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Fate of ^{60}Co at a sludge land application site

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ABSTRACT

Vertical distributions of ^{60}Co are determined in soil cores obtained from a 10-ha grassland, where anaerobically digested sludge was applied by surface spraying from 1986 to 1995 on the U.S. Department of Energy's Oak Ridge Reservation. These results, along with historical application records, are used to estimate vertical-migration rates and perform a mass balance. The presence of ^{60}Co results solely from the sludge-application process. Soil, vegetation, and surface-water samples were collected. Eleven soil cores were sectioned into 3-cm increments and analyzed by gamma-ray spectrometry. No ^{60}Co was detected in the vegetation or water samples. The downward migration rate of ^{60}Co in the upper 15 cm of soil ranged from 0.50 to 0.73 cm/yr. About 98%, 0.020 ± 0.011 Bq/cm^2, of ^{60}Co remained in the upper 15 cm of soil, which compared favorably with the expected ^{60}Co activity based on historical records of 0.019 ± 0.010 Bq/cm^2.

1. Introduction

The fate of land-applied radionuclides contained in wastewater-treatment sludge has received considerable recent attention (e.g., Bastian et al., 2005; O'Connor et al., 2005; Wolbarst et al., 2006), and the practice of land application of these biosolids is expected to continue to grow because of the perceived economic and societal benefits (Basta et al., 2005). Radionuclides are either present naturally or enter the wastewater stream from numerous anthropogenic sources, such as combustion, fertilizer runoff, mining and smelting operations, leachate from disposal facilities, industrial byproducts, research institutions, and hospitals, as well as from atmospheric fallout.

Historically, many investigators have studied the fate of radionuclides in the environment or laboratory, considering wind, soil, surface water, and groundwater, with numerous studies resulting from the 1986 Chernobyl release (e.g., Anspaugh et al., 2002; Arapis et al., 1997; Baes and Sharp, 1983; Boone et al., 1985; Bunzl et al., 1994; Tanaka and Ohnuki, 1996; Whicker and Pinder, 2002). Many of these studies involved acute application rates with high concentrations of radionuclides. In contrast, the study conducted at the Department of Energy's (DOE) Oak Ridge Reservation focused on the fate of ^{60}Co resulting from a chronic application rate at a low concentration. Results from previous studies of ^{60}Co are compared to the results from this investigation.

A study was undertaken during the summer of 1996 to characterize the movement of sludge-applied ^{60}Co on a land-application site near Oak Ridge, Tennessee, USA. Because of the types of industries in the Oak Ridge area, this sludge contained a variety of radionuclides; however, ^{60}Co was the radionuclide selected for investigation because its presence is unique to the sludge-application site. Anaerobically digested sludge from the Oak Ridge wastewater-treatment plant was applied to 11 grassland or forested sites on the Department of Energy's Oak Ridge Reservation beginning in 1983. This sludge typically contained 2–4% solids and was applied by either surface spraying or subsurface injection of sludge. One of these application sites, a 10.1-ha pasture designated as the Upper Hayfield site, was selected for study to determine whether all of the applied radionuclides could be accounted for through soil sampling. Sludge application by surface spraying at the Upper Hayfield site occurred intermittently from 1986 to 1995. With state and U.S. Department of Energy (DOE) oversight, the City of Oak Ridge and Oak Ridge National Laboratory worked cooperatively to minimize any adverse effects of potential accumulation of radionuclides in sludge-application areas (Gunderson et al., 1995).

2. Material and methods

The presence of ^{60}Co at this site was solely the result of surface spraying, and its occurrence at depth was investigated to determine vertical-migration rates and a mass balance between applied and measured soil activities and vegetative uptake.

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In addition, the percentage of applied ^{60}Co remaining in the soil, both in the top 15 cm and at depths below 15 cm, was estimated for the next few decades. Soil pH at the site ranges from about 5.0 to 6.0, and cation-exchange capacity is about 6 cmol/kg (P.R. Jardine, Ph.D., personal communication, 22 August 2007). Three soil samples from the Upper Hayfield site showed an average weight-percent distribution of 19% gravel (>2 mm), 25% sand (2 mm–63 μm), 47% silt (63–2 μm), and 9% clay (<2 μm). The soils are associated with the Knox Group and are generally formed from residuum that usually is uniform cherty silty clay (Hatcher et al., 1992).

2.1. Radionuclides applied to the site

Data on sludge application at the Upper Hayfield site between 1986 and 1995 were obtained from the City of Oak Ridge (City of Oak Ridge, 1991–1996c) and the Tennessee Department of Environment and Conservation (TDEC, 1996). A "sludge-application model" was developed to estimate present radionuclide activity resulting from past sludge application at the site. The model used reported application dates, weekly application rates, and weekly specific activity levels in applied sludge. The reported activity of sludge applied to the site during each week of application was decay corrected to 1 June 1996, using the following formula:

$$A_{\text{site}} = VA_{\text{sludge}}e^{-\lambda W} \tag{1}$$

where A_{site} is the 1 June 1996 activity resulting from a prior week's sludge application (Bq); V is the volume of sludge applied during that week (L); A_{sludge} is the reported specific activity of sludge during that week (Bq/kg); λ is the decay constant for the radionuclide of interest (1/week); and W is the number of weeks between the application period and the week of 31 May–6 June 1996. In using Eq. (1), it is assumed that 1 kg of sludge is equivalent to 1 L of sludge, which is a reasonable assumption given the low solids content of the sludge (i.e., 2–4%). The 1 June 1996 activity for ^{60}Co, as predicted by the model, was 19.3 ± 10.3 MBq, or 0.0191 ± 0.0102 Bq/cm^2. Variability in the model's results (i.e., ±sigma) may be attributed to several sources, including the radionuclide analyses, the sampling process, and actual variability in the source term. Prior to June 1988, a single sample was analyzed monthly. Subsequently, a sample was collected from the first sludge-application shipment made on a given day, from which a single 1-L composite sample was created from equal parts of that week's daily samples. The variability in sample activity from week-to-week was minimal, and only over multi-month periods is change noted. Results from curve fitting were used to provide weekly input to the model for time periods prior to June 1988. The variability associated with the available data used in the model, no matter the cause, was estimated by calculating the standard deviation of the data about a best-fit line using the following equation:

$$s_e = \sqrt{\frac{\sum(y-\bar{y})^2}{n-2}} \tag{2}$$

where s_e is the standard deviation; $\sum(y-\bar{y})^2$ is the sum of squared deviations from the best fit line $\bar{y}$; and n is the number of data points.

2.2. Sample collection

Soil samples were collected from the Upper Hayfield (11 cores, comprising 60 samples) and adjacent reference areas where no sludge had been applied (three cores, comprising 16 samples). Flora samples collected from the Upper Hayfield site were primarily above-ground grasses (11 samples), but included a variety of other leaves, stems, roots, and fruits (seven samples). The grass samples consisted primarily of *Muhlenbergia schreberi*, *Setaria faberi*, and *Panicum dichotomiflorum*. Other samples collected included *Asclepias syriaca*, *Allium tricoccum*, and various unidentified terrestrial macrophytes. Because there was no surface water on the Upper Hayfield, surface water (three samples) was collected from a pond bounded by another sludge-application site, from a depression area adjacent to the Upper Hayfield site, and from a pond near the Upper Hayfield site.

Soil cores were randomly collected to a depth of at least 15 cm when possible. Eleven soil cores were collected from the Upper Hayfield site, and one soil core was collected from a depression area northeast of the Upper Hayfield site. To help characterize radionuclide movement, the soil cores were sectioned into 3-cm increments and collected in 0.5-L Marinelli beakers.

The soil-coring device was a stainless-steel pipe with a cross-sectional area of 168 cm^2. This coring device was used to collect the 11 soil cores from the Upper Hayfield site. Soil compaction was considered negligible because of the relatively large diameter of the pipe used (i.e., 15 cm), and this conclusion was confirmed by comparing the extracted core lengths with the borehole depths. A soil-coring device with a cross-sectional area of 49 cm^2 was used to collect the soil sample from the depression area.

2.3. Sample analysis

Samples were analyzed using high-purity germanium and lithium-drifted germanium coaxial-photon detector systems that were calibrated using a certified mixed-gamma standard traceable to the National Institute of Standards and Technology. The most recent efficiency calibration used a mixed-gamma standard, QCY48, from Amersham Radiochemical Centre. A known quantity of this material was diluted in a 4-M HCl solution and placed in the 0.5-L and 1-L Marinelli beakers used for calibrating (Larsen and Cutshall, 1981).

A calibration check and contamination check were performed daily for each detector. To verify results, a quality-control blank-soil sample, a laboratory-control sample, and quality-assessment samples also were counted during the investigation. As an additional verification measure, Oak Ridge National Laboratory participates routinely in the Quality Assessment Program administered by the DOE Environmental Measurements Laboratory. The program is designed to test the quality of the environmental–radiological measurements being reported by DOE contractors and subcontractors.

All samples contained low concentrations of ^{60}Co. Count times of at least 1000 min were used to reduce statistical-counting uncertainties. To determine sample activities, the detector systems first calculated the photopeak count rate, which is a combined count rate from the sample itself and any ambient background. The ambient-background count rates were subtracted to determine the net-count rate introduced by the sample. The background-count rate was determined on a monthly basis by performing at least a 50-h count with no sample on the detector.

2.4. Vertical migration model

To characterize the downward movement of ^{60}Co at the Upper Hayfield site, residence half-times of ^{60}Co were estimated. The residence half-time is the time necessary for the activity present in a soil layer to be reduced by 50%. Ideally, the residence half-times would be estimated by measuring radionuclide soil-profile changes over time. However, time-sequenced data were not available, so a six-compartment model was used to estimate the ^{60}Co residence half-times based on the historical radionuclide-application rates and the present radionuclide-depth profile. Five of the "compartments" represented 3-cm segments of the soil cores at depths of 0–3 , 3–6 , 6–9 , 9–12 , and 12–15 cm. The sixth "compartment" represented soil at a depth greater than 15 cm. Many transport processes can contribute to migration of ^{60}Co through soil, but they can be generalized by considering diffusion and mechanical dispersion, advection, radioactive decay, retardation, and a source/sink term (Domenico and Schwartz, 1990). In the approach used for this study, radionuclide concentrations in each soil layer were assumed to be reduced only through radioactive decay and movement into deeper soil. The model estimates migration rates, but does not characterize the transport processes involved (e.g., diffusion and mechanical dispersion, advection, retardation). A visual representation of this simplified model is shown in Fig. 1. The ^{60}Co activity in the upper 0–3 cm soil layer is described by the following differential equation:

$$\frac{dA_1}{dt} = D - K_1A_1 - \lambda A_1 \tag{3}$$

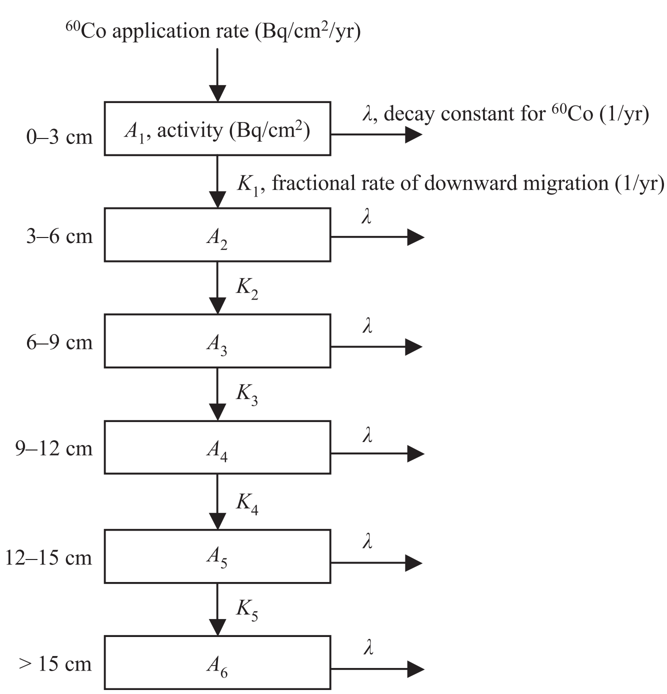

Fig. 1. Visual representation of the six-compartment model used to estimate ^{60}Co residence half-times at the Upper Hayfield site.