

URANIUM AND RADIUM CONCENTRATIONS IN PLANTS GROWING ON URANIUM MILL TAILINGS IN SOUTH DAKOTA

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Abstract

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Vegetation and soil samples were collected from a uranium mill tailings site and control sites in South Dakota. Uranium concentrations in soils from the mill tailings averaged $13.3 \mu\text{g g}^{-1}$ compared to $5.1 \mu\text{g g}^{-1}$ in soils from control sites. ^{226}Ra concentrations in soils averaged 111.0 pCi g^{-1} and 1.8 pCi g^{-1} from the mill tailings and control sites, respectively. Uranium concentrations in plants from the mill tailings averaged $3.6 \times 10^{-1} \mu\text{g g}^{-1}$, but only $3.4 \mu\text{g g}^{-1}$ from control sites. ^{226}Ra in plants from the mill tailings averaged 2.9 pCi g^{-1} compared to $3.7 \times 10^{-2} \text{ pCi g}^{-1}$ from the control sites. Concentration ratios (amount in plant/amount in soil) of radionuclides indicated that two grasses and one forb were not concentrating uranium or ^{226}Ra .

Introduction

Waste products from uranium milling processes (tailings) contain low amounts of natural radioactive materials. About 85% of the original radioactivity present in the ore is present in the tailings (Moffett and Tellier, 1977), of which the uranium content originally ranged from 0.05 to 0.25% (U.S. Nuclear Regulatory Commission, 1979). Some information exists on the uptake of uranium decay series radionuclides by plants (Moffett and Tellier, 1977; Rickard et al., 1977; Dreesen et al., 1978; Schreckhise and Cline, 1980; Rayno et al., 1980). Unless very thick soil coverings or imper-

meable barriers to plant roots are established, plants may disperse radionuclides from mill tailings disposal sites (Dreesen et al., 1978; Cline et al., 1980). Uptake of radionuclides by plants depends on the plant species, the radionuclide and on substrate characteristics (Dreesen et al., 1978). As a result, radio-ecologists need additional information on the uptake of specific radionuclides by various plant species growing in environments where they are in contact with radionuclides. The objective of this study was to evaluate the levels of uranium and radium in plants growing on mill tailings.

Study area and methods

This study was conducted on the Edgemont uranium mill tailings in southwestern South Dakota. The mill has not been operated since 1972. The mill tailings were re-processed mill sands, covered with topsoil and seeded to a mixture of crested wheatgrass (*Agropyron cristatum*), yellow sweet clover (*Melilotis officianalis*) and rye (*Lolium spp.*) in the autumn of 1973. Because the topsoil was not spread evenly over the tailing materials, the depth of the soil covering was about 10 cm near the center of the plot and about 30 cm near the edges. The control sites, west of the mill tailings, were selected for similarity in species vegetation on each site.

Samples of crested wheatgrass, alkali sacaton (*Sporoulus airoides*) and summercypress (*Kochia scoparia*) were clipped from five, 0.125-m² circular plots along six transects established on the topsoil-covered mill tailings. Additional collections of these plant species were taken from the study sites to ensure a sufficient sample for the radiochemical analyses (approximately 1.0 kg dry weight). Samples of the same plant species were collected from control sites. Except on the mill tailings, these plant species were not found occurring together on a single site. As a result, samples of crested wheatgrass were collected from a crested wheatgrass pasture grazed lightly during the winter months, alkali sacaton was collected from a lowland area below a water dam for livestock, and summercypress was collected from a disturbed site. Samples of vegetation were returned to the laboratory, washed in a 5% calgon solution, and rinsed in distilled water to remove externally impinged radionuclides from the samples (Moffett and Tellier, 1977). The samples were then combined to make two samples from each site, and were air-dried at 60°C.

Six soil samples from the top 2 dm were collected from six locations in a systematic grid covering the mill tailing site and crested wheatgrass pasture site. Two, 2-dm soil samples were also collected from the alkali sacaton and summercypress control sites. The soil samples were combined to make two samples per site.

The plant and soil samples were analyzed for total uranium and ²²⁶Ra. In addition, electrical conductivity (EC), sodium (Na), calcium (Ca), magnesium (Mg) and were determined from saturated extracts made from the

TABLE I

Mean chemical characteristics from the first 2 dm of soil from revegetated uranium mill tailings and control sites in western South Dakota^{1,2}

Site	EC (mmhos cm ⁻¹)	pH	Ca (p.p.m.)	Mg (p.p.m.)	Na (p.p.m.)
Mill tailings	5.10 (4.7–5.5) ^a	7.59 (7.5–7.7) ^a	418 (170–665)	373 (350–395) ^a	615 (460–770)
Control					
Alkali sacaton	11.35 (3.7–19.0)	7.64 (7.4–8.2) ^b	875 (800–950)	198 (270–225)	2932 (265–5600)
Crested wheatgrass	0.45 (0.4–0.5) ^b	8.00 (8.0–8.0) ^b	93 (75–110)	10 (8–11) ^b	3 (2.5–3)
Summercypress	3.85 (3.8–3.9)	7.75 (7.7–7.8) ^b	752 (735–770)	153 (150–155) ^b	370 (270–450)

¹ Means with different letters for superscripts for a parameter are significantly different ($\alpha < 0.05$).

² Numbers in parentheses following the means are the range.

soil samples. Laboratory analyses were performed by U.S. Testing, Inc.*, in Richland, Washington. The soil chemistry parameters and radionuclide concentrations from mill tailings and control sites were tested using *t*-tests. Significance was determined at $\alpha \leq 0.05$.

Results

Electrical conductivity of soils from the crested wheatgrass pasture were significantly lower than from mill tailings soils (Table I). Lower pH values were found in soils from the mill tailings compared to the three control sites, but magnesium concentrations were higher in mill tailings soils than either the crested wheatgrass or summercypress control-site soils. Other differences in the chemical composition of soils evaluated were probably masked by the limited sample sizes (e.g. sodium). However, overall, soils from the alkali sacaton and summercypress control sites were more similar to the mill tailings soils than soils from the crested wheatgrass control site.

Concentrations of both uranium and ^{226}Ra were higher in soils from the mill tailings than from the control sites (Table II). Uranium concentrations were significantly higher in all three species of plants growing on the mill tailings than in plants from the control sites (Table III). However, significant differences in ^{226}Ra concentrations were found only in crested wheatgrass. No comparisons could be made for ^{226}Ra concentrations in summercypress. Concentration ratios (concentration in plant tissue/concentration in soil) of uranium and ^{226}Ra were less than 1.0 (Table IV) and in the order of 10^{-2}

TABLE II

Mean concentration of uranium and radium in the first 2 dm of soils on a revegetated uranium mill tailing and on control sites in western South Dakota^{1,2}

Site	Total uranium ($\mu\text{g g}^{-1}$)	^{226}Ra (pCi g^{-1})
Mill tailings	13.286 (12.475–14.097) ^a	111.0 (107.0–115.0) ^a
Control		
Alkali sacaton	7.199 (5.160–9.239) ^b	2.905 (1.290–2.90) ^b
Crested wheatgrass	3.211 (3.003–3.419) ^b	1.007 (0.803–1.210) ^b
Summercypress	5.022 (4.618–5.427) ^b	1.640 (1.39–1.89) ^b

¹ Mean radionuclide concentrations in the same column with different letters for superscripts are significantly different ($\alpha < 0.05$).

² Numbers in parentheses following the mean are the range.

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TABLE III

Mean concentrations of uranium and radium in plants growing on a revegetated uranium mill tailings and on control sites in western South Dakota^{1,2,3}

Site	Total uranium ($\mu\text{g g}^{-1}$)	²²⁶ Ra (pCi g ⁻¹)
Mill tailings		
Alkali sacaton	4.381×10^{-1} (3.589–5.175 $\times 10^{-1}$) ^a	3.930 (1.88–5.98)
Crested wheatgrass	1.339×10^{-1} (0.820–1.858 $\times 10^{-1}$) ^a	1.795 (1.700–1.890) ^a
Summercypress	5.024×10^{-1} (4.840–5.207 $\times 10^{-1}$) ^a	ND
Control		
Alkali sacaton	5.237×10^{-2} (5.173–5.300 $\times 10^{-2}$) ^b	4.665×10^{-2} (2.51–6.82 $\times 10^{-2}$) ^b
Crested wheatgrass	2.349×10^{-2} (1.862–2.836 $\times 10^{-2}$) ^b	5.720×10^{-2} (3.39–8.05 $\times 10^{-2}$) ^b
Summercypress	2.774×10^{-2} (2.748–2.800 $\times 10^{-2}$) ^b	7.730×10^{-3} (7.773 $\times 10^{-3}$)

¹ Mean radionuclide concentrations in the same column in plants of the same species with different letters for superscripts are significantly different ($\alpha < 0.05$).

² ND = No data was reported for this sample.

³ Numbers in parentheses following the means are the range.

TABLE IV

Concentration ratios (\bar{x} concentration in the plant tissue/ \bar{x} concentration in the soil) of uranium and radium in plants growing on a revegetated uranium mill tailings and on control sites in western South Dakota^{1,2}

Site	Total uranium	²²⁶ Ra
Mill tailings		
Alkali sacaton	0.0248	0.0354
Crested wheatgrass	0.0075	0.0162
Summercypress	0.0577	ND
\bar{x}	0.0300 ^a	0.0258 ^a
Control		
Alkali sacaton	0.0074	0.0223
Crested wheatgrass	0.0077	0.0568
Summercypress	0.0048	0.0047
\bar{x}	0.0066 ^b	0.0279 ^b

¹ Mean concentration ratios in the same column with different letters for superscripts are significantly different ($\alpha < 0.05$) between mill tailings and control sites.

² ND = No data was reported for the ²²⁶Ra analysis in summercypress.

or less. Significantly higher concentration ratios were found for plants growing on the mill tailings compared to the control sites.

Discussion and conclusions

The amount of radionuclides taken up by plants from soils depends on the radionuclide form, soil moisture, and chemical and mineralogical composition

of the soil (Dreesen et al., 1978, 1982). Higher concentrations of uranium and ^{226}Ra were observed in plants growing on the mill tailings than in those on the control sites. However, the radionuclide concentrations of mill tailings soils were also higher. Rickard et al. (1977) suggested that plant uptake of the uranium was independent of soil concentration. The uranium and ^{226}Ra concentrations in mill tailings soil samples from this study were similar to those reported in Canada (Moffett and Tellier, 1977), but plant samples from the mill tailings in this study had higher uranium concentrations (by 1 order of magnitude) than those from Canada, which was probably related to differences in the chemical properties of the growing media in the two studies. Rickard et al. (1977) reported that concentrations of uranium in soils greater than 200 p.p.m. are toxic to some plants, and uptake of uranium from the soil by plants was in the range 10^{-1} - 10^{-2} $\mu\text{g g}^{-1}$. Uranium concentrations in plants from this study were within this range.

Apparently, plants do not readily concentrate elements of the uranium decay series. Concentration ratios for uranium varied in this study, but were on the order of 10^{-2} , as reported elsewhere (Schreckhise and Cline, 1980; Rayno et al., 1980; Garten, 1981; Dreesen et al., 1982). Concentration ratios for ^{226}Ra also were in the order of 10^{-2} ; similar to the concentration ratio reported by Moffett and Tellier (1977) of 0.03. The higher concentration ratios for both radionuclides on the mill tailing would suggest that there was some dependence of uptake of these radionuclides on the initial soil concentrations. However, this factor was also masked by potential differences in uptake related to soil characteristics. For example, ^{226}Ra is more mobile at higher pH (Dreesen et al., 1982).

Moffett and Tellier (1977) suggested that with concentration ratios of 0.03 or less (as reported here), plants can hardly be ^{226}Ra accumulators. Therefore, plants do not constitute a significant mode for spread of these uranium decay-series radionuclides from covered mill tailings to adjacent environments, at least in the short-term. The long-term implications of radionuclide uptake involve the accumulation of uranium and ^{226}Ra on the soil surface from many years of litter fall and decomposition, and off-site movement by wind and water.

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