SCDHEC and EPA regulations to determine the cleanup standards for ^{137}Cs at SRS, which is 28 pCi/g [DHEC2016], desired uncertainty, which is an order of magnitude [EPA2000]. To accurately measure the soil volume, we developed a method for configuring the detector, shielding, and calibration source such that the detector could be held in a consistent position. To determine the concentration in a specified volume of soil (1 cm, 5 cm, 15 cm, and infinite from the surface) we measured the volume "seen" by the detector and used nuclear physics code to distribute a standard quantity of ^{137}Cs throughout these volumes and created scaling factors to relate actual to modeled measurements. We validated these factors on certified phantoms and tested the method in the field.

To make this system more portable (controllable from smartphone or tablet), we evaluated several commercially available USB-based multi-channel analyzers on the basis of ease-of-use, performance, programmability, and price.

To determine the depth of a buried contamination layer, we used the peak-to-valley ratio of a ^{137}Cs spectra obtained at different depths to create a calibration curve. To use this curve with confidence requires knowledge of a site's history.

Results/Discussion

Due to the (relative) low molecular weight of soil minerals, there were no differences in the modeled detector response for soil composed of different types of solids. Different soil saturation levels do impact the detector response, due to the different densities. However, an error of 13% in estimating the density (completely saturated or complete dry soil) results in less than a 7% error in reported concentration at "infinite" depth. Using the cleanup standards, we calculated a minimum count time of 10 minutes. This time will enable detection of contamination as low as 1 pCi/g, but this system will not see global-fallout levels of ^{137}Cs, 0.1 pCi/g.

The soil volume "seen" by the shielded detector was experimentally measured and determined to be equivalent to a right cylinder with a radius of 10 cm and a height of 20 cm. We used the nuclear physics models to provide scaling factors to convert counts to pCi per gram at different depths and densities. We then confirmed the model by measuring certified phantoms, which had a range of concentrations, and converting the counts into concentration.

We took measurements in several locations near monitoring site PSC-002-D1, beside Steel Creek in SRS P-area, which is a known contamination area. At this location, we determined the contamination was within 27 feet of the center of the stream, in contrast to gamma-overflight data showing it to be within 400 feet (93% smaller). As shown in Figure 1, we calculated the soil concentration to be between 1 and 6 pCi/g 18 feet from the center of the creek, between 0.7 and 4 pCi/g 6 feet south-west of that location (24' from center of the creek) and not measurable beyond at 27' from the center of the creek. Approximately 150' upstream, in the middle of the creek-bed, in an area covered in rip-rap, we calculated the soil

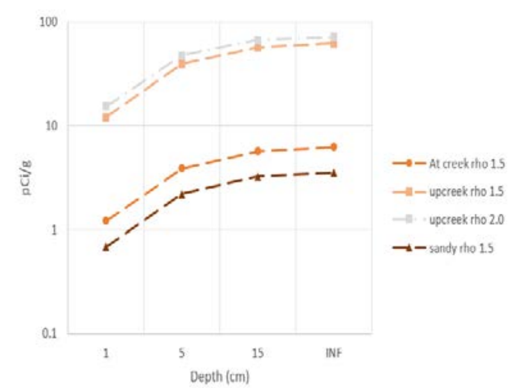

Figure 1. Calculated soil contamination at depth at Steel Creek

concentration to be between 12 and 63 pCi/g. We used the peak-to-valley relationship as reported in SRNL-MS-2018-00120 to see if the contamination was under the rip-rap. We estimated the depth of contamination to be between 1 and 3 inches, which corresponds to a concentration between 12 and 57 pCi/g.

The ease-of-use, performance, programmability, and price was evaluated for the Ortec Digibase, Canberra Osprey, Amptek TB-5, and the Bridgeport usbBase. The Digibase is the easiest to setup and use on conventional systems, for use with Android-based operating systems, the easiest is either the TB5 or usbBase. For programmability, the usbBase or TB5 is easiest, followed by the Digibase and Osprey. Price scales with analog-to-digital converter (ADC) size: the Digibase (10-bit), usbBase (12-bit), TB5 (13-bit), and Osprey (14-bit ADC). The performance of these systems was similar, but mainly driven by the ADC "bitness". This was assessed by comparing measurements of a standard source, ^{166m}Ho. ^{166m}Ho was selected because it has many intense peaks across the range of 80 – 1000 keV. In addition to each other, we compared the performance to the Canberra DSA-LX and the XIA PIXIE-4. Several figures-of-merit were analyzed to compare the data-acquisition systems, including the resolution of the peaks, and the integral and differential linearity of the ADC.

FY2018 Accomplishments

- Developed a simple-to-use method that with minimal further development could be deployed for use in the field.
- Demonstrated that this method is appropriate for determining the footprint of a contaminated area (> 1 pCi/g), it is not appropriate for measuring global-background levels of contamination (0.1 pCi/g).
- Soil composition is not a major source of error in reporting overall concentration of contamination.
- The performance of all USB-based instrumentation is nearly equal. The primary difference between them is the ease of setup (Digibase), simplicity of custom programming (TB-5), and cost (Digibase).
- Depth to contamination can be determined with prior knowledge of a site, however it may not be necessary for practical cleanup purposes.

Future Directions

- Explore the applicability of this work to other radionuclides, specifically Uranium-235/238
    - Backwater Basin, OH (DOE-LM)
    - Tim's Branch at SRS (Clemson University)
- Develop common control software and integrate with driver software for the various USB-based detector hardware. This will enable rapid instrumentation development using the most appropriate equipment for the application.

FY 2018 Publications/Presentations

1. SRNL-STI-2018-00510: Calculating Cs-137 concentration in soil using nuclear modeling and laboratory standards
2. SRNL-STI-2018-00524: Evaluation of multichannel analyzers for gamma-radiation measurements
3. SRNL-MS-2018-00120: Improving the Field Portability of Neutron and Gamma Counting Techniques
4. SRNL-L4120-2018-00023: Uranium measurements in upper Tim's Branch

References

1. DHEC2016 Environmental Surveillance and Oversight Program 2016 Data Report.
2. EPA2000 Environmental Protection Agency Soil Screening Guidance for Radionuclides: User's Guide EPA/540-R-00-007

Acronyms

DOE-EM	Department of Energy Environmental Management
DOE-NNSA	Department of Energy National Nuclear Security Administration
EPA	Environmental Protection Agency
MCA	Multi-Channel Analyzer
SCDHEC	South Carolina Department of Health and Environmental Control
SRS	Savannah River Site
SRNL	Savannah River National Laboratory
USB	Universal Serial Bus

Intellectual Property

None

Total Number of Post-Doctoral Researchers

None