INVESTIGATING CADMIUM BIOAVAILABILITY IN NW ONTARIO USING BOREAL FOREST PLANTS

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ABSTRACT: Here we document cadmium concentrations in terrestrial and aquatic vegetation growing in boreal forested terrains of NW Ontario. Cd concentrations in plant samples from our study areas do not exceed the range (0.1 to 1 µg/g) normally found in land plants, except when growing in soils or sediments formed or deposited in close association with Cd-mineralized rocks. We also discuss the viability of higher herbivorous fauna such as moose as bioindicators.

ALCES VOL. 30 (1994) pp.71-80

This study was prompted by reports of cadmium levels in liver and kidney tissue of deer and moose exceeding FAO/WHO daily intake levels recommended for humans (for example, Crête *et al* 1987). In these studies Cd was considered carried to moose and deer habitats in air masses from industrial areas. Our study explores the bioavailability of Cd to herbivores in NW Ontario where regional air streams generally do not derive from industrial regions. In the first part of our research (1987 and 1988) we measured Cd concentrations in terrestrial and aquatic plants growing in soils and sediments formed in close association with Cd-mineralized rocks. In 1988 and 1989 this work was extended to include plants growing along major highways in NW Ontario.

Over the past 15 years several researchers reported Cd concentrations of up to 60 µg/g in kidney and liver tissues of northern hemisphere cervides (Crête et al. 1987; Frank and Petersson 1984; Froslie *et al.* 1986; Glooschenko *et al.* 1988; Grodzinska 1978; Sawicka-Kapusta 1981; Scanlon *et al.* 1986). Negligible amounts (<0.1 µg/g) of cadmium were found in moose and deer muscle (Glooschenko and Burgess 1987). Frank and Petersson (1984) designed an extensive (>4,000 animals) study to obtain reference values for Cd bioavailability to higher Swedish fauna using moose as bioindicators. Their findings indicate kidney tissue as the target organ for Cd accumulation.

Biological and geochemical pathways of Cadmium (Cd) at the Earth's surface are poorly understood. In the biosphere Cd is usually chemically tethered to the nutrient element, Zn, due to its being in the same group (IIB) of chemical elements. This close association is prevalent in rocks and soils; however, in the biosphere Cd generally exhibits a stronger bioaffinity than Zn, i.e. Cd/Zn concentration ratios are higher in plants than in surface rocks. Cd like Zn is sometimes considered a bioessential trace element (Kovalevsky 1984; Mertz 1981), and the inclusion of Zn data in this study of soil (sediment)/plant cadmium partitioning provides important chemical information for assessing the viability of Cd as a bioindicator.

STUDY AREA and METHODS

Site Selection - In the initial research (1987-88), we selected 2 sites to compare Cd levels in "moose-preferred" terrestrial and aquatic plants growing in soil and sediment associated with cadmium-mineralized rocks with Cd concentrations in plants of the same species growing in non-mineralized soil and sediment. These sites are located near Manitouwadge, NW Ontario (Fig. 1 and 2), where cadmium-bearing rocks are mined (Bakker *et al.* 1984). Atmospheric loading of cadmium in this area is close to background, except within a few hundred meters of the concentrator at the current mine site (Racette and Griffin 1979).

The mineralized terrestrial site (Willroy Area, Figure 1b, 49° 10'N, 85°50'W) is adjacent to an abandoned mine pit. Most vegetation samples from the mineralized site grew a few meters from the edge of the pit; the remainder were from the pit itself. The mineralized aquatic site (Garnet Lake, Fig. 1) receives water directly from creeks draining the mineralized area. The control site (One Otter Lake, Fig. 2) is located ~20 km to the northeast of the Willroy area. At One Otter Lake both terrestrial and aquatic vegetation samples were taken from within a few meters of the shoreline. Here bedrock is covered by several meters of carbonate-rich glacial sediments commonly observed in the Manitouwadge area (Hicock 1986). The vegetation cover at all sites, including the shallow mine pit, has not been significantly disturbed for at least 2 decades. There was little variation in topography at all sites.

For the roadside study (1988-89), we chose five sites with a range (1,050 to 17,000) of AADT (averge annual daily traffic) along major highways in the vicinity of Thunder Bay (Fig. 3). We collected only terrestrial vegetation samples at these sites.

Vegetation Sampling and Analysis - The selection of terrestrial species was based on information gathered by McNichol and Gilbert (1980) on moose upland browse in the vicinity of Thunder Bay, NW Ontario. These species included balsam fir (*Abies balsamea*), white birch (*Betula papyrifera*), trembling aspen (*Populus tremuloides*), mountain maple (*Acer spicatum*), red osier dogwood (*Cornus stolonifera*), bush honeysuckle (*Diervilla lonicera*), pin cherry (*Prunus pensylvanica*) and american mountain ash (*Sorbus americana*). At the Manitouwadge

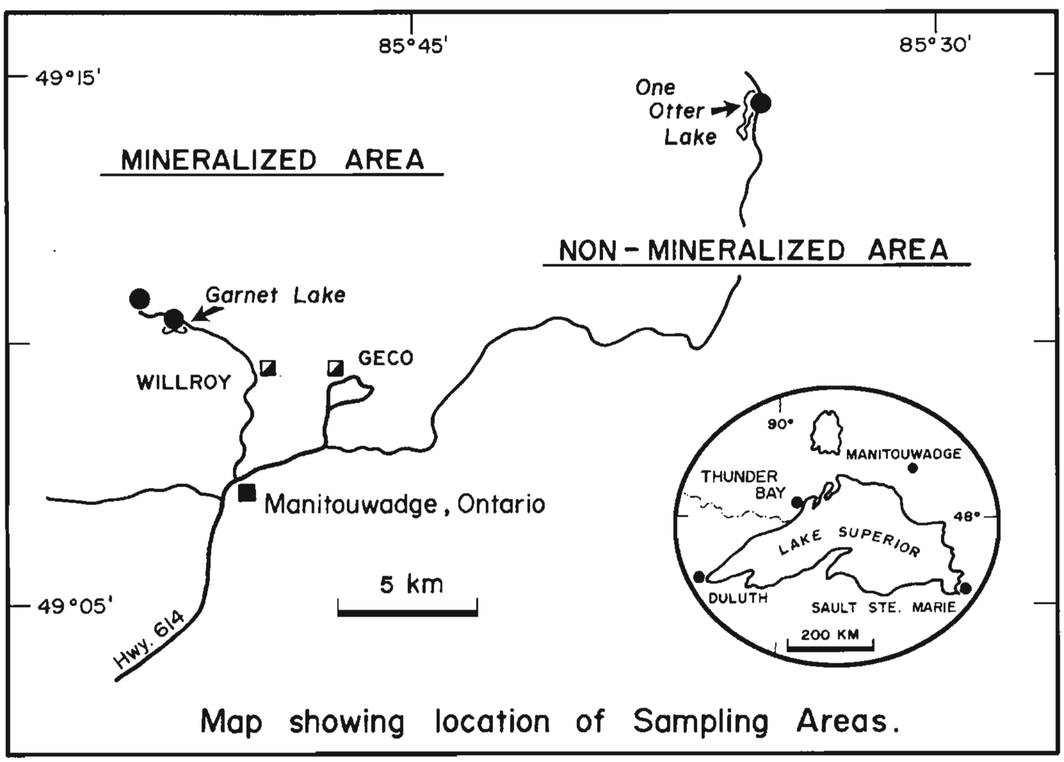

Fig. 1. Sketch Map Showing Regional Context of Mineralized and Non-mineralized Sites.

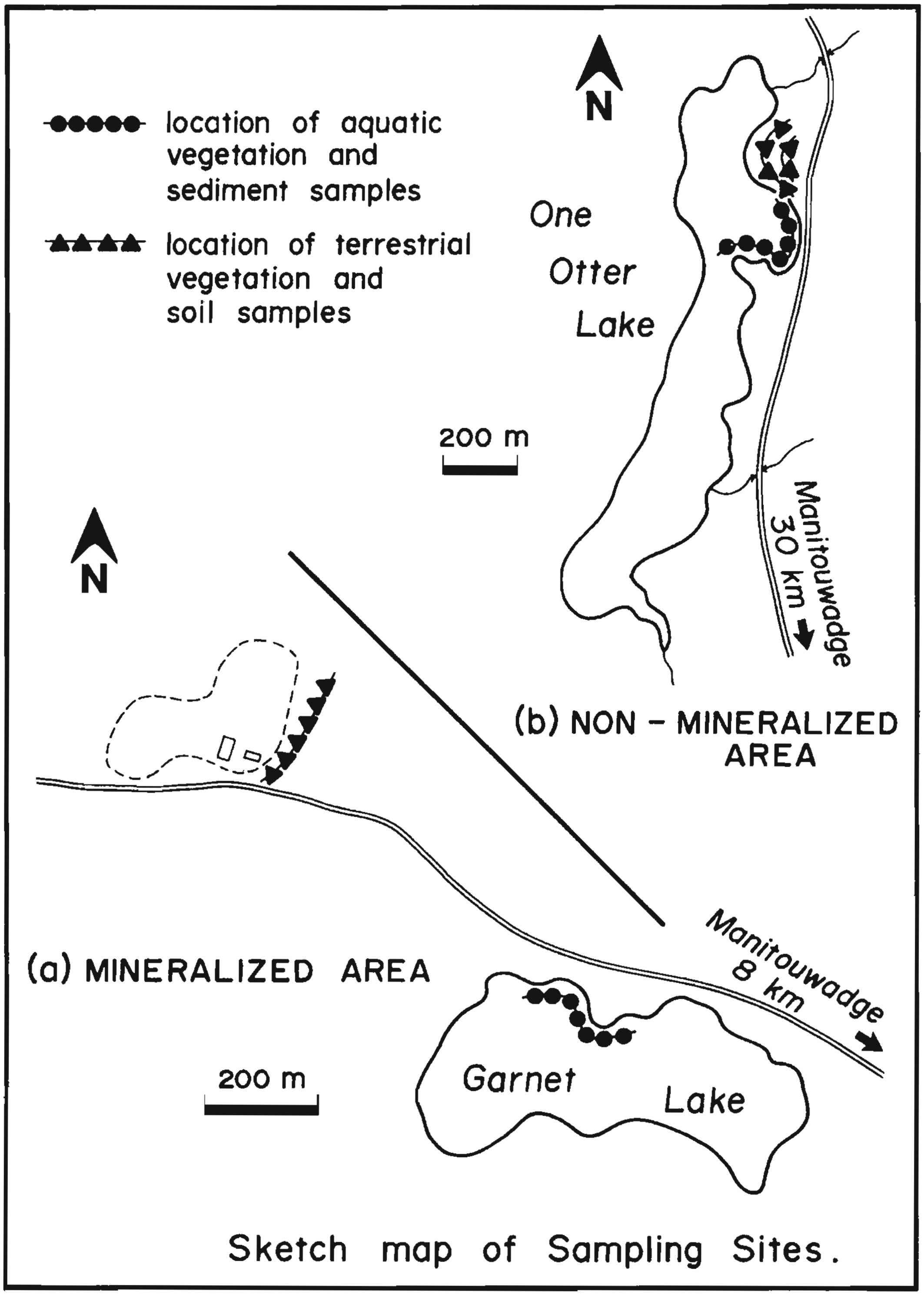

Fig. 2. Sketch Map Showing Mineralized Terrestrial (Willroy) and Aquatic (Garnet Lake) and Non-mineralized (One Otter Lake) Sampling Sites.

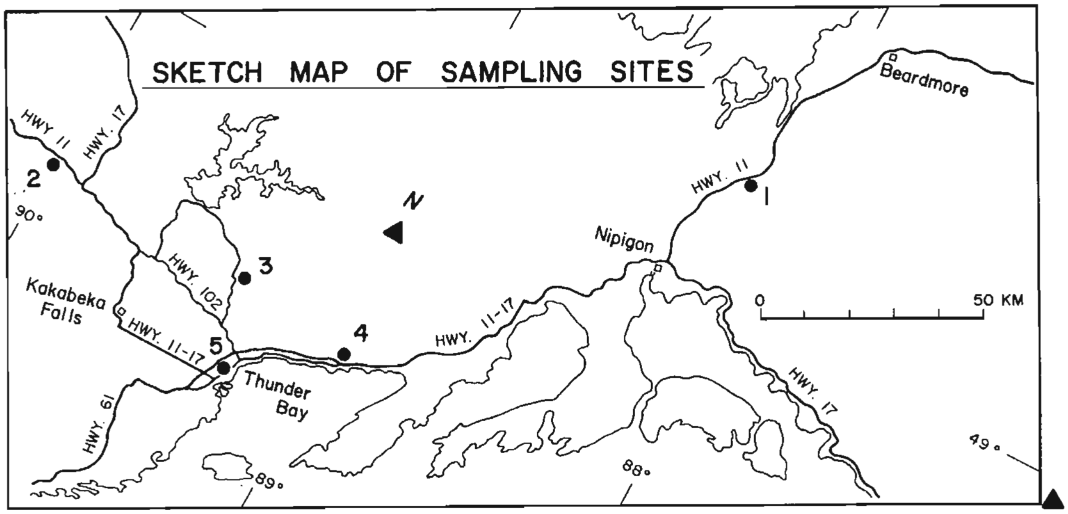

Fig. 3. Map Showing Location of Roadside Sites.

sites all vegetation samples were collected within 3 days in July, 1987. Terrestrial plants collected in the roadside study include only those plants (trembling aspen, balsam fir, white birch, bush honeysuckle), for which >1.0 µg/g Cd levels were noted in the first part of the study.

Selection of aquatic plant samples was based on observations of moose feeding habits in lakes and ponds in NW Ontario (Fraser *et al.* 1984). Plants designated as "preferred" or "intermediate preferred" were sampled: milfoil (*Myriophyllum exalbescens*), floating pondweed (*Potamogeton natans*), Richardson's pondweed (*P. richardsonii*) and red-disced waterlily (*Nuphar rubrodiscum*). Samples of cattail (*Typha latifolia*) designated as "unpreferred" were also collected. At each lake we measured pH, dissolved oxygen, conductivity, temperature and took water samples for subsequent dissolved cadmium analysis.

Terrestrial vegetation samples included leaves and twigs from most recent growth. These were air-dried for ~3 weeks followed by oven-drying for 3 days at 40° C. Aquatic samples were put in plastic bags and frozen. For analysis these samples were thawed for one day, then dried for 2 days at 40°C and for 3 more days at 60°C. To avoid leaching of metals all samples were left unwashed and adhering soil and sediment particles were shaken off or removed by hand. We divided terrestrial vegetation samples into twigs and leaves (needles) and some roots and flower heads of aquatic plants were also treated as separate samples.

Vegetation samples were ground in a Tomas-Wiley mill with a 2 mm mesh screen and stored in sterilized whirlpak bags. The prepared samples were digested by wet ashing for cadmium and zinc analysis by atomic absorption spectrometry (AAS). Samples, for which AAS concentration measurements did not exceed 1 µg/g, were analysed by inductively coupled plasma spectrometry (ICP). (AAS and ICP are standard analytical chemical techniques for determining metal concentrations in aqueous solutions.)

Soil and Sediment Sampling and Analysis - At each terrestrial site, we sampled soils along the traverse used for vegetation samples. Each composite soil sample was taken from 8 compass points along 2 concentric circles (radii 0.5 m and 1 m). Sediment samples at the aquatic sites (Fig. 2) were taken with a spring-loaded grab sampler. Soil and sediment samples were air dried, then ground to ~100 mesh using a mortar and pestle. Concentrations of major minerals and major elements were determined by x-ray diffraction (XRD) and x-ray fluorescence (XRF) respectively. (XRD and XRF are standard analytical techniques for identifying mineral constituents and element concentrations, respectively, in solid samples.) Concentrations of cadmium and zinc were measured using AAS.

Airborne Cadmium Fluxes - Cadmium atmospheric inputs were monitored using moss bags, hung for 6 weeks from tree branches ~1.5 m above ground. This procedure is followed routinely by the Ontario Ministry of Environment (MOEE).

Long-term atmospheric fluxes were assessed with samples of Schreber's moss (*Pleurozium schreberi*) collected at the study areas. Moss samples were prepared and analysed using the same procedures as those for vegetation samples.

RESULTS

Cadmium Concentrations in Vegetation - The highest Cd value (60 µg/g) was measured in one organic soil sample collected from within a few metres of the abandoned Willroy mine pit (Fig. 1). Of the terrestrial plants sampled (Table 1), the highest Cd levels were found in trembling aspen (19 and 24 µg/g) growing in the mine pit, in milfoil (19 µg/g) from Garnet Lake at the mineralized site and in common bladderwort (16 µg/g) from One Otter Lake at the non-mineralized site. Only these five samples have Cd concentrations well outside the range (0.1-1 µg/g) generally found in land plants (Wedepohl, 1970). Otherwise Cd levels in samples from both mineralized and non-mineralized sites were in the low, one to five, microgram per gram range (balsam fir, white birch, trembling aspen, bush honeysuckle, Richardson's pondweed, Table 1) or even lower (<0.05 to 0.4 µg/g) in samples of mountain maple, red osier dogwood, cattail, common bladderwort and red disced waterlily. For 2 species (balsam fir and bush honeysuckle) found at both mineralized and non-mineralized sites mean Cd concentrations in the former site (1.0-3.0 µg/g) were enhanced relative to the latter (0.3-0.4 µg/g) (Table 1). Trembling aspen and some white birch samples found only at the mineralized site also exhibit elevated mean values (2.8-5.3 µg/g) relative to background values (0.1-1 µg/g; Wedepohl, 1970). Aquatic species sampled give a mixed signal in that Richardson's pondweed for the mineralized site contains more Cd at the non-mineralized site while the reverse is noted for samples of common bladderwort. No variations in Cd concentrations were noted for different parts (leaves, roots, stems, flower heads) of aquatic plants. For every case of enhanced Cd concentrations in both terrestrial and aquatic samples Zn levels were also greater relative to background Zn values. Because of the close chemical association between Zn and Cd these concomitant measurements are important to corroborate concentration patterns.

Cadmium Concentrations in Soil and Sediment - Cadmium concentration measurements in 15 soil samples and 10 lake sediment samples from the mineralized (Willroy) area yielded means of 7.7 and 7.1 µg/g respectively (Table 1). Corresponding data from the non-mineralized site yielded means of 0.6 and 0.5 µg/g Cd respectively. The mean Cd concentration (0.2 µg/g) from 48 roadside samples is of the same order of magnitude as the value for non-mineralized soil samples.

Concentration values for Cd and Zn in soil and sediment samples are closely correlated (r>0.9) and their concentration ratios ~200:1 (Zn:Cd) is of the order of magnitude generally observed in rocks and soils (between 500:1 and 900:1; Fairbridge 1972).

Mineralogy and Chemistry of Soil and Sediment Samples - Major mineral components in all soil and sediment samples are quartz and feldspar as is typical in continental rocks and sediments. At the control site (One Otter Lake) calcite and dolomite were minor components and their presence is evident in the higher pH

Table 1. Cadmium Concentrations (μg/g) in Terrestrial and Aquatic Plant Samples
Data include means, ranges and sample sizes in brackets.
Standard error (se) calculated when N>10.
m=mineralized sites; n=non-mineralized (control) sites; l=leaves;
t=twigs; r=roadside plants; se=standard error.

	Terrestrial					Aquatic	
spp.	m (l)	m (t)	n (l)	n (t)	r(l)	m (l)	n (l)
Balsam Fir	1.5 1-2 (6)	3.0 3 (2)	<1 <1 (6)	<1 <1 (2)	0.1 .1-.3 (21) se .01		
White Birch	2.8 2-3 (7)	1.0 1-2 (5)	plant not found	plant not found	0.5 .06-.9 (19) se .07		
Trem. Aspen	5.3 .7-24 (6)	4.4 .7-19 (5)	plant not found	plant not found	0.6 .08-2 (24) se .08		
Mount Ash	<1 <1 (6)	<1 <1 (5)	<1 <1 (4)	<1 <1 (4)			
Mount. Maple	0.2 .1-.2 (6)	0.3 .2-.4 (5)	0.3 .1-.5 (6)	0.3 .2-.5 (6)			
Red O Dogwd	<0.05 <0.05 (5)	<0.05 <0.05 (4)	<0.05 <0.05 (6)	<0.05 <0.05 (5)			
Bush Honey- suckle	0.8 0.6-1 (5)	1.0 0.7-1 (4)	0.3 .2-.5 (6)	0.4 .3-.5 (3)	0.2 .05-.4 (24) se.02		
Milfoil						17 16-19 (3)	plant not found
Red D Water- lily						<1 <1 (6)	<1 <1 (3)
Rich. Pond Weed						4.3 4-5 (4)	<1 <1 (2)
Cattail						<1 <1 (4)	<1 <1 (4)
Comm Bladd- erwor						<1 <1 (2)	16 16 (1)

Table 1 continued ...

	Terrestrial					Aquatic	
spp.	m (l)	m (t)	n (l)	n (t)	r(l)	m (l)	n (l)
Soil	7.7		0.6		0.2		
	0.5-11		0.5-1.2		.04-.9		
	(15)		(15)		(48)		
	se 3.9		se .05		se 0.05		
						7.1	0.5
Sedmt			0.6			1-21	.5-.7
			0.5-1.2			(10)	(10)
			(15)			se 2.0	se 2.0
			se .05				

levels in One Otter Lake (8.1) relative to Garnet Lake (pH, 6.3) at the mineralized site. Concentrations of major element oxides (Na2O (1 to 6%), MgO (1 to 5%), SiO2 (30 to 70%), Al2O3 (5 to 10%), K2O (1 to 2%), CaO (2 to 8%), Fe2O3 (1 to 7%) reflect the mineral compositions with the higher Ca, Mg and lower Si, Al levels in samples from the control site.

Atmospheric Cadmium Inputs - Cadmium levels (0.5 to 0.7 µg/g) in samples taken from moss bags deployed in the study sites differ only slightly from those in the control moss bags (<0.3 to 0.5 µg/g). Samples of Schreber's moss collected during our field work contain 0.3 µg/g Cd.

Measurements of Lake Waters - Values for pH, temperature, dissolved oxygen and conductivity were 6.3, 20°C, 8.1 µg/ml and 130 mhos respectively at Garnet Lake and 8.1, 20°C, 7.5 µg/ml and 125 mhos respectively at One Otter Lake. Cadmium levels in water samples from both lakes were below the detection limit of 0.0005 µg/ml.

DISCUSSION

Cadmium Availability to Moose in NW Ontario - The general finding from this inquiry is that Cd levels in the vegetation samples collected in this study do not exceed the range of levels (0.1 to 1 µg/g) normally found in land plants (Wedepohl 1970), except when growing in soils or sediments developed in close association with Cd-enriched rocks. Cadmium is made available to plants principally via weathering reactions between soil waters and rocks. While soils may receive atmospheric Cd inputs, there is no evidence for any significant atmospheric loading in this study.

For example, very low Cd concentrations (~0.013 µg/m^3 (24 hr); about ten times less than in southern Ontario) are found in suspended particulates (<100 µ) collected in the Thunder Bay area using high volume air sampling (MOEE, 1991). These data are corroborated by low levels of Cd found in Schreber's moss (~0.3 µg/g). Slight differences in Cd contents in the deployed mossbags (0.5 to 0.7 µg/g) over those in the control (i.e. unexposed) mossbags (<0.3 to 0.5 µg/g) likely reflect natural background Cd inputs from soil particulates in atmospheric dust. Negligible Cd accumulations in snow meltwater (<0.2 µg/l; MOEE atmospheric monitoring program) also indicate negligible Cd contributions from atmospheric dust. Another study (Rinne and Barclay-Estrup 1980) in NW Ontario designed to assess atmospheric inputs of heavy metals using samples of Schreber's moss, showed Cd levels (0.5 to 0.6 µg/g) were similar to those found in this study. Further indication of very low to negligible Cd atmospheric inputs in NW Ontario are Cd concentrations (<0.0005 μg/ml) in lake waters from the study sites as well as in NW Ontario bog waters (<0.002 μg/ml, Harris 1989). Bog waters have acidic pH levels (normally 3 to 4) compared to the lake waters at the study sites and, therefore, could sustain higher concentrations of dissolved Cd. The low soil Cd levels (means of 0.2 and 0.6 μg/g for non-mineralized sites) noted in this study suggest moose would not significantly enhance their rate of Cd ingestion from soil entrained in their forage. In this study soil and sediments contained cadmium levels typical of surface rocks, except in highly localized lake sediments and soils at the mineralized site. It appears then, that Cd taken in by moose foraging in NW Ontario terrains, similar to our study sites, is derived by natural pathways.

Accumulation Times of Cd Burdens in Moose - The time required for moose to acquire their cadmium burden may be estimated from their daily consumption of plant matter (10-20 kg for an adult; H.R. Timmermann, personal communication) and Cd levels in plant tissues they consume (0.1 μg/g, general minimum level). At this rate their daily consumption of Cd is 1 to 2 mg and the highest adult (>5 years old) Cd burdens in NW Ontario moose (26 μg/g kidney and 5 μg/g for liver; Glooschenko *et al.* 1988; Glooschenko and Burgess 1987) could be accumulated in a few months, assuming that Cd is concentrated mainly in kidney and liver tissue (total weight ~10-20 kg; H.R. Timmermann, personal communication).

Biomagnification of Cd Relative to Zn - The concept of Cd "barrier-free" plants could be useful in exploring Cd bioavailability to higher fauna. In an extensive study of Cd and Zn levels in over 100 parts of plant species, Kovalevsky (1984) distinguished between "barrier-free" plants (those that accumulated elements in direct proportion to their concentration in the nutrient medium and, hence, do not appear to have uptake limits) and "barrier" plants (those that appear to limit their uptake of an element). Some moose "preferred" species (e.g. *Betula spp.*, *Populus, spp.*) were included in Kovalevsky's list of Cd "barrier-free" plants. His results showed that more species were found to be Cd "barrier-free" than Zn "barrier-free". Thus, Cd appears more useful, for example, in geochemical exploration for rocks enriched in Cd and Zn. Kovalevsky concluded that the biogeochemical behaviour of Cd was closer to that of Pb than that of Zn.

Moose as Bioindicators of Cd Bioavailability - The results of this study raise the possibility of using moose as indicators of natural background Cd levels. Due to their herbivorous feeding habits and large sizes they are efficient samplers of vegetation. Analyzing moose tissues on a regular basis could be useful for monitoring changes to background Cd levels and may serve as a proxy for other heavy metals (e.g. Pb, Hg) considered to have significant bioaffinities. European studies show that, in this century, increasing Cd inputs to the biosphere correlated with increases in industrial cadmium production (Frank and Petersson, 1984). In one study Cd levels in wheat grown in the same place in Sweden doubled between 1916 and 1972. Another study discussed by (Frank and Petersson 1984) shows sharp rises over this century in Cd levels in renal human tissue from Sweden, Germany and Japan. These "indicator" studies provide useful global reference data. Regions such as NWO receiving background Cd levels are important in this global context to track trends in background levels of indicator elements.

ACKNOWLEDGEMENTS

The authors acknowledge the technical assistance both in the field and in the laboratory of S.A. Moogk and L.G. Zorn. Field study assistance was provided by A. Harris, G. Hazenberg and L. Malek. At Manitouwadge F. Bakker and R.G. Friesen (Noranda-GECO) suggested suitable study sites and arranged logistical support. From Noranda Research M.R. Speyer (now of Technitrol, Montreal), C.S. Wood and K.C. Wheeland assisted with gathering data. D.J. Racette (Ontario Ministry of Environment and Energy, Thunder Bay) provided mossbags and holders as well as Cd analyses. H.R. Timmermann (Ontario Ministry of Natural Resources, Thunder Bay) was an advisor throughout the project. At Lakehead University gratitude is due to T.J. Griffith and A. MacKenzie for assistance with analyses, P.Barclay for loaning equipment and S. Spivak for drafting figures. Funding for this project was provided by the Ontario Ministry of Natural Resources.

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