Accumulation of Metals and Radium-226 by Water Sedge Growing on Uranium Mill Tailings in Northern Saskatchewan

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ABSTRACT

The Lorado uranium mill tailings, near Uranium City, Saskatchewan are characterized by low pH, high salinity and high concentrations of elements such as copper, nickel, lead, uranium, vanadium and zinc. In 1982, we measured the concentrations of 30 elements in plant tissues of water sedge (<u>Carex aquatilis</u> Wahl.), a natural invader of wet tailings. Concentrations of copper, lead, uranium and vanadium in plants on the tailings were higher than in plants from background sites off the tailings. More detailed analyses of these elements in 1983 showed that relative concentrations in plant parts for all four metals were: roots > rhizomes > green leaves. Radium-226 and uranium were elevated in aboveground tissue (leaves) compared to samples from background sites off the tailings.

INTRODUCTION

The first uranium claims in the Uranium City area, staked in 1945 at Fish Hook Bay on the northern shore of Lake Athabasca, led to intensive exploration in the region. Mills at Lorado, Gunnar and Eldorado operated from 1957-60, 1955-63 and 1953-82 respectively, before abandonment (Woods 1981, Kupsch 1978).

The Lorado mill was designed to process 635 tonnes of ore per day from the Lorado mine and several other small mines, however this capacity was never fully realized. While in operation the mill processed a total of 545,000 tonnes of ore with an average grading of 0.24% U₃0₈. Ore composition varied greatly because of the diversity of sources. Ore was crushed to 60% minus 200 mesh and leached with sulphuric acid to dissolve the uranium minerals. Sulphuric acid was initially produced at the Lorado mill site using pyrite mined at the Lorado mine site, and later from sulphur shipped from Alberta (Whiting et al. 1982).

After removal of most of the uranium, the waste materials were pumped in a slurry to the waste disposal pond adjacent to Nero Lake. There were no presite disposal preparations and the tailing slurry (pH of 1.7-2.0) received no treatment prior to disposal. The tailings are acidic, saline and have elevated levels of several heavy metals including lead, uranium and vanadium (Kalin 1981). No stabilization of the tailings area (14.2 ha) has been attempted since the termination of the operation and exposed tailings material is subject to transport by air, surface and ground water (Swanson and Abouquendia 1981).

An additional problem associated with uranium mill tailings is the presence of radionuclides. Of particular concern are radium-226, radon, thorium-230 and lead-210. Some uranium and most of the radionuclides pass from the mill to the tailings area (Merritt 1971). Ripley and Redmann (1978) estimate that 14% of the total radioactivity is contained in the final processed concentrate, and 50 to 86% in the tailings, depending on the proportion of radon lost.

The entry of radionuclides into food chains is of particular concern in the case of uranium mining. Abouguendia et al. (1979) recorded elevated levels of U^{238} , Pb^{210} and Ra^{226} in meadow voles collected from a stand of sedges (<u>Carex rostrata and C. aquatilis</u>) and grasses over a uranium ore body in the Cluff Lake area of Saskatchewan. Meadow voles feed on insects, seeds and the vegetative parts of sedges, which dominate the site. Analysis of the sedges indicated elevated levels of U^{238} , Pb^{210} and Ra^{226} , which may have contributed to the elevated level of these elements in the meadow voles. Thus, tailings revegetation, although providing an effective means of reducing wind and water erosion, can create additional pathways for the movement of radionuclides and heavy metals into the biosphere.

Natural revegetation of the Lorado tailings site is sparce, and restricted to areas along the tailings edges. The main species include bluejoint (<u>Calamagrostis canadensis</u>), water sedge (<u>Carex aquatilis</u>), common cattail (<u>Typha latifolia</u>) and birch (<u>Betula papyifera</u>); secondary species are: <u>Betula neoalaskana</u>, <u>Calamagrostis inexpansa</u>, <u>Equisetum</u> spp., Festuca rubra and Salix planifolia (Swanson and Abouguendia 1981).

<u>C. aquatilis</u> is also a natural invader of uranium tailings in other regions of Saskatchewan and Canada (Harms 1982, Kalin and Caza 1982).

Densely tufted, with long creeping rhizomes, it occurs throughout Canada, commonly growing in slough margins, river flats, marshes and wet meadows (Moss 1983, Porsild and Cody 1980).

The objectives of this study were twofold: (1) to determine if \underline{C} . <u>aquatilis</u> growing at the tailings site accumulated elevated levels of heavy metals and radionuclides; and (2) to determine if metals and radionuclides were concentrated in particular plant parts.

MATERIALS AND METHODS

Sampling Procedure

Samples of <u>C</u>. <u>aquatilis</u> were collected from the Lorado mill tailings site and the Uranium City area in August 1982 and June 1983. Tailings and soils were collected from the root regions of sample plants. Plant samples collected in August 1982 were separated into leaves and a composite of roots and rhizomes. Samples collected in June 1983 were separated into three fractions: leaves, roots and rhizomes.

Sample Analysis

Oven dry (105°C) plant material, tailings and soils collected in 1982 were analyzed for 30 elements using plasma emission spectroscopy. Samples collected in 1983 were analyzed for copper, lead and vanadium using atomic absorption spectroscopy, for uranium using delayed neutron counting and for radium-226 using alpha counting.

Tailings and soils were air dried and sieved to remove particles greater than 2mm. Hydrogen ion activity (pH) and conductivity were determined in a 1:1 mixture of 20 g of air dry material and 20 ml of

deionized distilled water agitated for 1 h. Moisture content was determined after a fresh weight sample was oven dried at $105^{\circ}C$ for 24 h. Available sulphate was extracted by agitating 25 g of sample with 50 ml of 0.001M CaCl₂ for 0.5 h. The extract was analyzed for sulphate using atomic absorption spectroscopy. Particle size analysis was performed using the hydrometer method (Boyoucos 1951).

An unpaired t-test (0.05 level, population variances assumed unequal) was used to compare element concentrations in tailings, soils and vegetation samples from on and off the site. This test was also used to compare concentrations in different plant parts.

RESULTS

Some of the ores milled at Lorado contained pyrite which when exposed to the environment is oxidized producing sulphuric acid. This and the use of sulphuric acid in the milling process has resulted in acidic conditions and extremely high available sulphate levels throughout the tailings site (Table 1). According to Richards' (1954) classification of saline soils, the tailings site is moderately saline. Tailings contained less sand and more silt and clay than local soils. Available nitrogen, phosphorus and potassium levels were very low while copper and zinc levels were elevated in tailings compared to soils vegetated with C. aquatilis (Frankling 1984).

Tailings and soils from the root regions of <u>C</u>. <u>aquatilis</u> were analyzed for 30 elements in 1982. Tailings had significantly higher concentrations of several elements including copper and lead, compared to soils in the Uranium City region (Table 2). In 1983, elevated concentrations of copper, lead, radium-226, uranium and vanadium were observed in tailings, compared

	рĦ	Conductivity (ms cm ⁻¹)	Moisture Content (%)	s - s0 ₄ (μg g ⁻¹)	Sand	Silt (%)	Clay
Tailings	3.6(4.0)*	3.4(1.2)*	27.6(7.5)	2400 (1860) *	56.1(22.1)*	37.7(21.6)*	6.0(2.4)*
Soils	5.6(5.7)	1.5(1.5)	48.1(38.3)	420 (260)	91.0(2.5) ^Δ	6.0(2.1) [∆]	2.9(0.5) ^Δ

Table 1. Chemical and physical characteristics of tailings and soils from the root regions of \underline{C} . aquatilis.

*Significant difference (0.05 level) between tailings and soil

 $^{\Delta}n = 6$

Element	Tailings $(n^2 = 4)$ $(\mu g g^{-1} or)$	Soils (n = 4) ven dry wt)	Significance level
Al	56500 (7593)	14550 (6681)	.0004*
в	12.6(47)	77 (16.4)	.0054*
Ba	502 (69)	100(41)	.0006*
Ca	17600 (19675)	30650 (1837)	.2783
Cr	116(28)	21.5(8.8)	.0083*
Cu	645 (256)	12.5(6.4)	.0161*
Fe	43750 (11176)	12517 (8711)	.0126*
K	5925 (1654)	3625 (1406)	.0876*
Mg	9725 (1105)	6550 (2693)	.1172
Min	235 (73)	87 (30)	.0334*
Na	26250 (6291)	5312(332)	.0069*
Ni	239(141)	14.5(2.3)	.0839
Pb	330(121)	12.9(3.4)	.0458*
Ti	2875(512)	438(180)	.0029*
Zn	465(210)	100 (53.6)	.048 *
Zr	260 (84)	NA ³	

Table 2. Concentrations of elements in tailings and soils from the root regions of <u>C</u>. aquatilis collected in 1982.

¹The following elements were below detection limits in tailings: Ag, As, Au, Be, Cd, Co, Mo, P, Sb, Se, Th, U, V and W.

²Sample size

³Not analyzed

*Indicates significant difference (0.05 level) between tailings and soil.

to soils vegetated with C. aquatilis (Table 3).

Several elements including lead, uranium and vanadium appear to be elevated in roots/rhizomes from the tailings site compared to the background sample (Table 4). Leaves of <u>C</u>. <u>aquatilis</u> collected from the tailings site had higher concentrations of several elements including nickel, lead and uranium compared to background samples, however, manganese was the only element to be significantly elevated in leaves from the tailings site based on statistical tests. After entering <u>C</u>. <u>aquatilis</u> plants, elements appeared to accumulate in belowground tissue. Aluminum and iron were significantly higher in roots/rhizomes than leaves collected at the tailings site, and several other metals, including copper, lead, uranium and vanadium displayed a similar trend, although not statistically significant (Tables 4 and 5).

Roots and rhizomes of <u>C</u>. <u>aquatilis</u> from the tailings site had relatively higher concentrations of copper, lead, vanadium and particularly uranium compared to samples collected in the Uranium City area (Table 6). A comparison of copper and lead concentrations in <u>C</u>. <u>aquatilis</u> leaves collected on and off site indicated no significant differences (Table 6). Uranium and radium-226 concentrations were elevated in leaves collected at the tailings site (significant at the 0.05 and 0.08 levels respectively).

A comparison of roots, rhizomes and leaves was performed to determine where in plants elements were accumulating. Vanadium was statistically higher at the 0.05 level in aboveground tissue (leaves) compared to belowground tissue (roots) (Table 7). However, lead and uranium also appear to accumulate in roots compared to leaves. Differences in metal levels between roots and rhizomes and rhizomes and leaves are less evident (Table 7). The general trend is for element concentrations to decrease in the order: roots > rhizomes > leaves.

	Cu	Pb	U	v	Ra-226 (Bq g ⁻¹ Oven
	dry wt.)				
Tailings (n ¹ =4)	71(18)	307 (145)	112(43)	430 (158)	26.5 (12.0)
Soils (n=4)	22(4.3)	11.5(5.8)	6.6(2.5)	55(17.3)	.24(.17)
Significance level	.0146*	.0266*	.0167*	.0183*	.0224*

Table 3. Concentrations of elements in tailings and soils collected in 1983.

¹Sample size

*Significant difference (0.05 level) between tailings and soil.

Element	Tailings site (n ² =4) 1 (µg g ⁻¹ Oven d	
Ag	.6(.2)	.1
AL	667 (122)	900
в	7.2(3.7)	7
Ba	10.6(2.2)	28
Be	.4(-)	.7
Ca	860 (157)	1900
Co	3(1.8)	2.3
Cr	3.1(1.3)	3
Cu -	21(9.8)	10
Fe	1500 (522)	3000
ĸ	6875(573)	5800
Mg	1675(573)	930
Min	182(43)	190
Na	355(189)	340
Ni	8.6(5)	5
P	1087(154)	970
Pb	21(10)	6
Se	40(-)	1
Ti	6.9(2.2)	52
U	63 (65)	3
v	12.5(2.8)	4
Zn	24 (29)	29
Zr	1.3(.4)	1.4

Table 4. Concentrations of elements¹ in roots and rhizomes of \underline{C} . <u>aquatilis</u> collected in 1982.

The following elements were below detection limits in tailings: As, Au,

	Tailings	Background	Significance values					
Element	site area $(n^{2}=4)$ $(n = 4)$ $(\mu g g^{-1}Oven dry wt.)$		Leaves on site-off site	Roots/rhizomes - leaves on site				
Ag	1.6(2.3)	< .1(-)	4	.4858				
Al	247 (92)	132(50)	.0931	.0031*				
в	17(7.4)	31.2(3.5)	.0261*	.0796				
Ba	6.1(2.8)	15.2(2.2)	.0038*	.0563				
Be	.5(-)	.2(-)	-	-				
Ca	2325 (464)	4400 (374)	.0009*	.0094*				
Co	1.4(0.8)	.48(-)		-				
Cr	1.1(0.4)	2.4(1)	.1059	÷				
Cu	12.7(3.8)	8.4(2.3)	.4926	.1945				
Fe	305 (86)	205 (57)	.1131	_0204*				
K	10175 (2275)	10800 (1036)	.6433	.0587				
Mg	2765 (1602)	1475(95)	.2319	.3248 -				
Mn	710 (200)	77 (22)	.0082*	.0143*				
Na	267 (167)	350(21)	.4009	.5203				
Ni	5.9(3.4)	1.6(1.2)	.1017	.4714				
P	1067(47)	1227 (400)	.4854	.8207				
Pb	6.8(4.3)	1.6(1.1)	.1061	.0617				
Ti	1.4(0.1)	2.8(1.1)	.8060	.0159*				
U	19.6(22)	0.3(.08)	.2813	.3044				
v	2(-)	< 2(-)	.5443	.5188				
Zn	29 (24)	37.5(2.6)		-				

Table 5. Concentrations of elements ¹ in in 1982.	leaves	or \underline{C} .	aquatilis colle	ected
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¹The following elements were below detection limits in tailings: As, Au, Cd, Mo, Sb, Se, Th, W and Zr.

²Sample size.

*Indicates significant difference (0.05 level) between on and off site samples.

Location	Plant part	Cu	Cu Pb U			Ra-226 (Bq g-1			
	(µg g ⁻¹ Oven dry wt.) Oven dry wt)								
Tailings site	roots (n ¹ = 3)	61 (24)	19.3(5.6)	252 (138)	19.6(4.0)	NA ²			
Background area	roots (n = 2)	11 (8.4)	7.0(4.2)	0.6(0.1)	<5.0	NA			
Tailings site	rhizomes (n = 4)	34.5(8.8)	13.7(7.0)	148 (106)	15.7(8.7)	NA			
Background area	rhizomes (n = 2)	4.4(3.0)	7 (0)	0.5(0.07)	<5.0	NA			
Tailings site	leaves $(n = 4)$	26.5(13)	9 (5.3)	61(39)	6.2(1.5)	0.13(0.07)			
Background area	leaves $(n = 4)$	14.5(2.5)	5.1(2.1)	0.26(0.05)	<5.0	0.03(0.			
Significance level	leaves	0.1750	0.4863	0.0518	÷	0.0749			

Table 6.	Concentrations of elements in roots, rhizomes and leaves of	
	C. aquatilis collected in 1983.	

¹Sample size, except U: n = 4. ²Not analyzed.

AR				
AR	Cu	Pb	U	v
Rhizomes roots	0.56	0.70	0.58	0.80
Leaves rhizomes	0.76	0.65	0.41	0.39
Leaves roots	0.43	0.46*	0.24*	0.31**

Table 7. Accumulation ratios (ARs) for roots, rhizomes and leaves of C. aquatilis growing at the Lorado tailings site, collected in 1983.

*Significant difference between levels in roots and leaves at the 0.10 level.

** Significant difference between levels in roots and leaves at the 0.05 level.

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DISCUSSION AND CONCLUSIONS

A number of trace elements, including arsenic and vanadium, are frequently enriched in uranium ore deposits. Other trace metals, including copper, nickel and zinc are also often enriched due to the presence of base metal sulphides associated with pyrite (Dressen et al. 1982). Tailings beneath <u>C. aquatilis</u> growing at the site had elevated concentrations of several metals, including copper, lead, uranium and vanadium. The radionuclide radium-226 was also elevated in tailings.

Belowground tissue of <u>C</u>. <u>aquatilis</u> growing at the tailings site had elevated levels of copper, lead, vanadium and particularly uranium compared to background samples. Copper, uranium and vanadium values were also higher than values considered normal for land plants, as reported in Bowen (1979). Lead concentrations were similar to values for land plants.

Aboveground tissue of <u>C</u>. <u>aquatilis</u> growing at the tailings site had elevated levels of uranium and radium-226 compared to background samples. Uranium levels also were elevated compared to reported values for <u>Carex</u> spp. (<3.0 μ g g⁻¹ash weight) growing near uranium anomalies on the Precambrian Shield (Sheppard et al. 1981), and in <u>C</u>. <u>aquatilis</u> and <u>C</u>. <u>rostrata</u> (0.04-1.2 and 0.04-7.0 μ g g⁻¹ ash weight respectively) growing adjacent to Cluff Lake, Saskatchewan in the vicinity of the Cluff Lake uranium mine (Abouguendia et al. 1979). Radium-226 values in leaves were similar to values for aboveground tissue of <u>C</u>. <u>aquatilis</u> (0.001-0.921 Bq g⁻¹ ash weight) and <u>C</u>. <u>rostrata</u> (0.004-0.307 Bq g⁻¹ ash weight) growing in the Cluff Lake area (Abouguendia et al. 1979). Concentrations of copper, lead, uranium and vanadium in plants growing at the tailings site tended to decrease in the order: roots > rhizomes > leaves. This is in agreement with the behaviour

of lead in ryegrass and uranium in barley (Jones et al. 1972, Van Netten and Morley 1983).

In conclusion, copper, lead, uranium, vanadium and radium-226 levels were elevated in <u>C</u>. <u>aquatilis</u> growing at the tailings site. Belowground tissue provides an effective barrier to the movement of elements to aboveground plant parts, and only uranium and radium-226 appeared to be elevated in aboveground tissue. The presence of these two elements in leaves of <u>C</u>. <u>aquatilis</u> suggests that further study on their fate is appropriate in tailings areas in Canada where this species is growing.

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