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ABSTRACT

Radiochemical analysis was performed on samples of soil and eight species of common vegetation growing on the Anaconda uranium mill site, located in New Mexico. The concentrations of the long-lived radionuclides U-238, U-234, Th-230, Ra-226, and Pb-210 in these forage plants were determined. The sampling procedures and analytical laboratory methods used are described. The highest radionuclide concentration found in a forage species was 130 pCi of Ra-226 per gram dry weight for grass growing on the main tailings pile at Anaconda, where the surface soil activity of Ra-226 was 236 pCi/g. A comparison of shoots activity with that of roots and soil was used to determine a distribution index and uptake coefficient for each species. The distribution index, the ratio of root activity to shoot activity, ranged from 0.30 (Th-230) in galleta grass (Hilaria jamesii) to 38.0 (Ra-226) in Indian ricegrass (Oryzopsis hymenoides). In nearly all instances, the roots contained higher radionuclide concentrations. The uptake coefficient, the ratio of vegetation activity to soil activity, ranged from 0.69 (U-238) in Indian ricegrass roots to 0.01 (U-238) in four-wing saltbush (Atriplex canescans) shoots. The range of radionuclide concentrations in plants growing on the Anaconda mill site is compared to that in vegetation from a control site 20 km away.

INTRODUCTION

The radiological assessment of the food pathway which originates from uranium milling operations is of current concern. There is a lack of data concerning forage uptake of U-238 series nuclides, especially the long-lived isotopes U-238, U-234, Th-230, Ra-226, and Pb-210. This information is essential for a realistic estimation of food-chain contamination. Some vegetation uptake data for the U-238 series has been reported (1-10), as well as for other radionuclides such as Sr-90 and plutonium (11-13). But, the uptake of U-238 series radionuclides by many forage plants has not been studied.

This investigation was conducted at the Anaconda uranium mill in Bluewater, New Mexico, and the main considerations were: identification of forage species consumed by livestock, the concentration of

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each radionuclide in each forage species, the ratio of activity in roots to that in shoots (which are above ground), and the uptake coefficient (plant-to-soil activity ratio) for each plant species.

TECHNIQUE

Representative plant species were sampled at several locations adjacent to uranium mill tailings, within a circular area about 15 meters in diameter. These samples were collected within 0.5 km of the embankment of the Anaconda tailings pond. At the same sites, composite soil samples were collected from the top 15 cm. Also, samples were collected at the northern boundary (NB) of the site, at a distance of 1.5 km from the tailings pond, and at a control location (C) about 20 km from the Anaconda mill and other uranium mines and mills. The main vegetation species collected were perennial grasses (Hilaria jamesii, Sporobolus asper, Bouteloua gracilis, Sitanion hystrix, and Oryzopsis hymenoides) and shrubs: Russian thistle (Salsola paulsenii), four-wing saltbush (Atriplex canescans), and sticky snakeweed (Gutierrezia lucida).

Each composite plant sample was washed in mild soap and water to remove foliar particulates and soils, followed by rinsing and thorough drying. Then, each composite was wet-ashed and dissolved in nitric-perchloric acid to make stock solutions. The radiochemical techniques for the measurement of each radionuclide have been previously reported (14). U-238, U-234, and Th-230 were measured using solvent extraction separation followed by thin electrodeposition on stainless-steel disks and alpha spectrometry. Pb-210 was separated using solvent extraction, and its activity was measured by beta-counting its Bi-210 daughter with a gas proportional counter. Ra-226 activity was determined using the de-emanation method, alpha-counting radon and its daughters (Po-218, Po-214) with an alpha scintillation detector. Soil activity for each composite sample was measured by either extracting the radionuclides from the soil with nitric acid and mild heat, followed by radiochemical determinations as above, or by gamma spectroscopy using a Ge(Li) detector.

Any system for the analytical measurement of radioactivity is limited to a minimum detection level in a given counting period. A measurable quantity of radioactivity above this detection level can be achieved with either a high specific activity of a small sample, or a larger sample of lower specific activity. In this study, the sample size was limited to the available collected forage. Also, the specific activity was not known before measurement. Thus, the only available parameter for exceeding the detection level was the counting period. Measurable radioactivity was not observed in some vegetation samples. The minimum detectable limits and analytical errors* for each radionuclide determination were calculated using the procedures previously reported (14).

$$^{*}\pm \text{ error} = \frac{\sqrt{\text{total counts/count time (min)}}}{\text{net count/min}}$$

RESULTS

In the rangeland surrounding the Anaconda mill, the predominant livestock are cattle and sheep. Table 1 indicates that all grasses sampled are valued as forage for livestock, especially Indian rice-grass and blue grama grass. The ranges of radionuclide concentrations (pCi/g) for dried forage grasses on the mill site are: 0.23 to 3.92 (uranium in shoots), 0.63 to 12.5 (uranium in roots), 0.12 to 13.2 (Th-230 in shoots), 0.75 to 51.8 (Th-230 in roots), less than detectable to 3.43 (Ra-226 in shoots), 0.18 to 130 (Ra-226 in roots), less than detectable to 16.0 (Pb-210 in shoots), and less than detectable to 81.5 (Pb-210 in roots). Grasses from the control site have not been analyzed.

Concentrations (pCi/g) in the shoots of shrub samples obtained from the control location range from less than detectable to 0.38 (uranium), less than detectable to 0.09 (Th-230), and less than detectable to 0.08 (Ra-226). In these shrubs from the control location, Pb-210 concentrations were less than the detectable limit. As shown in Table 2, the ranges of radionuclide concentrations (pCi/g) in dried composite soil samples from the mill site are 2.70 to 18.2 (U-238), less than detectable to 114 (Th-230), 0.87 to 236 (Ra-226), and 2.56 to 209 (Pb-210). For the dried soil composites from the control location, the radionuclide concentrations (pCi/g) are 5.18 (U-238), less than detectable (Th-230), 0.61 (Ra-226), and 1.40 (Pb-210).

The uptake coefficient is defined in this study as the ratio of activity concentration in the dried forage to that in the dried soil, both collected at the same site. Table 3 gives the uptake coefficients calculated for both grasses and shrubs. The uptake coefficient ranges are: 0.01 to 0.12 (U-238), 0.01 to 0.25 (Ra-226), and 0.12 to 0.55 (Pb-210). Th-230 concentration in background soil cannot be readily measured by gamma spectroscopy and was not measured by alpha spectroscopy. Thus, only one uptake coefficient for Th-230 is reported (Table 3).

DISCUSSION

The contribution of any forage species to human exposure is directly related to its linkage to the human food chain, i.e. selection by foraging animals which, in turn, provide food for human consumption. A plant which is not preferred as forage, even with a high radionuclide-uptake coefficient, is not radiologically significant in the food pathway: plant-animal-man. An excellent forage (Table 1) is defined in this study as a plant palatable to livestock, widely distributed, and consumable either fresh or dried. A good forage consists of vegetation that is consumable only fresh or dry, lower in distribution and seasonal availability, or less palatable to grazing livestock. A bad forage is vegetation with a combination of very low distribution and poor palatability (poisonous, such as sticky snake weed). Almost all grasses collected are excellent forage material.

The turnover cycle of a plant is important, because it controls both the duration of forage availability to animals and the period during which the plant accumulates radionuclides. All the forage vegetation in this study consisted of perennial plants--species that endure from year to year. In the case of grasses, the top portion of the plant decays at the end of the first season of growth, and then the grass renews itself with shoots from the base. Shrubs such as four-wing saltbush have less of their overground vegetation decaying due to their predominantly woody stems. Because of this, grasses probably recycle more of their accumulated activity back to the soil than shrubs. Because all the grasses under consideration are perennials, they are readily available for foraging from year to year.

The growth of vegetation on mill tailings, such as those at Anaconda, is much slower than that on undisturbed soil. This is probably due to the lower pH of the tailings and/or the presence of toxic substances (10, 15). Thus, in areas with high tailings concentrations, less forage is available, although the activity of the existing plants is elevated. At the southeast corner of the tailings pond at Anaconda, the area with highest tailings contamination, only a small amount of Indian ricegrass was found.

The uptake of thorium, radium, lead, and uranium by vegetation on the mill site varies from less-than-detectable amounts to concentrations of over 100 pCi/g. Many plants growing on tailings-contaminated soil concentrate more Pb-210 than the other nuclides (Table 1). This has also been observed by Winsor and Whicker (16).

Table 1 shows three sites (the southeast, northeast, and southwest corners of the tailings pond) that contain the vegetation most contaminated with tailings. The plants with the highest activity were obtained from the southeast location. The areas of highest soil activity are the east and southeast sides of the pond. Ra-226 and Pb-210 constitute about 70% of the measured soil activity at these locations. These two radionuclides are in near equilibrium.

The uptake coefficients for the forage species studied are given in Table 3. The uptake (transfer) coefficient is an indicator of the fraction of activity that nas moved from the soil to the plant. The Pb-210 coefficients are the highest for most species; uranium and radium are taken up in lesser amounts. The lack of data for Th-230 concentrations in soil prevents estimations of thorium uptake coefficients for most of the plants. Table 1 seems to indicate that thorium, considered not readily assimilated by vegetation, can be accumulated significantly in some instances. This can be inferred from a comparison of the radionuclide concentrations within the same plant species, e.g. Indian ricegrass or blue grama grass (Table 1).

Many foraging animals graze only on the shoots, or aboveground portions, of a plant. Other grazing livestock, such as sheep, often tear the roots out of the ground and consume the whole plant. This is particularly true with grasses. Table 4 lists the distribution index, i.e., the ratio of root activity to shoot activity, for various species. In nearly all cases, the root activity is greater than that of the shoots.

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TABLE 1. Concentration vs. Dry Weight (pCi/g) of Radionuclides in the U-238 Series in Samples of Forages Collected Around the Anaconda Tailings Pond, Northern Boundary (NB), and at a Control Location (C)

Vegetation Species	Forage Value	Location	U-238	U-234	Th-230	Ra-226	Pb-210
Galleta Grass (*Hilaria jamesii*)	Excellent	NW	0.25±0.03	0.42±0.03	0.16±0.01	BDL	1.27±0.28
Galleta Grass roots		NE	0.91±0.02	1.54±0.01	4.52±0.06	0.95±0.01	2.30±0.04
Russian Thistle (*Salsola paulsenii*)	Bad	W	0.96±0.02	0.59±0.02	0.03±0.003	0.08±0.002	4.16±0.30
Russian Thistle		W	0.29±0.01	0.24±0.01	0.01±0.004	0.22±0.004	BDL
Blue Grama Grass (*Bouteloua gracilis*)	Excellent	NW	0.23±0.02	0.52±0.01	0.12±0.003	BDL	BDL
Blue Grama Grass		SW	3.38±0.01	3.92±0.01	2.69±0.06	2.15±0.01	4.16±0.07
Indian Ricegrass (*Oryzopsis hymenoides*)	Excellent	SE	2.11±0.01	2.63±0.01	13.2±0.25	3.43±0.01	16.0±0.02
Indian Ricegrass roots		SE	9.89±0.01	12.5±0.01	51.8±1.3	130±0.18	81.5±0.03
Squirreltail Grass (*Sitanion hystrix*)	Good	NB	0.44±0.01	0.39±0.02	0.19±0.007	0.06±0.002	1.54±0.24
Dropseed Grass (*Sporobolus asper*)	Excellent	NB	0.30±0.04	0.64±0.03	0.24±0.02	0.04±0.002	0.45±0.40
Dropseed Grass roots		NB	0.99±0.03	0.63±0.04	0.75±0.03	0.18±0.008	BDL
Russian Thistle		C	0.33±0.008	0.38±0.008	0.09±0.006	0.03±0.001	BDL
Four-Wing Saltbush (*Atriplex canescans*)	Excellent	C	0.04±0.07	0.07±0.05	BDL	BDL	BDL
Sticky Snakeweed (*Gutierrezia lucida*)	Bad (Toxic)	C	BDL	BDL	0.03±0.003	0.08±0.002	BDL

BDL = below detectable limits

Table 2. Concentration (pCi/g in Dry Soil ± Error) of Radionuclides in the U-238 Series Measured by Gamma Spectroscopy (U-238, Th-230, Ra-226, and Pb-210) and Radiochemical Techniques (Ra-226, Pb-210)

	Gamma Spectroscopy				Radiochemical	
Location	U-238	Th-230	Ra-226	Pb-210	Ra-226	Pb-210
E	14.9±8.38	76.0±11.0 86.9±0.06(L)	177±4.1	166±7.5	150±0.01	116±0.01
E	18.2±7.76	104±31.1	166±3.6	156±8.3	104±0.02	71.7±0.15
S[a]	2.80±0.44	BDL[b]	3.51±0.71	5.02±0.67	BDL	BDL
E	17.7±8.55	78.5±29.8	133±2.5	120±2.8	131±0.002	105±0.12
E	10.9±6.15	45.5±18.3	77.5±2.09	78.3±4.30	95.8±0.003	93.01±0.22
SE	18.0±9.41	115±44.6	236±5.0	209±10.5	176±0.002	153±0.08
W	2.70±1.77	BDL	0.87±0.50	2.56±0.47	BDL	BDL
S	6.24±3.53	BDL	22.9±1.24	23.56±1.3	70.5±0.002	BDL
C	2.97±1.72	BDL	0.61±0.18	1.40±0.30	0.76±0.05	BDL

[a] Two kilometers to the southwest.

[b] BDL = below detectable limits.

$$\pm \text{ error} = \frac{\sqrt{\text{total counts/total count time (min)}}}{\text{net count/min}}$$

Table 3. Uptake Coefficients of Selected Radionuclides in Forage

Vegetation Species	Location	U-238	Th-230	Ra-226	Pb-210
Russian Thistle	W	0.11		0.25	
Indian Ricegrass	SE	0.12	0.11	0.01	0.08
Russian Thistle	C	0.11		0.04	0.54
Four-Wing Saltbush	C	0.01			
Sticky Snakeweed	C			0.11	0.53

Table 4. Comparison of Roots with Shoots Expressed by Distribution Index

Vegetation Species	U-238	Th-230	Ra-226	Pb-210
Galleta Grass	0.82	0.30	6.61	1.15
Blue Grama Grass	1.65	10.50		
Indian Ricegrass	4.69	3.93	38.0	5.08
Dropseed Grass	3.30	3.13	4.50	

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