Environmental Characterization Scans Using Sodium-Iodide Gamma Spectroscopy to Determine Ra-226 Concentration in Surface Soil – 16326

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ABSTRACT

The recent natural gas drilling boom has brought heightened attention to the issue of TENORM waste management. Radioactive residues from the extraction, treatment, and purification of materials containing uranium and thorium, or their progeny, pose a public health hazard primarily in the form of ground water contamination. Hydraulic fracturing can liberate formation waters from shale; the high salinity of which can cause radium to move with the water to the surface. The disposal and handling of this water has been a subject of public controversy. For soil remediation projects, radium-226 is generally one of the radionuclides of concern due to its alpha emissions. However performing a survey scan for Ra-226 is difficult because its photon emissions are low energy and have minimal probability of emission per decay (186.2 keV, 3.3% abundance). Additionally resolving its emission from U-235 (185.7 keV, 57% abundance) with field instrumentation is not feasible. Therefore detection of Ra-226 relies on the presence and photon emissions of its progeny. This paper outlines methodology for calculating the minimum detectable concentration of Ra-226 in characterization scans using a sodium-iodide detector array coupled with a multi-channel analyzer.

Using progeny ingrowth as a detection proxy requires knowing decay time in order to determine progeny emission rates. Knowledge of site history, process, and prior survey results can be key in determining an accurate decay time. However in the absence of such information in-situ measurements can be used to estimate decay time by comparing observed progeny emission ratios against established values. These values are determined using Oak Ridge National Laboratory's ORIGEN-ARP program to generate photon energy spectra for Ra-226 and its progeny at specified time intervals. Once decay time is established, the detector's minimum detectable concentration can be calculated by synthesizing the expected emission spectra and Los Alamos National Laboratory's Monte Carlo N-Particle transport code to simulate detector response.

Traditionally, one of the steps in calculating a system's minimum detectable concentration is to place an appropriate button source under the detector, record the response, and then move the source by a specified increment. This process is repeated until the detector's spatial efficiency has been mapped. Due to the impracticality of performing this with Ra-226 button sources of varying decay times, Monte Carlo N-Particle transport code was utilized to determine the detector's spatial efficiency. A dime sized source with the established emission spectra was placed into

the center of a rectangle of soil. This rectangle, including the source, was used as a lattice element to fill the region of space below the detector. The simulation was designed to track the energy distribution of pulses created in the sodium iodide crystal using the F8 tally, and was setup to isolate each source; thereby correlating detector response to source position.

The simulation output required post-processing to determine conversion factors from counts per second to disintegrations per second. Post-processing accounted for subtraction of the Compton continuum, and average the data over the scanning pattern to obtain the expected spatial sensitivity during a characterization study. This was performed for the following photopeaks: 609 keV (Bi-214), and 1764 keV (Bi-214). For calculation of the scan minimum detectable concentration, the surveyor efficiency coefficient from NUREG 1507 was supplanted in favor of a scan speed deviation factor. This was used because the detector utilizes computerized data collection (spatial and spectral). This report finds that, for two shielded 10.16 cm x 10.16 cm x 40.64 cm sodium-iodide crystals with integrated multi-channel analyzers, the minimum detectable concentration in surface soils is less than 1 pCi per gram Ra-226 for a 1 second survey measurement. These results assume a volumetric source with a maximum depth of 15.24 cm and a minimum decay time of 21 days.

Verification of the system's capabilities was performed by sending soil samples from scanned areas, and locations where in-situ measurements were performed, to an independent laboratory for analysis. The laboratory results demonstrated that scanned area was principally comprised of soil containing less than 1 pCi per gram Ra-226. This borders the system MDC and it underestimated the concentration of radium in the soil. The probable causes of this is escape of radon gas from the surface soil. Radon is the daughter of radium, and has a high mobility in soil. If the gas escapes, then the progeny used as the detection proxy will be present in lower than expected concentrations.

Radionuclide identification using high volume sodium-iodide crystals in low level characterization scans is demonstrated to be possible. Further system refinement will focus on developing a methodology to estimate a radon escape factor for site specific use. Accounting for radon escape in the top 15.24 cm of soil will allow the system to better estimate Ra-226 concentrations in soil. This is a significant advancement over the gross counting statistics used in most studies to satisfy MARSSIM survey requirements, because it will increase stakeholder confidence.

INTRODUCTION

The recent natural gas drilling boom has brought heightened attention to the issue of TENORM waste management. Radioactive residues from the extraction, treatment, and purification of materials containing uranium and thorium, or their progeny, pose a public health hazard primarily in the form of ground water contamination. Hydraulic fracturing can liberate formation waters from shale; the high salinity of which can cause radium to move with the water to the surface. The disposal and handling of this water has been a subject of public controversy. For soil remediation projects, radium-226 is generally one of the radionuclides of concern due to its alpha emissions. However, performing a survey scan for Ra-226 is difficult because its photon emissions are low energy and have minimal probability of emission per decay (186.2 keV, 3.3% abundance). Additionally resolving its emission from U-235 (185.7 keV, 57% abundance) with field instrumentation is not feasible. Therefore, detection of Ra-226 relies on the presence and photon emissions of its progeny. Historical knowledge of a site can be used to estimate if the soil has been disturbed within 24 hours and some level of progeny ingrowth can be assumed and used as a detection proxy. Bi-214 and Pb-214 were selected for use as Ra-226 proxy because of their abundant photon emissions. The 352 keV (Pb-214), 609 keV (Bi-214), 1120 keV (Bi-214), and 1764 keV (Bi-214) photopeaks were initially selected for used to determine radium concentrations. This paper outlines methodology for calculating the minimum detectable concentration (MDC) of Ra-226 in characterization scans using a sodium-iodide detector, NaI(Tl), array coupled with a multi-channel analyzer.

The detector system used in this report is the Radiation Solutions Inc. (RSI), RS-700 Radiation Mapping System (RMS). The RMS consists of the RS-701 console, two 10.16 cm by 10.16 cm by 40.64 cm sodium iodide (NaI) crystals, a Trimble Ag global positioning system (GPS), a shielded trailer modified to carry the detectors, a laptop computer, and a locomotive device as shown in Figure 1. The detectors are oriented parallel to the ground and end-to-end with their narrow axis oriented in the direction of travel. The bottom of the detector casing is 12.55 cm above the ground and the gap between detectors is 0.32 cm.

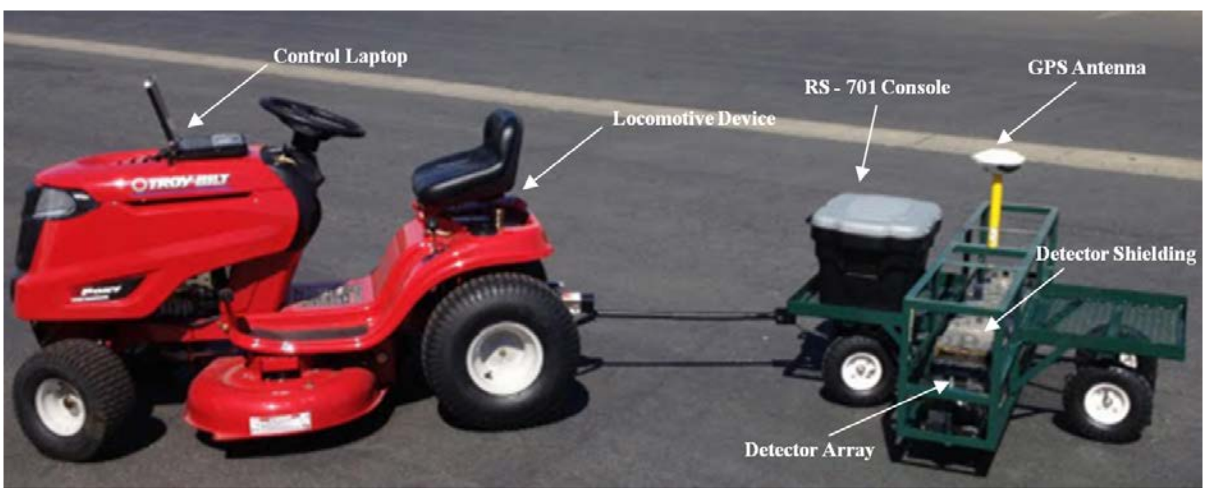

Fig. 1: Trailer mounted RMS system with locomotive device and gps.

The RMS records the operator's location to within 10 cm, in areas of open areas, and associates it with the spectral data from the detectors once per second. The system's position is determined by the onboard Trimble unit, configured as a rover, and another configured as a base station. The rover unit communicates to the base station via radio to determine position correction in real time. The RS-700 system uses internal multichannel analyzers (MCA) for each detector. Data is retrieved from the onboard laptop using RSI's RadAssist software and binned according to pre-specified regions of interest (ROI). The comma separated value (CSV) file is exported to facilitate post processing before being mapping using ArcGIS.

METHODS

Using progeny ingrowth as a detection proxy requires knowing decay time in order to determine progeny emission rates. Knowledge of site history, process, and prior survey results can be key in determining an accurate decay time. However, in the absence of such information in-situ measurements can be used to estimate decay time by comparing observed progeny emission ratios against established values. These values are determined using Oak Ridge National Laboratory's ORIGEN-ARP program to generate photon energy spectra for Ra-226 and its progeny at specified time intervals. ORIGEN-ARP is a computer program that performs nuclear irradiation and decay calculations with problem dependent cross sections [1].

A 1 Ci source of Ra-226 was input into ORIGEN-ARP to generate spectral data for various decay times. Due to program limitations the results include Bremsstrahlung production in water instead of soil, however this is an acceptable approximation. The total photon energy spectra for Ra-226 and its progeny over time is presented in Figure 2.

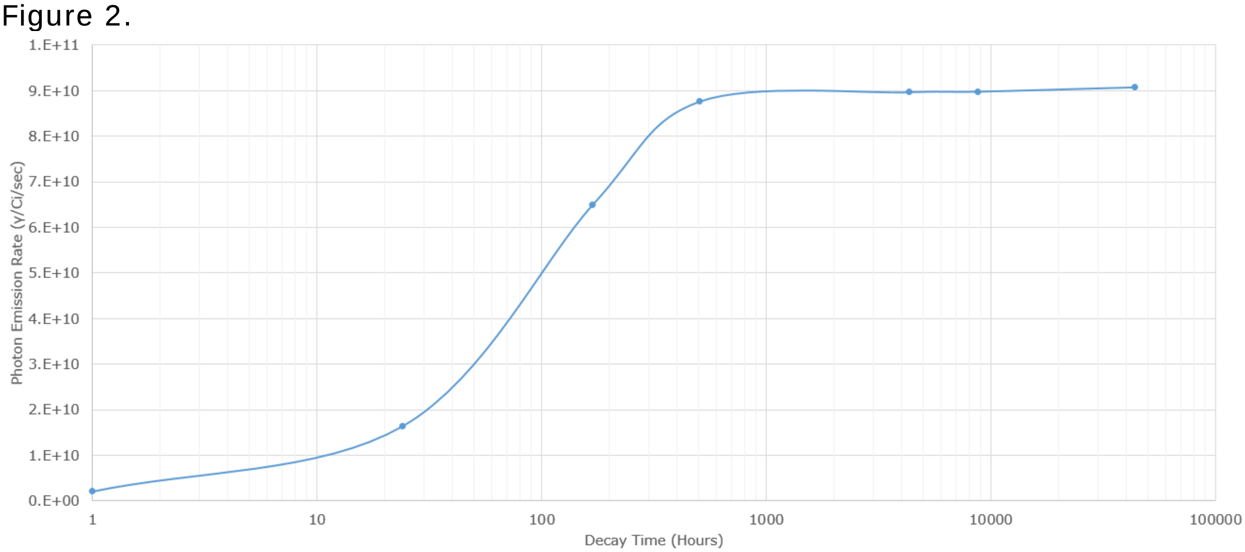

Fig. 2. Ra-226 progeny photon emission rate

The photon emission rate shows a strong dependence on decay time as Ra-226 progeny are produced and approach secular equilibrium at 21 days (98%). This corresponds with the 21-day ingrowth period used in EPA method 901.1 for a hermetically sealed matrix containing radium. Splicing the decay dataset into energy groups and plotting the results as a percentage of total emissions illustrated that the relative proxy photopeak ratios (Bi-214/Pb-214) are approximately constant after 24 hours, as shown in Figure 3. Ingrowth ratios for decay times less than 24 hours have a strong time dependence; however, it is unlikely that a survey scan of a new Ra-226 contaminated area will be performed within 24 hours. For the purposes of generating a Ra-226 progeny source term, a decay time of 21 days is assumed.

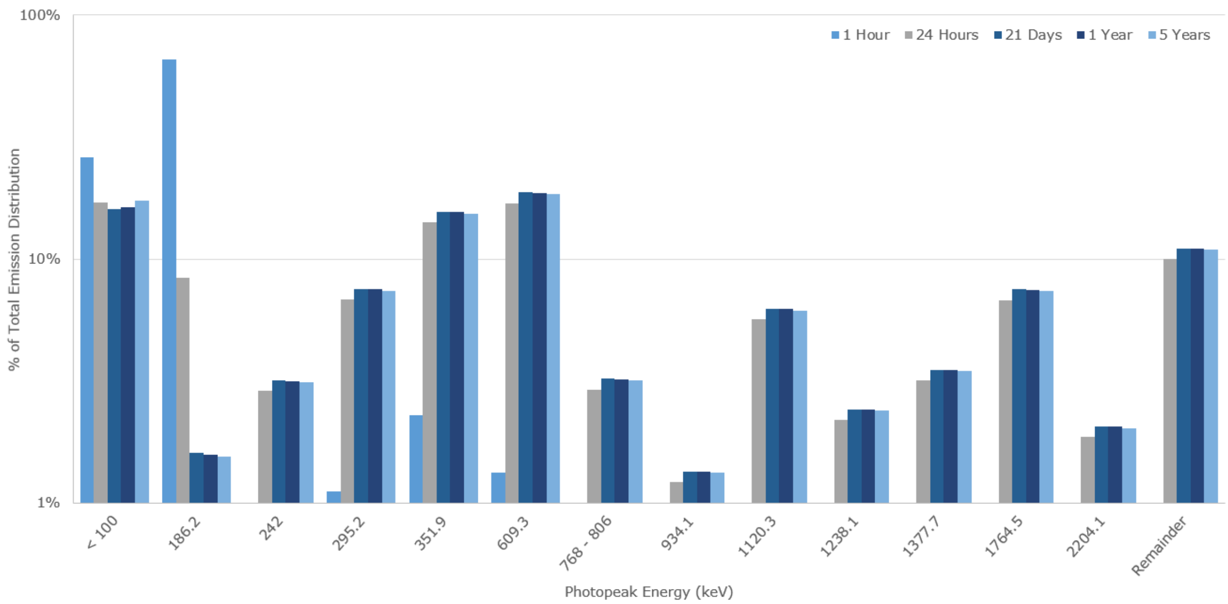

Fig. 3. Ra-226 progeny relative photon emission rates

Radiation Transport Modeling

The 21-day source emission spectra was input into Monte Carlo N-Particle (MCNP) transport code for modeling. Developed by Los Alamos National Laboratory, MCNP is a general-purpose computer program that can be used for neutron, photon, electron, or coupled transport. The program is frequently used for dosimetry, radiation shielding, detector design and analysis, decontamination and decommissioning [2]. MCNP was used to simulate the RMS response to various source geometries in order to determine the MDC and field of view for the RMS.

The simulation was designed to track the energy distribution of pulses created in the sodium iodine crystal. This is performed by recording the energy deposited in the crystal by each source particle and its secondary particles [2]. The simulation creates a bin for each source, thereby providing the spatial location of the source, and each bin contains the energy distribution of pulses created by that source, thereby measuring detector response. This is comparable to placing a source near the detector, measuring response, moving the source by a set increment, re-measuring the response, and repeating the process until the detector's field of view has been mapped. The results are processed into the detector field of view.

Simulation Geometry

The MCNP simulation was constructed using the detector and secondary shielding design specifications to create a problem geometry that radiation is transported through. The detector was modeled as a sodium iodine crystal, 10.16 cm by 10.16 cm by 40.64 cm incased in aluminum. The model assumes that the space between the crystal and the outer detector dimensions specified by RSI, 16.26 cm by 17.27 cm by 68.07 cm, is solid aluminum. This assumption is conservative because it increases the amount of self-shielding in the detector, thereby decreasing the sensitivity of the RMS. The secondary detector shielding consists of 0.16 cm of copper, 0.32 cm of lead, and 0.16 cm of carbon steel. This shielding wraps around 3 sides of the detector array shown in Figure 4 as orange, and primarily covers detector crystal, as light blue. The soil facing side of the shielding is a 0.32 cm layer of acrylic glass used to provide structural support while minimizing shielding in the direction of interest.

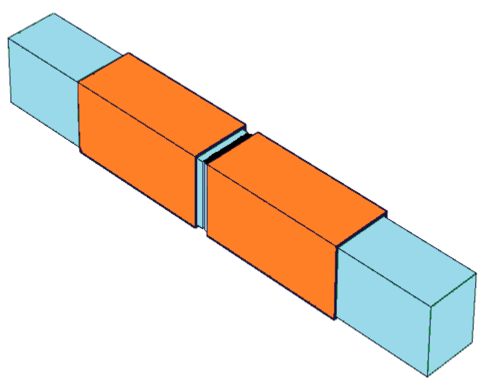

Fig. 4. Simulated isometric view of detector array

The detector array is positioned 12.54 cm above a layer of soil, as shown in Figure 5. The soil layer is 137.16 cm by 25.4 cm by 320.04 cm, and is assembled from a lattice of soil cells 15.24 cm wide, 5.08 cm thick, and 15.24 cm long. Each of these lattice cells contains a dime sized region that is used as geometric location of the source for the simulation, also illustrated in Figure 5. A dime sized source region was selected because volume is equivalent to 1 gram of soil. Emissions from these sources are tracked individually to map the spatial sensitivity of the detector array. Material compositions and densities were taken from the Pacific Northwest National Laboratory compendium [3].

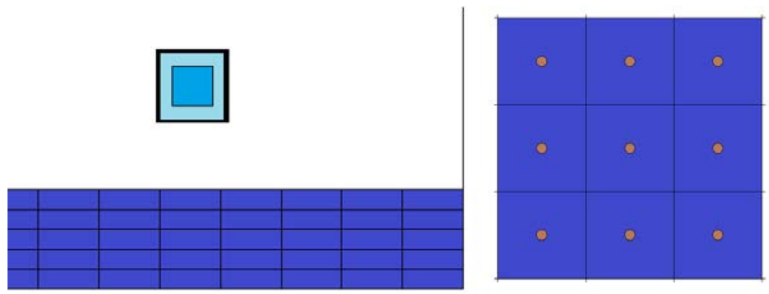

Fig. 5. (Left) Cross section of detector over soil (Right) Soil lattice with source

Preliminary Simulation Results

RMS static measurements of the site showed that, in addition to Ra-226 progeny, Th-232 progeny and K-40 were present and needed to be taken into account. Preliminary simulations detector response were plotted against a field measurement to determine if simulations accurately reflected field observations. The field measurement count data was normalized to the MCNP output for a qualitative comparison. Two simulations were plotted against the measurement, one using Ra-226 progeny emissions, and the other with Th-232 progeny emissions. Figure 4 illustrates the results from these preliminary efforts.

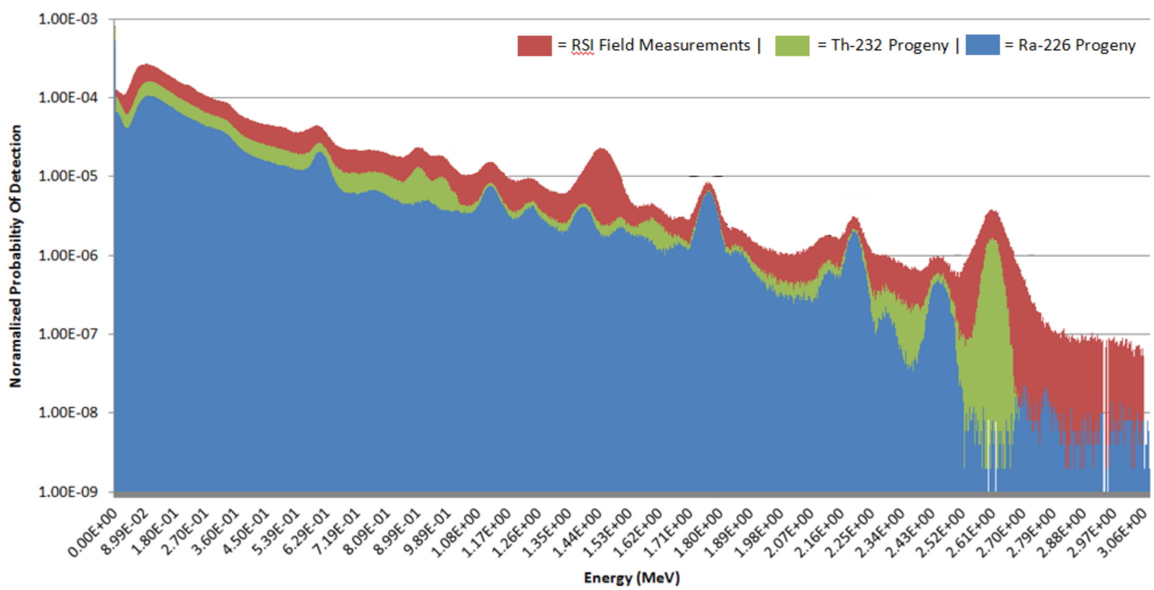

Fig. 4. MCNP simulated results overlaid with RMS static field measurement

K-40 was not simulated because based on comparison with the field measurement it will interfere with the Bi-214 1120 keV photopeak. Additionally, both the simulation and the field measurement show that the Pb-214 352 keV photopeak is indistinguishable from the Compton continuum. Therefore, only the Bi-214 609 keV and 1764 keV photopeaks can be used as a proxy for Ra-226 activity. Figure 4 also demonstrated that the simulation underestimated the full width half maximum (FWHM) of high energy photopeaks. The simulation calculates detector response using the F8 tally, and the FT GEB card is used to modify the Gaussian distribution used to estimate the FWHM of a detector system. This card was modified to more accurately match the observed FWHM observed in the RMS field measurement.

Minimum Detectable Concentration

RMS response to soil matrix contamination was calculated using the MCNP simulation described in previous sections. The output of the simulation requires post processing to calculate the RMS MDC in pCi per gram. This conversion requires the following additional inputs: scan speed of 50 cm per second, and a background count rates for the 609 keV and 1764 keV photopeaks and their standard deviations. Background data was taken from a previously established clean area of the survey site. RMS scan MDC supplants the surveyor efficiency coefficient in favor of a scan speed deviation factor [4]. This is acceptable because the RMS system utilizes computerized data collection for surveys (spatial and spectral); the only factor the surveyor influences is the scan speed of the system. The RMS MDC is calculated as using the following set of equations [6]:

$$MDC\left[\frac{pCi}{g}\right] = \frac{L_D\,[counts]}{K\left[\frac{counts*g}{pCi*sec}\right]*T[sec]*\epsilon_{speed}} \qquad \text{(Eq. 1)}$$

$$L_D = 3 + 4.65*\sqrt{C_{BKG} + 2\sigma_{bkg}}\,[counts] \qquad \text{(Eq. 2)}$$

$$K = SR\left[\frac{counts*g}{nps*photon}\right]*NPS*SF\left[\frac{photons}{Ci*sec}\right]*10^{-12}\left[\frac{Ci}{pCi}\right] \qquad \text{(Eq. 3)}$$

$$T = \frac{10.16\,[cm]}{50\left[\frac{cm}{sec}\right]} \qquad \text{(Eq. 4)}$$

where L_D = Detection Limit established with a 0.95 detection probability and a probability of 0.05 false positive [5], T = Observation Interval duration source is directly underneath the detector, C_{BKG} = Area Background field measured detector response in previously established clean area, K = Instrument Response calculated instrument response to source, SR = Simulation Result, output of MCNP tally for the simulation, NPS = Number of particle histories run during the simulation, SF = Source Factor from ORIGEN-ARP photon emission rate for 1 Ci Ra-226 decayed for 21 days,

ε_{speed} = Scan Speed Deviation two sigma deviation from desired scan speed: $0.78 = \sqrt{Speed}/(\sqrt{Speed}+2)$.

DISCUSSION

Figure 5 illustrates the calculated field of view for the RMS, where the overlaid orange box and black box are the positions of the detector crystal, and detector array respectively.

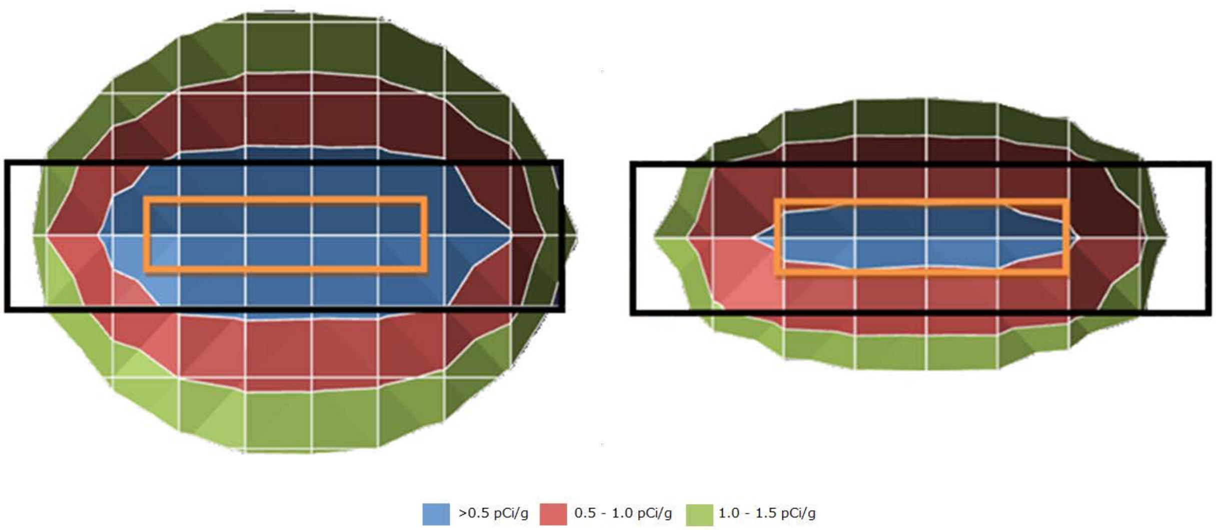

Fig. 5. RMS MDC field of view | (Left) Surface Soil | (Right) 5.08 cm Soil Depth

Table I displays the results of the simulated MDC calculation and the California Radiologic Health Branch (CRHB) [7]. The CRHB operated the RMS using the gross energy bounds 45 keV - 1980 keV and a Ra-226 decay time of 1 year. To determine the system's spatial efficiency, the CRHB placed a Ra-226 point source under the detectors at ground level and moved in increments of 10 centimeters until a field of 1 square meter was measured. MicroShield was used to model a Ra-226 source distributed over the over one square meter to a depth of 15 cm. Detector responses were calculated by dividing the experiential count rates by the MicroShield fluence. The CRHB estimated a lower MDC than the simulated result; however, the CRHB result uses a gross gamma count rather than energy specific photopeaks. Therefore, the CRHB Ra-226 estimate will be more sensitive to the presence of other radioactive materials.

TABLE I. Summary of RMS MDC (pCi/g)

	Simulated MDC			CRB MDC
Soil Depth (cm)	Minimum	Direct Crystal Passover	Detector Array Passover	Minimum
0	0.22	0.50	2.5	0.32
5.08	0.38	0.50	2.5	
10.16	0.74	1.0	4.5	
15.24	1.46	3.0	9.0	
Average	0.70	1.25	4.63	

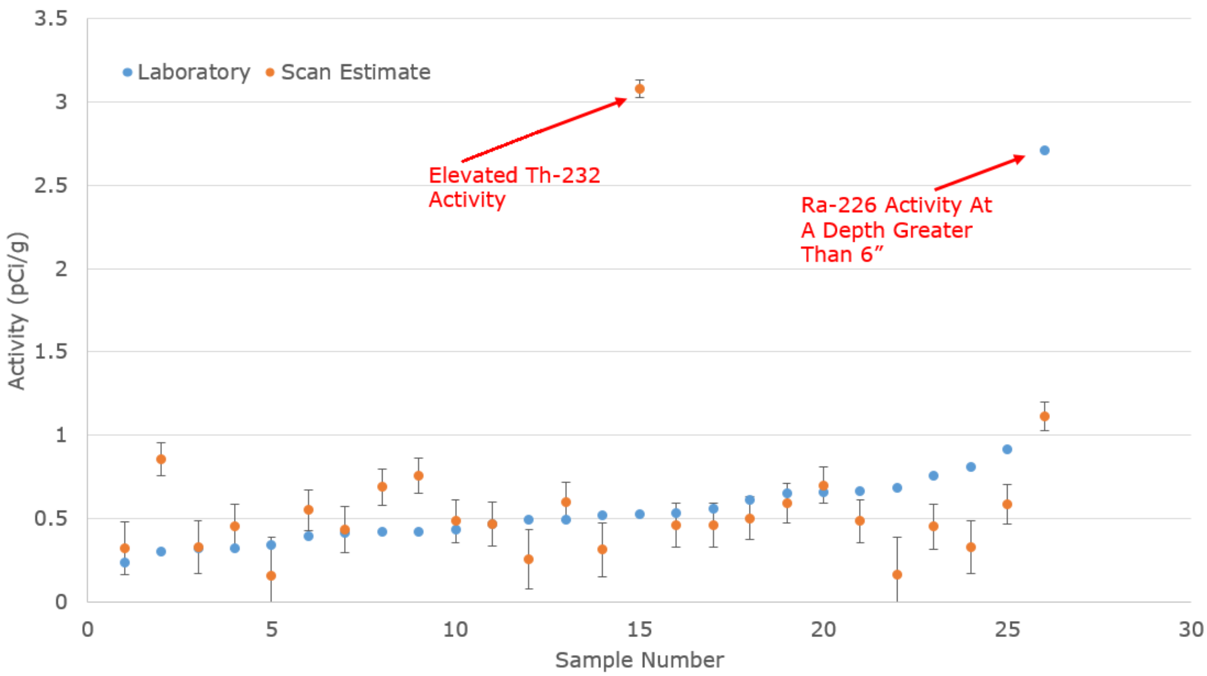

Fig. 6. Laboratory Ra-226 activity results against RMS scan estimates

The laboratory results demonstrated that scanned area was principally comprised of soil containing less than 1 pCi per gram Ra-226. Although this borders the system MDC, it underestimated the concentration of radium in the soil. There are two probable causes of this: escape of radon gas from the surface soil, and uneven distribution of activity as a function of soil depth. Radon is the daughter of radium, and has a high mobility in soil. If the gas escapes, then the progeny used as the detection proxy will be present in lower than expected concentrations. Figure 3 illustrated the high dependence of activity depth on detection capabilities. Ra-226 concentrations were calculated using the average of probability of photon detection over the 15.24 cm soil depth. If there is less activity in the surface soil than at depth, then the RMS will underestimate the true concentration.

CONCLUSIONS

Radionuclide identification using high volume sodium-iodide crystals in low level characterization scans is demonstrated to be possible. Further system refinement will focus on developing a methodology to estimate a radon escape factor for site specific use. Accounting for radon escape in the top 15.24 cm of soil will allow the system to better estimate Ra-226 concentrations in soil. This is a significant advancement over the gross counting statistics used in most studies to satisfy MARSSIM survey requirements, because it will increase stakeholder confidence.

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