A Method for In Situ Depth Profiles of Alpha and Beta Contaminants in Soil Using Scintillators and Fiber Optic Light Guides

D.M. Hamby and J.R. Zometsky*

Department of Environmental and Industrial Health
School of Public Health
University of Michigan
Ann Arbor, MI 48109-2029

ABSTRACT

This work examines the potential use of two scintillation detectors for in-situ measurements of alpha and beta particles at various depths in soil. Use of an organic (plastic) scintillator was investigated for beta-particle measurements and a ZnS(Ag) inorganic scintillator was investigated for alpha-particle measurements. Each detector was connected to a photomultiplier tube by varying light-guide lengths of 20 cm, 50 cm, and 100 cm. Soil standards, each containing a known quantity of a single radionuclide, were prepared to characterize the detectors. Efficiency of the alpha and beta detectors ranged from 0.0021 to 0.0041 cps/Bq/g and 0.064 to 0.18 cps/Bq/g, respectively. Detection limits (L_D) for a 15 minute count ranged from 0.33 to 1.6 Bq/g and from 8.1 to 15 Bq/g for the beta and alpha detectors, respectively. Resolution in the alpha detector was poor with values ranging from 85 to 150%. Detectors of this type have the potential advantage of providing depth-profile information in situ, but would generally be useful only in their estimation of gross alpha or gross beta activity.

* Current address: U.S. Department of Agriculture, Washington, DC

INTRODUCTION

There are many waste sites around the country managed by the U.S. Department of Energy which contain radioactive contamination in excess of current limits. The decommissioning of commercial power reactors, DOE sites, and other nuclear facilities requires accurate, efficient, and cost effective methods of site assessment. Traditionally, soil characterization has involved direct measurements of soil samples in a laboratory using gamma spectroscopy or alpha and beta quantification following some form of radiochemical sample preparation. The method of soil sampling, radiochemical procedures and subsequent counting could be quite cost prohibitive and time consuming since many samples normally require analysis, even in moderately sized areas. In most cases, in-situ field measurements provide a more cost effective means of site characterization. Similarly, field measurements also have the advantage of providing real-time results when determining soil radionuclide concentrations.

The decommissioning and decontamination (D&D) process requires site characterization before, during, and at the end of the decontamination efforts. Measurements are required before remediation activities begin in order to provide an initial characterization of the extent of contamination. During the decommissioning process, characterization is needed to determine what further decontamination efforts are required. And, each site must be assessed after remediation in order to ensure regulatory compliance for free release of the site. By characterizing soil in situ at various depths, one can determine the full extent of contamination during the initial stages of site assessment, thereby reducing or even eliminating the need for soil measurements during the D&D process.

Within the past decade, there have been technological advances in the area of in-situ alpha and beta detection and measurements. Schilk et al. (1994) have developed a beta detector that uses scintillating fibers to measure ^{90}Sr/Y and ^{238}U on the ground surface by detecting the high energy beta particles emitted by their daughter products ^{90}Y and ^{234m}Pa, respectively. MacArthur et al. (1992) have developed the Long Range Alpha Detector (LRAD) that detects ions created by alpha interactions in air. The LRAD transports the ions either by an air current or by electrostatic forces into an ionization chamber where they are measured. The LRAD detector can be used for site characterization purposes on flat soil surfaces (MacArthur et al. 1993). Segovia et al. (1990) have placed solid state nuclear track detectors (SSNTD) (cellulose nitrate foils) beneath the surface to measure radon concentrations in soil gas. Hutter (1995) has used stainless steel tubes inserted into the ground to obtain soil gas samples and measure thoron (Rn-220) concentrations in ZnS(Ag) scintillation cells located at the surface. Hsu et al. (1991) placed calcium sulfate ($CaSO_4$:Dy) thermoluminescent dosimeters in the soil to measure exposure rates beneath the surface. Even with these advances, however, research in the area of in situ environmental assessment is still in its infancy.

This work demonstrates the potential effectiveness of two scintillation detectors for real time, in-situ quantification of alpha and beta emitters in soil. It employs the use of an organic plastic scintillator for beta detection and an inorganic ZnS(Ag) scintillator for alpha detection. The scintillators are coupled to a photomultiplier tube by fiber optic light guides of varying length for generating depth profiles in soil.

MATERIALS AND METHODS

Two scintillation detector systems were examined to assess their usefulness in measuring alpha and beta contaminants at various depths in soil. Organic scintillators are typically used for beta particle detection, and, due to their low Z constituents, they have an extremely low photoelectric cross-section resulting in a very low detection efficiency for gamma rays. Additionally, electron backscatter is reduced because of the low density material. The beta detector sub-assembly, therefore, is made of an organic plastic scintillator (Bicron model BCF-12) 3 mm in diameter by 50 mm long and covered in reflective mylar. On one end of the sub-assembly there is a threaded coupling to connect the scintillator to a light guide.

The alpha detector (Bicron model BCF91A-ZNS) is made of wave shifting fibers covered with 8 mg/cm^2 of ZnS(Ag) and then covered with reflective mylar. Its dimensions are also 3 mm dia. x 50 mm in length and it, too, has the threaded coupling on one end. Silver-activated zinc sulfide is used primarily for the detection of alpha particles or other heavy ions. Thicknesses above approximately 25 mg/cm^2 cannot be used due to the opacity of the crystal to its own luminescence wavelength (Knoll 1989). A wave shifting fiber is used with this detector to re-radiate the absorbed light from the scintillator at a different wavelength since the PM tube is more sensitive to a wavelength other than that given off by the ZnS(Ag) scintillator.

Scintillators were coupled to 3 mm dia. fiber optic light guides 20, 50, or 100 cm in length and sheathed in light-tight coverings. Light guides typically have a high index of refraction in order to minimize the critical angle and, thus, also serve to maximize the internal reflection of the scintillation light (Knoll 1989). One end of the light guide is fitted with threads to connect with the scintillator sub-assembly. The other end is flat and slides into a tightening ring making direct contact with the photocathode of the PM tube. The phototube (Hamamatsu model R628) is 28.6 mm in diameter and sheathed in an aluminum housing of 3.5 cm diameter and 18.4 cm in length. The PMT voltage divider is integral to the tube design.

Both the organic and ZnS(Ag) scintillators were characterized and calibrated using soil standards that were prepared in our laboratory according to the procedure by Sill and Hindman (1974). These soil standards were tested for homogeneity by the one-way analysis of variance method. The detectors were characterized for efficiency, accuracy and precision, and minimum detectable activity. The alpha detector was also characterized for spectroscopy resolution. Accuracy and precision measures were determined with soil standards obtained from the National Institute of Standards and Technology (NIST) and the US Environmental Protection Agency (EPA).

Efficiency. Absolute efficiency, being dependent on counting geometry, was calculated for both detectors with each of the three light guides in place. The detectors were inserted into our soil standards, giving a consistent geometry for all measurements. The dependence of energy on efficiency was also determined. The sources used to determine the efficiency of the organic scintillator were ^{134}Cs, ^{90}Sr/Y, and ^{144}Ce/Pr, and those used to determine the efficiency of the ZnS(Ag) detector were ^{239}Pu, ^{243}Am, and ^{226}Ra. Each soil standard and background standard was counted five times for equal counting times. These counts were averaged and the net count rate was used in the efficiency calculation. Because of the amount of activity in each sample, the alpha counting intervals were 15 minutes each and the beta counting intervals were 30 minutes each. The average beta energies were used in the determination of energy dependence, and both the beta and alpha energies were weighted and averaged by the radiation yields. The ^{90}Sr/Y and ^{144}Ce/Pr energies were also averaged according to their secular equilibrium relationship. Since these detectors respond to the ^{226}Ra daughter radiations, the ^{226}Ra energy was specified as the average energy of ^{226}Ra and its daughter products.

Accuracy and Precision. Measurements were made for both detectors and all three light-guide lengths in two different soil types. The detectors were tested in contaminated soils obtained from the U.S. National Institute of Standards and Technology (NIST) and from the U.S. Environmental Protection Agency (EPA). The NIST soil standard was originally obtained from the U.S. DOE's Rocky Flats Site and contains radionuclides included in the U-238 and Th-232 decay series. The EPA soil standard contains monazite ore and, therefore, also contains the Th-232 decay products.

Minimum Detectable Activity. Currie (1968) and Lochamy (1981) each have defined successive counting limits describing the concept of minimum detectable activity (MDA). These limits refer to various degrees of confidence when reporting the activity that is detectable in a given sample by a particular detector. The most common limit used to describe the MDA in a low-level counting system is the detection limit, L_D. For equal counting intervals of background and the sample, the detection limit, in units of counts, can be calculated using,

$$L_D = 2.71 + 4.65\sqrt{B}, \tag{1}$$

where B is the mean number of background counts for a given system configuration, whether it be total counts or counts in a particular interval of a multichannel analyzer (MCA). The detection limit is specified for a given detection system or analytical procedure, and is the point at which a signal is almost certain to be detected. This form of the MDA is usually referred to as the Lower Limit of Detection (LLD). The number of counts given by the detection limit can be related to concentration, C_{LLD} (in Bq/g), by:

$$C_{LLD} = \frac{L_D}{\varepsilon T}, \tag{2}$$

where ε is the detector efficiency and T is the effective counting interval and is equal to $(1-e^{-\lambda t})/\lambda$, where t is the counting time. For long-lived radionuclides, relative to the counting interval, T is essentially equal to t.

Resolution of the Alpha Scintillator. Resolution was determined from the differential pulse height distribution obtained from the spectral output of the MCA for the alpha ZnS(Ag) probe. Detector energy resolution, R, is defined as:

$$R = \frac{FWHM}{H_0} \tag{3}$$

where FWHM is the width of the full-energy peak at half its maximum amplitude, and H_0 is the peak centroid (Knoll 1989). Both the FWHM and the peak centroid are expressed as MCA channel numbers. Energy resolution is a dimensionless fraction typically expressed as a percentage. For consistency, the spectra containing the maximum number of counts were used for the resolution calculations. Resolution of the beta detector was not determined since the incident beta spectra were not monoenergetic.

RESULTS AND DISCUSSION

Efficiency. Absolute efficiencies for both detectors and all three light-guide lengths are given in Table 1. Since the detectors are placed into soil containing a contaminant of known concentration, efficiency is given in units of net count rate per unit concentration, i.e. cps/Bq/g. Plots of efficiency as a function of energy for all detector configurations are given in Figs. 1 and 2. Efficiency increases proportionally with increasing energy at all three light guide lengths for the beta detector, whereas efficiency generally decreases with increasing energy for the alpha detector. Figs. 3 and 4 give a graphical representation of how efficiency changes with the light guide length. Efficiency of the beta detector remains constant over all light-guide lengths, however, the efficiency of the alpha detector appears to increase from 20 to 50 cm and then decrease from 50 to 100 cm.

The dependency of efficiency on energy is based on the attenuators of soil, air and mylar through which the particles must pass before reaching the detector. That is, the higher energy results in a greater range of the incident radiation, allowing a larger number of particles to pass through the soil and reach the detector with enough energy to induce a signal. The detector efficiencies are dependent on the detector size and geometry in that a larger detector with greater surface area would intercept more alpha and beta particles, resulting in increased absolute efficiency. The beta detector efficiencies are significantly greater than the alpha detector efficiencies due to the low LET nature of the beta particles and their longer range through soil, resulting in a larger effective sampling area.

Accuracy and Precision. Accuracy and precision results are given in Table 2 for the EPA and NIST soil standards. Cember (1983) states that an accuracy of ±200% is acceptable for most radiation detection instruments when levels being measured are less than 10% of the appropriate standard, however, when levels approach the standard, an accuracy of ±30% is expected. As seen in Table 2, measurements of the EPA soil standard generally meet the more liberal criteria for alpha quantification and meet the more restrictive limit for beta quantification. Measurements using the NIST standard, however, were much less accurate, yet very conservative. Although the accuracy of the two scintillating detector systems is questionable, precision or reproducibility for both detectors is very good. The propagated error from three measurements in each configuration is quite small, showing less than about 25% and 10% variability for alpha and beta measurements, respectively. “Certified” concentrations in Table 2 are as provided by EPA and NIST and “measured” concentrations were those determined using the alpha and beta scintillators. Efficiency, being energy dependent, was estimated by interpolation of the data in Figs. 1 and 2 for the weighted mean energies of beta or alpha emitters in each soil sample.

The comparison of certified and measured concentrations is highly dependent on the efficiency used to determine the measured value. As stated above, the efficiency in these detectors is energy dependent, therefore, it would be expected that the use of a single efficiency value would bias the result. It is quite clear that the accuracy of both the alpha and beta systems is markedly worse in the analysis of the NIST soil relative to the EPA soil. Contamination of the detector element between measurements was controlled by conducting background measurements between each source analysis. Hot spots in the soil standards are possible; even though each sample was thoroughly mixed prior to analysis, the physical size of the scintillator and the source volume being analyzed are both very small, thus magnifying the effects of inhomogeneities. A complex spectra of incident alpha and beta particle energies may also affect the response of these scintillators, making a single efficiency factor all the more difficult to use. As stated earlier, interpolation of the data in Figs. 1 and 2 resulted in efficiency estimates as a function of the weighted average beta and alpha energies present in the two soil standards. The functions used to estimate beta efficiency were fairly well defined, however, those used to determine alpha efficiencies were extremely varied.

Minimum Detectable Activity. The values in Table 3 show that the detection limit (L_D) for the beta detector is between 0.33 and 1.6 Bq/g depending on the beta particle energy being measured, a counting time of 15 minutes, and the level of statistical accuracy desired. The beta detection limit is constant with depth (light-guide length) and decreases with increasing average beta energy. The detection limit (L_D) for the alpha detector ranges from 8.1 to 15 Bq/g and also appears to be fairly consistent with light-guide length. The alpha detection limit with this detector configuration is shown to vary greatly with energy. Detection limits in terms of activity are dependent on detector efficiency according to Eqn. 2.

Resolution of the Alpha Scintillator. Resolution estimates for each light-guide length and the ZnS(Ag) scintillator are given in Table 4. The resolution of the alpha detector is quite large. Generally, two energies that are separated by more than one value of the detector FWHM can be resolved and analyzed (Knoll 1989). As can be seen in Table 4, however, the alpha energies would have to be separated by 85% to 150% in order for this system to distinguish between two energies and be used as a radiation spectroscopy system. In addition to the generally poor spectroscopic qualities of scintillators, energy straggling of the alpha particles contributes to the lack of resolution. Since the energy loss of an alpha particle along its track is a stochastic process, a range of energies results as the alpha particles pass through the soil, air, and mylar absorbers. The energy straggling increases with distance between the source and the detector and with increased absorber thickness.

CONCLUSIONS

The absolute efficiency of the scintillation detector systems are dependent on radiation type, incident energy, light-guide length, and detector surface area. Although the intrinsic efficiency is high, the physical size of the scintillators limits the absolute efficiency to between 0.064 and 0.18 cps/Bq/g for the organic plastic and between 0.0021 and 0.0041 cps/Bq/g for the ZnS(Ag) scintillator. The efficiency of the beta detector is quite good, it increases with energy and is very stable over the light-guide lengths tested. The efficiency of the alpha detector, however, is very low and irregular with alpha energy and light-guide length. The alpha detector's very low efficiency limits its use for field measurements.

While the precision of both the alpha and beta detectors is high, their accuracy is poor. High precision is a reflection of stable operating characteristics and a sign of reproducibility between measurements. However, poor accuracy will certainly limit their use in the field unless a method for utilizing an efficiency function can be developed. This function would be energy dependent and would be more indicative of detector response. The lack of accuracy of measurements in this study is very likely the result of areas of high activity (hot spots) within the soil standards.

The lower limits of detection estimated for both the alpha and beta scintillator systems are relatively low for in situ field screening, yet they are several orders of magnitude higher than levels achievable with sophisticated laboratory-based analyses. If used as a screening tool, however, simple gross alpha or gross beta quantification may be appropriate for determining whether the concentration at a given location is near the applicable standards.

Like most other scintillators used for beta detection, spectroscopy would be quite difficult, if not impossible. Likewise, resolution with the alpha probe is not of the quality needed for nuclide identification. The lack of resolution essentially prohibits the use of this detector configuration for spectroscopy. The two systems do, however, work well for quantification of gross alpha or beta, assuming that the contamination level is high enough to result in reasonable counting times, given the very low efficiency of the alpha detector.

In very simple field trials, it was determined that the instruments perform well, with a marked dependency of ambient temperature on performance of the photomultiplier tube. The longer light guides, however, are unstable and a deployment device is necessary to permit field use with consistent geometry and protection against probe contamination. Additionally, the reflective mylar coverings lack the ruggedness necessary for field work.

The results of our investigation suggest that these scintillators may be an effective means for in situ quantification of total beta and total alpha contamination as a function of depth in soils. A lack of efficiency and resolution in the alpha detection system (ZnS(Ag)), however, limits its usefulness as an in situ device at low concentrations. Before field use can be realized, both detectors require further investigation into their response characteristics for radiation types other than that for which they are designed, e.g., gamma or alpha response in the organic scintillator.

About the Authors

David M. Hamby is currently an Assistant Professor of Radiological Health at the University of Michigan's School of Public Health, He received the PhD and MS degrees from the Department of Environmental Sciences and Engineering at the University of North Carolina, Chapel Hill. Dr. Hamby has worked as a Principal Research Scientist in the Environmental Technology Division for Westinghouse Savannah River Company.

John R. Zometsky is a graduate of the Radiological Health program with an MS in Environmental Health from the UM School of Public Health. This paper is based on a major portion of his Master's thesis under the direction of Dr. Hamby. Mr. Zometsky is currently employed by the U.S. Department of Agriculture in Washington, DC.

REFERENCES

Cember H. Introduction to Health Physics. 2nd ed. New York: Pergamon Press. 1983.

Currie L.A. Limits for Qualitative Detection and Quantitative Determination. Analytical Chemistry, vol. 40, no. 3, pp. 586-93, March 1968.

Hsu P.C., Weng P.S., Wang H.E., and Tseng P.W. Monitoring Underground Radiation Fields Using Powdered Calcium Sulfate Thermoluminescent Dosimeters. Health Physics, vol. 61, no. 3, pp. 367-71, September 1991.

Hutter A.R. A Method for Determining Soil Gas Rn-220 (Thoron) Concentrations. Health Physics, vol. 68, no. 6, pp. 835-39, June 1995.

Knoll G.F. Radiation Detection and Measurement. 2nd ed. New York: John Wiley & Sons, Inc. 1989.

Lochamy, J.C. The minimum detectable activity concept. EG&G Ortec System Application Studies. PSD No. 17; 1981.

MacArthur D.W., Allander K.S., Bounds J.A., Butterfield K.B., and McAtee J.L. Long-Range Alpha Detector. Health Physics, vol. 63, no.3, pp. 324-30, September 1992.

MacArthur D.W., Allander K.S., Bounds J.A., Catlett M.M., Caress R.W., and Rutherford D.A. Alpha Contamination Monitoring of Surfaces, Objects, and Enclosed Areas. IEEE Transactions on Nuclear Science, vol. 40, no.4, pp. 840-5, August 1993.

Schilk A.J., Knopf M.A., Thompson R.C., Hubbard C.W., Abel K.H., Edwards D.R., and Abraham J.R. Real-time, in situ detection of Sr-90 and U-238 in soils via scintillating-fiber-sensor technology. Nuclear Instruments & Methods in Physics Research, Section A, vol. 353, no.1-3, pp. 477-81, 30 Dec. 1994.

Segovia N., Gaso M.I., Chavez A., Tejera A., Gutierrez A., Azorin J., Balcazar M., Lopez A., Tamez E., Cervantes L., and Monnin M. Environmental Alpha and Gamma Survey. Radiation Protection Dosimetry, vol. 34, no.1/4, pp. 219-22, 1990.

Sill C.W., and Hindman F.D. Preparation and Testing of Standard Soils Containing Known Quantities of Radionuclides. Analytical Chemistry, vol. 46, no.1, pp. 113-18, January 1974.

FIGURE CAPTIONS

Fig. 1. Efficiency as a function of energy for the beta scintillator.

Fig. 2. Efficiency as a function of energy for the alpha scintillator.

Fig. 3. Efficiency as a function of light-guide length for the beta scintillator.

Fig. 4. Efficiency as a function of light-guide length for the alpha scintillator.

Table 1. Absolute efficiency (cps/Bq/g) as a function of light guide length.

	Energy (MeV)	20 cm	50 cm	100 cm
^{134}Cs	0.157	0.075	0.070	0.064
$^{90}Sr/Y$	0.565	0.181	0.181	0.177
$^{90}Sr/Y$†	0.565	0.180	0.180	0.177
$^{144}Ce/Pr$	0.645	0.326	0.327	0.314
^{226}Ra	5.96	0.0023	0.0024	0.0021
^{239}Pu	5.15	0.0037	0.0041	0.0031
^{243}Am	5.27	0.0030	0.0030	0.0030
^{243}Am†	5.27	0.0030	0.0030	0.0030

†second sample, different concentration

Table 2. Accuracy/precision results.

Detector	Light Guide Length (cm)	Net Count Rate (cps)[†]	Efficiency (cps/Bq/g)	Certified Conc. (Bq/g)	Measured Conc. (Bq/g)	Difference (%)
EPA Soil Standard						
Beta (weighted avg. energy = 300 keV)						
	20	2.1 (0.09)	0.088	22.2	24	8.1
	50	2.2 (0.09)	0.082		27	22
	100	2.2 (0.08)	0.079		28	26
Alpha (weighted avg. energy = 6.0 MeV)						
	20	0.27 (0.03)	0.0022	33.3	120	260
	50	0.24 (0.05)	0.0024		100	200
	100	0.18 (0.02)	0.0019		95	190
NIST Soil Standard						
Beta (weighted avg. energy = 450 keV)						
	20	1.1 (0.08)	0.12	1.18	9.2	680
	50	1.2 (0.09)	0.11		11	830
	100	0.80 (0.10)	0.11		7.3	520
Alpha (weighted avg. energy = 5.7 MeV)						
	20	0.069 (0.02)	0.0030	0.748	23	3000
	50	0.081 (0.01)	0.0027		30	3900
	100	0.087 (0.02)	0.0034		26	3400

[†]one standard deviation counting error (3 samples)

Table 3. L_D values in concentration units (15 minute count time).

		Lower Limit of Detection (Bq/g)*		
	Energy	20 cm	50 cm	100 cm
Beta				
^{134}Cs	0.210	1.5 (0.04)	1.5 (0.03)	1.6 (0.03)
$^{90}Sr/Y$	0.934	0.62 (0.02)	0.60 (0.01)	0.59 (0.01)
$^{144}Ce/Pr$	1.22	0.34 (0.01)	0.33 (0.01)	0.33 (0.01)
Alpha				
^{226}Ra	4.78	15 (0.8)	14 (0.7)	14 (0.9)
^{239}Pu	5.16	9.1 (0.5)	8.1 (0.4)	9.7 (0.6)
^{243}Am	5.28	11 (0.6)	11 (0.6)	10 (0.6)

*mean of 5 measurements; value in parentheses is one standard deviation about the mean.

Table 4. Measured energy resolution (percent) for the alpha scintillator.

Nuclide	20 cm	50 cm	100 cm
^{243}Am	132	151	148
^{239}Pu	93.3	104	85.2
^{226}Ra	96.6	96.2	93.1