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THE FATE OF RADIONUCLIDES IN SEWAGE SLUDGE APPLIED TO LAND^o

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ABSTRACT

Municipal sewage sludge containing up to 12 pCi/g (dry wt) ¹³⁷Cs, 20 pCi/g ⁶⁰Co, and 300 ppm U was injected in a pasture (43 Mg/ha) and sprayed over a young pine plantation (34 Mg/ha). In the pasture, radionuclides were largely retained in the upper 15 cm of the soil, and only about 15% moved below 15 cm. Sludge rapidly infiltrated the soil on the pine plantation. One year after application, at least 85% of the ¹³⁷Cs, ⁶⁰Co, and U were found in the upper 7 cm of the pine plantation, with only about 15% moving into the 7- to 15-cm strata. On-site total added radiation dose was 2 to 6 mrem/year. Radionuclides were not detected above background in soil solutions at ~ 50 cm depth or in shallow down-gradient groundwater wells. Surface runoff from application areas did not have elevated radionuclide concentrations. Concentrations of these radionuclides increased slightly in vegetation on treated sites, and uranium was notably higher in earthworms.

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INTRODUCTION

Sewage treatment plants receive radionuclides from industry, hospitals, and from atmospheric fallout that may be removed and concentrated in the sewage sludge (ref 1-3). Current U.S. regulations for radionuclide releases to sanitary sewers consider neither accumulation of radionuclides in sewage sludge nor implications for sludge disposal. Therefore, radionuclides have accumulated in sludge destined for disposal by land application (ref 4). The environmental fate of sludge constituents has been the subject of numerous investigations (ref 5,6). However, the fate of radionuclides in sewage sludge applied to land has received little attention thus far.

Digested sewage sludge from the City of Oak Ridge was applied to a pasture and a 4-year-old loblolly pine plantation on the U.S. Department of Energy's Oak Ridge Reservation (Oak Ridge, TN USA) to improve soil fertility and plant growth (ref 7). During this time the sludge was contaminated with ^{137}Cs , ^{60}Co , ^{131}I , and depleted U from industrial and medical sources. We measured the activity of ^{137}Cs and ^{60}Co and the concentration of U in soils, soil water, groundwater, surface runoff, vegetation, and earthworms to determine the fate of these nuclides.

METHODS

Anaerobically digested sludge from the City of Oak Ridge was injected to 10 cm soil depth in the pasture at a rate of 43 Mg/ha from December 1986 through May 1988. The sludge contained about 12 pCi/g (dry wt) ^{137}Cs , 20 pCi/g ^{60}Co , and 110 ppm U from industrial releases. Sludge was sprayed over a 4-year-old pine plantation (34 Mg/ha) from June 1988 through June 1989. This sludge contained about 2 pCi/g ^{137}Cs , 5.7 pCi/g ^{60}Co , and 300 ppm U. In the pasture, the upper 15 cm and the 15 - 30 cm soil depth were sampled following application. Vegetation (grass) was sampled at the end of application and about 2 years later. The upper 15 cm of soil on the pine plantation was sampled following application and again in May 1990. In November 1989, the upper ~45 cm of soil was sampled. During May 1990, new-growth pine needles were collected.

During sludge application, soil solution at 40- to 60-cm depth was collected with suction lysimeters. Samples of surface runoff from active application areas were collected during rainstorms. Groundwater was collected from shallow wells (<10 m) up- and down-gradient of the pine plantation. Earthworms were also collected from the pine plantation. Similar samples were collected from untreated (reference) areas adjacent to the application sites.

^{137}Cs , ^{60}Co , ^{40}K , and ^7Be were determined by spectrometry (IGe detector) calibrated with standards of known activities and geometries. When possible samples were counted to reduce counting error to <10%. Total U was determined by neutron activation (sludge and soil) or ICP-mass spectrometry (water, vegetation, and earthworms) and calibrated with reference materials.

RESULTS

Aside from the radionuclide content, Oak Ridge sludge was typical of municipal sewage sludge in the U.S. (ref 7) and was applied at rates typical for land application. We found that sludge metals were rapidly immobilized and retained in the upper layers (15 cm) of the soil (ref 7), as has been observed elsewhere (ref 5). On the tree plantation, U, ^{137}Cs , and ^{60}Co were retained in the upper 15 cm of the soil (Table 1). In the pasture about 15% of the added U, ^{137}Cs , and ^{60}Co occurred below 15 cm. We could not determine if this finding reflected migration or variable injection depth. The upper 15 cm of soil on the pine plantation was sectioned (Table 1). The profile showed that radionuclides from sludge sprayed over the surface rapidly moved into the soil. Although U averaged 300 ppm in the sludge, the concentration in the surface 2 cm of the soil averaged only about 25 ppm (Table 1) when application ended. Following treatment, ^{137}Cs seemed to be retained in the upper 2 cm of soil, whereas some ^{60}Co clearly moved to the 7 to 15 cm strata. About one year after application, the majority of U, ^{137}Cs , and ^{60}Co was still in the upper 7 cm of the soil, and the extent of movement of each seemed similar. Although the average concentrations of ^{137}Cs , ^{60}Co , and U in the 7- to 15-cm depth were not significantly greater for the treated area (Table 1), there was evidence of radionuclide movement to this strata from individual

soil cores.

Mass balance calculations for sludge radionuclides on the pine plantation (based on soils collected in May 1990) accounted for all of the added radionuclides in the upper 15 cm of the soil (ref 8). Total added radiation dose (all pathways, continuous occupancy) was about 2.5 and 5.5 mrem/year for the pine plantation and pasture, respectively, and was primarily due to short-lived ^{60}Co (ref 8).

Vegetation on application sites had elevated concentrations of N, P, K, and Ca but not of heavy metals (ref 7). Grass collected from the pasture site was slightly enriched with ^{137}Cs following application and was slightly enriched in U and ^{137}Cs almost 2 years later (Table 2). New growth pine needles collected about 1 year after application had higher concentrations of ^{137}Cs , ^{60}Co , and U than plants on the reference site. The concentrations of these elements were not greatly above background levels and were small compared with naturally occurring isotopes (e.g., ^{40}K and ^7Be).

Soil water on the application sites had high NO_3^- concentrations but did not contain elevated concentrations of metals (ref 7). Soil water analyzed for ^{137}Cs and ^{60}Co on two occasions contained <5 pCi/L of each nuclide. At times, some down-gradient groundwater wells at the pine plantation contained up to 2 times the concentration of U of up-gradient wells, but concentrations were generally low (<0.1 ppm). ^{137}Cs and ^{60}Co were never detected in down-gradient wells (<2.5 pCi/L). Surface runoff from application sites was enriched in NO_3^- , P, BOD, and fecal coliform bacteria but never contained detectable ^{137}Cs or ^{60}Co (i.e., <5 pCi/L).

Earthworms and other soil organisms can accumulate metals (ref 9) and may transfer these to other organisms. Earthworms from the treated area of the pine plantation contained 0.4 pCi/g (dry wt) ^{137}Cs and 0.6 pCi/g ^{60}Co , concentrations near our limit of detection. Earthworms from the treated area contained about 3 ppm U (dry wt), compared with about 0.5 ppm U for the reference area.

Radionuclides applied to soils in sewage sludge are diluted and immobilized in the soil. On these sites the radionuclides applied with sludge increased radiation dose by only about 2 to 6 mrem/year. Concentrations of radionuclides in the vegetation were not greatly increased. Radionuclides were neither moving off-site with surface runoff nor moving into the groundwater.

Table 1 Radionuclides in soils (dry wt basis). Values are means \pm 1 S.D. or ranges in [] for ^an = 6 treatment, 3 reference; ^bn = 3 treat., 3 ref.; or ^cn = 6 treat., 4 ref. samples.

Soil depth (cm)		U (ppm)		¹³⁷ Cs (pCi/g)		⁶⁰ Co (pCi/g)	
		Treatment	Reference	Treatment	Reference	Treatment	Reference
Pasture ^a	0-15	6.2 \pm 1.4	3.3 \pm 0.3	1.24 \pm 0.44	0.43 \pm 0.13	0.26 \pm 0.09	<0.07
	15-30	3.8 \pm 0.4	3.3 \pm 0.4	0.28 \pm 0.26	<0.11	[<0.03-0.06]	<0.02
Pine Plantation ^b	Nov. 1989 0-15	7.9 \pm 0.6	3.8 \pm 0.7	0.48 \pm 0.06	0.43 \pm 0.01	0.09 \pm 0.01	<0.04
	15-30	3.6 \pm 0.4	3.4 \pm 0.7	0.04 \pm 0.01	0.05 \pm 0.02	<0.011	<0.015
	30+	3.7 \pm 0.4	3.4 \pm 0.5	<0.012	<0.012	<0.010	<0.010
Pine Plantation (0 - 15 cm only)							
July 1989 ^c	0-2	25.3 \pm 12.2	3.9 \pm 0.6	0.88 \pm 0.10	0.45 \pm 0.01	0.53 \pm 0.16	<0.05
	2-7	11.4 \pm 4.0	4.0 \pm 1.4	0.49 \pm 0.14	0.42 \pm 0.04	0.12 \pm 0.02	<0.03
	7-15	5.4 \pm 1.0	4.0 \pm 1.1	0.33 \pm 0.19	0.43 \pm 0.15	[<0.02-0.10]	<0.03
May 1990 ^c	0-2	23.0 \pm 11.9	4.1 \pm 1.2	0.77 \pm 0.18	0.44 \pm 0.04	0.31 \pm 0.16	<0.05
	2-7	9.7 \pm 5.3	4.0 \pm 1.1	0.66 \pm 0.12	0.44 \pm 0.14	0.10 \pm 0.07	<0.01
	7-15	5.0 \pm 1.4	4.0 \pm 1.1	0.52 \pm 0.15	0.36 \pm 0.19	[<0.01-0.05]	<0.01

Table 2 Radionuclides in vegetation (dry wt basis). Values are means \pm 1 S.D. or ranges in [] for ^an = 3 treatment and 3 reference or ^bn = 6 treatment and 2 reference samples. NA = not available.

	U (ppm)	¹³⁷ Cs (pCi/Kg)	⁶⁰ Co (pCi/Kg)	⁴⁰ K (pCi/Kg)	⁷ Be (pCi/Kg)
Pasture-1988 ^a					
Treatment	NA	61 \pm 52	[59-<200]	27400 \pm 5800	5360 \pm 2930
Reference	NA	[61-<200]	<86	18900 \pm 4420	13800 \pm 8200
Pasture-1990 ^a					
Treatment	[<0.01-0.03]	[<14-34]	<16	26100 \pm 5200	1490 \pm 662
Reference	<0.01	[<16-26]	<24	22300 \pm 1660	3440 \pm 383
Pines-1990 ^b					
Treatment	0.11 \pm 0.08	[<15-34]	[<12-29]	3618 \pm 287	1913 \pm 67
Reference	0.03 \pm 0.01	<15	<15	3811 \pm 602	2381 \pm 359

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