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A method for determining the analytical form of a radionuclide depth distribution using multiple gamma spectrometry measurements

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ABSTRACT

When characterizing environmental radioactivity, whether in the soil or within concrete building structures undergoing remediation or decommissioning, it is highly desirable to know the radionuclide depth distribution. This is typically modeled using continuous analytical expressions, whose forms are believed to best represent the true source distributions. In situ gamma ray spectroscopic measurements are combined with these models to fully describe the source. Currently, the choice of analytical expressions is based upon prior experimental core sampling results at similar locations, any known site history, or radionuclide transport models. This paper presents a method, employing multiple in situ measurements at a single site, for determining the analytical form that best represents the true depth distribution present. The measurements can be made using a variety of geometries, each of which has a different sensitivity variation with source spatial distribution. Using non-linear least squares numerical optimization methods, the results can be fit to a collection of analytical models and the parameters of each model determined. The analytical expression that results in the fit with the lowest residual is selected as the most accurate representation. A cursory examination is made of the effects of measurement errors on the method.

1. Introduction

This paper introduces an innovative method designed to determine the one-dimensional radionuclide depth distribution in a media. When performing in situ gamma spectroscopy, one of the unknowns with the greatest impact on the measured activity is the depth distribution. It may itself inform both the optimal approach and cost of remediation and decommissioning work. For example, tasks could involve radioactive contamination migrating in cracks through concrete in structures being decommissioned, or the selection of excavation techniques in the case of soil contamination. Furthermore, it can be used to determine the total effective dose to individuals from environmental radionuclides (Beauvais et al., 2009). Although in many cases the horizontal source distribution is either known or constant over a large area (Sowa and Martini, 1989; Zombori and Andrasi, 1992; MacDonald et al., 1997), it is often unknown (Rybacek et al., 1992; Benke and Kearfott, 2002).

The depth distribution can be measured directly using soil sampling (EML Procedures Manual, US DoE, 1997). Estimates can also be made based upon the past soil history or based upon soil samples taken from depths at other locations. However, such results are often highly inaccurate or impossible if the site history is unknown. Furthermore, this measurement method presents additional challenges that make accurate determination difficult (Benke and Kearfott, 1999a,b, 2000).

An alternative to soil sampling is in situ measurement, which yields data within ±15% of the soil sample results (Benke and Kearfott, 1997). For the multiple photopeak method of determining depth distribution, the attenuation of various energy photons from a single radionuclide (Sowa and Martini, 1989; Beck et al., 1972; Karlberg, 1990), or the primary photopeak and the X-ray lines (Rybacek et al., 1992), are used with information about attenuation coefficients to determine the average photon path length through the media and thus estimate source depth. This method is limited to radionuclides that emit photons of two or more energies. The number of unknowns that can be determined in

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an assumed analytical representation of the distribution, shown as an exponential distribution for the examples presented, is limited by the number of photons.

Other investigators proposed using the ratio of the photopeak to the adjacent scatter to determine a planar source depth (Zombori and Andrasi, 1992; Tyler and Sanderson, 1996). The method relies on a single ratio, therefore it is possible to determine only one unknown in an analytical expression. Because the detection of scattered radiation requires higher activity and may be affected by interfering photopeaks with scattered photons in the same valley region as the radionuclide of interest, the method proved limited (Tyler, 1999). The addition of a cylindrical collimator to the method, however, was of some benefit (Chesnokov et al., 1997; Golosov et al., 2000).

A single lead disk was placed in varying distances in front of a detector to vary the detector field of view (FOV) and obtain measurements dependent upon the depth distribution which could then be fit to a Gaussian curve (Korun et al., 1994). This fundamental idea could be further improved by modifying the design so that the detector response depended even more strongly with the depth dose distribution. This was achieved through an in situ gamma spectrocopic system design consisting of cylindrically symmetric collimators, each with a differing FOV (Benke and Kearfott, 2001). For that system, the detector responses for each collimator and geometry were calculated using In Situ Object Counting System (ISOCS) software.[1] The method was used to determine the depth of a variety of planar sources and to estimate the uniform source distribution within several material layers. The method was tested in a laboratory environment for a limited number of source configurations (Benke and Kearfott, 2002) and in the field at Hanford, Washington (Van Riper et al., 2002).

Much prior work directed at determining radionuclide depth distributions assumed a distribution shape characterized by a specific mathematical function and fit the parameters for that function. The calibrated responses of in situ detectors for those approaches are typically determined by using a pre-determined analytical model of the distribution (EML Procedures Manual, US DoE, 1997; Helfer and Miller, 1988; Faller, 1992; Whetstone et al., 2011). The distribution that is selected is based upon historical knowledge of the site, previous sampling results at the location or similar locations, the chemical and physical radionuclide characteristics, and the site's properties. There is no fixed procedure for distribution selection and it is often based upon user knowledge and software limitations. A poor distribution characterization can lead to inaccurate sampling results (Sowa and Martini, 1989; Zombori and Andrasi, 1992; Rybacek et al., 1992).

This paper presents a method that can be employed to select the analytical model that best represents the true radionuclide distribution. After obtaining a series of measurements with varying detection system geometries, it is possible to determine the analytical expression that most closely matches the source distribution present. Each geometry has a response that can be calculated theoretically (Whetstone et al., 2011) and is directly dependent upon the source distribution. Once a series of measurements is taken, a system of non-linear equations is developed for each of several analytical models selected. Using non-linear optimization methods, each set of non-linear equations is solved for its unknown parameters. The measured data are then fit to each analytical model under consideration. The analytical model that represents the closest fit to the measured data is assumed to best represent the source distribution. The work presented here is a preliminary attempt to explain and understand the method. As such, all input values used are based on calculated theoretical detector responses as opposed to actual measurements. This allows for a focus on the method without the random uncertainty associated with real measurements.

2. Methods

Multiple measurements can be made using an adjustable cylindrical collimator and circular lead shield described elsewhere (Dewey et al., 2010). The geometry is shown in Fig. 1. The detector responses to a non-uniform vertical source distribution and a semi-infinite uniform horizontal distribution, $R_{\mathrm{volume}}(E,D,t_{\max})$, in counts s^{-1}, can be calculated using the equation presented by (Whetstone et al., 2011), namely:

$$R_{\mathrm{volume}}(E,d,t_{\max}) = \int_0^{t_{\max}} \int_{\beta_{\min}}^{\pi} \frac{S_{\mathrm{p}}(t)A_{\mathrm{R}}(E,\theta)C_{\mathrm{R}}(E,\theta,c_{\mathrm{d}})R_{\mathrm{c}}(E)}{2\left[\frac{d+t}{\cos(\theta)}\right]^2} e^{-\mu_{\mathrm{media}}(E)t/\cos(\theta)}\, e^{-\mu_{\mathrm{air}}(E)[p-t/\cos(\theta)]}\, \mathrm{d}\theta\, \mathrm{d}t, \quad (1)$$

where E is the incident gamma ray energy; d is the distance, in cm, from lower detector face to media surface; $S_{\mathrm{p}}(t)$ is the source activity, in Bq cm^{-3}, at depth t in the media; $A_{\mathrm{R}}(E,\theta)$ is the dimensionless angular detector response at angle θ, in radians; R_{c} is the centerline detector response, in counts $\mathrm{s}^{-1}\,\mathrm{cm}^5\,\mathrm{Bq}^{-1}$; t is the depth in the media, in cm, from the surface; μ_{media} is the media's attenuation coefficient in cm^{-1}; μ_{air} is the attenuation coefficient of air, in cm^{-1}; p is the distance, in cm, from the point of interest to the lowest point on the detector centerline; $t_{\max}$ is the maximum depth of interest, in cm; C_{R} is the ratio of the detector response with no collimator to a point source at a particular angle, θ, to the detector response with the collimator to the same point source at the same angle; and c_{d} is the adjustable distance from the lower detector face to the plane of the lower collimator face.

The function $S_{\mathrm{p}}(t)$ represents the radionuclide depth distribution, approximated using one of several model analytical forms. The functions typically used include exponential, Gaussian, double exponential, polynomial, and double Gaussian. Each model

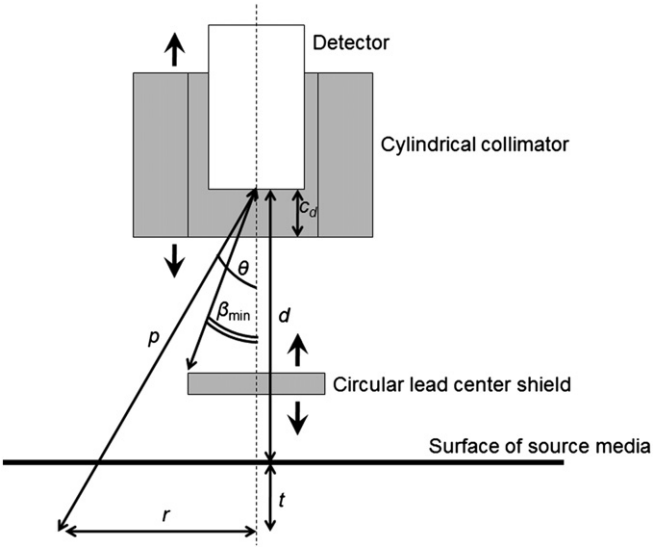

Fig. 1. Diagram of the configurations used to perform the multiple in situ measurements. The positioning of the cylindrical collimator is varied by moving it vertical up and down in relation to the detector. The lead circular center shield can be used for some of the measurements and omitted in others.

[1] In Situ Object Counting System (ISOCS), Canberra 800 Research Parkway, Meriden, CT 06450.