MERCURY IN HAIR OF LARGE ALASKAN HERBIVORES: ROUTES OF EXPOSURE

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ABSTRACT: Atmospheric-soil cycling of mercury is a process ultimately affecting mercury distribution to upper trophic levels in terrestrial ecosystems. In the Arctic and sub-Arctic, there is a paucity of information on processes and pathways related to mercury accumulation in terrestrial environments. We reviewed data on mercury levels in resident species of caribou (*Rangifer tarandus*) and muskox (*Ovibos moschatus*). Comparison of the inhaled and ingested contribution of mercury from the environment in those species, as well as concentration factors, show accumulation of total mercury (THg) at higher trophic levels in the terrestrial food webs in Alaska, USA. Higher THg concentrations in free-ranging caribou support the hypothesis that caribou, with a major component of lichen in their diet, have higher levels of Hg. Captive muskox showed little biomagnification with a concentration ratio close to 1, whereas caribou had a concentration ratio of 12.

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Caribou and reindeer (*Rangifer tarandus*) as well as muskox (*Ovibos moschatus*) generally are associated with the tundra biome (White et al. 1987, 1995; Bunnell et al. 1981). In the wild, these ungulate species are all known to eat low-growing plant species, such as lichens, grasses, and herbaceous vegetation. Sedges and cotton grasses (*Eriophorum* spp.) are a major component in the diet of caribou during summer (Ewing 1996). Lichens (*Cetraria* spp.) play a more important role in food habits of caribou during winter (Parker 1978). Lichens are slow-growing plants and accumulate contaminants in a nonselective manner (Thomas et al. 1992). Information on levels of contaminants such as mercury (Hg) in terrestrial species of wildlife in Arctic and sub-Arctic regions was limited prior to 1991, and data on temporal trends was almost nonexistent (Thomas et al. 1992). Mercury is a heavy metal that is readily absorbed through respiratory and gastrointestinal tracts. Mercury can accumulate over time in body tissues and deposition of Hg in hair is a possible means of loss of this heavy metal (Neuman 1998). Studies of Hg accumulation in Arctic food chains have been performed mostly on aquatic and marine biota, whereas terrestrial food webs have been studied less well (Braune et al. 1999, Gnamus et al. 2000). Nonetheless, past estimates of atmospheric transport of Hg indicate that total mercury (THg) is deposited on terrestrial environments (Fitzgerald and Mason 1996). The transport of Hg from plant and seed biomass to large terrestrial herbivores is low compared with conversions in predators (Lodenius 1994); however, higher rates of consumption of grasses and lichens in Arctic herbivores may intensify Hg uptake.

Herein we examine Hg accumulation in Arctic herbivores by calculating the contribution by inhalation and ingestion in the intake of Hg. Ambient air concentrations of Hg were measured as starting points of mercury accumulation in the terrestrial food web (Gnamus et al. 2000), with our goal being to establish concentration factors at the trophic level of caribou and muskox in both captive and free-ranging populations.

METHODS

Sampling of Air

Fifteen samples were collected by pumping air at a constant rate of 2.10 L/min through a gold-coated quartz tube for 4 hrs with an air-sampling pump (Mine Safety Appliances; Escort Elf Pump, Pittsburgh, Pennsylvania, USA). A portable electronic thermometer-barometer (Oregon Scientific, Portland, Oregon, USA) recorded initial and final temperatures and the barometric pressures. Trapped Hg^0 was analyzed with a Tekran detector. Descriptions of the Tekran and its operating parameters have been reported by Lindberg et al. (2000); the device collects airborne Hg from Teflon-filtered air by gold amalgamation, then trapped Hg is released by thermal desorption for quantification by cold vapor atomic fluorescence spectroscopy (CVAFS).

Study Site

Captive caribou, reindeer, and muskox used in this study were bred and raised at the Large Animal Research Station (LARS) at the University of Alaska Fairbanks (UAF; 65° N, 146°W; White et al. 1995). Sixteen muskoxen from Nunivak Island were taken to LARS in April 1980 to establish that herd (White et al. 1995). Colonies of reindeer and caribou were established within 3 years. Muskox at LARS are fed brome hay, which is supplemented by the UAF Textured Ration (Barboza and Blake 2001). This textured ration is composed of pellets of corn and barley (Alaska Pet and Garden, Anchorage, Alaska, USA). All herbivores at LARS graze on seasonally available vegetation. Reindeer and caribou forages were supplemented with barley, corn, minerals, and vitamins (Textured Reindeer Ration; Alaska Pet and Garden, Anchorage, Alaska, USA). The supplement contained 88% dry matter, which was composed of 18% neutral fiber, 7% acid fiber, 19% crude protein, 3% crude fat, and 8% ash (Barboza and Blake 2001). Hair from the large herbivores was collected during summer 1998 at LARS, placed in plastic bags and stored cold until analysis. Samples from free-ranging caribou were collected during 1997-1998 in the Teskekpuk Lake area of northwestern Alaska, USA.

Sample THg and Statistical Analysis

Seven milliliters of a 7:3 volume-to-volume mixture of concentrated nitric and sulfuric acid was added to vials and allowed to dissolve approximately 0.5 g of hair. Samples were placed on a 90°C hotplate for 4 hrs to dissolve organic matter. Samples were diluted to 37 ml with a 10% solution of 0.2 N BrCl. Aliquots of digested samples then were placed in the bubbler along with reverse osmosis (RO) water (100 ml) and 0.3 ml SnCl. The gold trap was then placed onto the bubbler and allowed to collect for 20 min. Mercury was released from the amalgam by heating, and total levels of mercury were determined with a Tekran detector by cold vapor atomic fluorescence (CVAFS). Samples of feed were collected in plastic zip-lock bags and analyzed by the same methodology at Frontier Geosciences (Seattle, Washington, USA). When sample size permitted, *t*-tests were used for comparison (Zar 1984).

RESULTS

Atmospheric mercury, mostly Hg^0 by our methodology, was measured during late summer and early autumn 1999 in interior Alaska, USA (Healy, Fairbanks, and Yukon River). Table 1 compares measurements 100 km north of Fairbanks at the Yukon River to 100 km south of Fairbanks at Healy. Those results showed levels slightly > 2 ng/m^3 for widely separate sites in interior Alaska. Mercury concentrations in air fluctuated < 1 ng/m^3 and temperature changes from 8° to -4°C did not substantially affect mercury concentrations.

Mercury in Alaskan Herbivores

Hair from caribou and reindeer differed between captive and free-ranging animals (Table 2). Mercury levels in the hair of caribou and muskox at LARS varied between 2.4 and 11.7 ng/g. The free-ranging caribou from the Teskekpuk Lake region of northwest Alaska showed not only higher mean levels, but also a wider range of values for THg (7.5 - 85.1 ng/g). The lower levels in the captive herbivores at LARS indicated a need for analysis of the feed used at the research station. Table 3 indicates very low levels of THg in feed with the highest level in imported brome hay. Hg concentrations of feed in caribou at LARS were used to calculate minimum or baseline intakes from air inhalation and ingestion. Amount of mercury inhaled by caribou at LARS was 500 fold less than that ingested (Table 4).

DISCUSSION

Information available on mercury in the Arctic and sub-Arctic has increased over the last few years, especially in aquatic and marine biota (Hansen et al. 1998). Nevertheless, the major focus for terrestrial mammals has been on other metals (Table 5), mostly cadmium (Glooshenko et al. 1988, Braune et al. 1999, Crichton and Paquet 2000, Gustafson et al. 2000). Mercury levels in tissues of caribou, such as kidney and liver, are generally low in Northwest Territories and Yukon, Canada (Braune et al. 1999). Larter and Nagy (2000) recently reported Hg levels in kidneys from caribou in the Northwest Territories with means of 1.45 µg/g for Banks Island and a 2.41 µg/g (wet weight) for the Bluenose caribou herd. Elkin and Bethke (1995) reported Hg levels for other caribou herds in the Northwest Territories with values that ranged between 0.52 and 2.93 µg/g. The Hg value from our hair samples are below that range, similar to the lower levels reported in Finnish reindeer by Lodenius (1994; 0.09 µg/g in liver and 0.01 µg/g in muscle). On Isle Royale, Michigan, USA, mercury in molars of moose (*Alces alces*) increased from 135 to 254 ng/g between 1966 and 1970 (Eide and Peterson 1997).

We observed even lower levels of mer-

Table 1. Mean atmospheric mercury concentration in air at 3 surveyed locations in Alaska, USA.

Location	Date	*n*	Hg Concentration (ng/m^3) $\bar{x}$ (Range)
Fairbanks	August 1998	5	2.1 (1.5-2.7)
	October 1998	4	2.4 (1.9-2.9)
	September 1998	3	2.3 (2.0-2.4)
Yukon river	August 1998	2	2.3 (2.0-2.6)
Healy	August 1998	1	2.0
Mean ± SD		15	2.3 ± 0.4

Table 2. Mean total mercury concentrations (ng/g) in large herbivores, Alaska, USA.

Species	n	$\bar{x}$ (Range)
Caribou		
LARS (1998)[1]	2	3.6 (2.5-4.6)
Teskekpuk (1997)	11	47.1 (7.5-66.2)
Teskekpuk (1998)	11	62.7 (44.2-85.1)
Reindeer (LARS 1998)	4	4.1 (2.4-6.1)
Muskox (LARS 1998)	5	8.0 (6.1-11.7)

[1]LARS caribou are tame. For the wild caribou at Teskekpuk, the difference in the yearly means was not significant ($t = 0.21, P > 0.05$).

cury in the hair of caribou and muskox at LARS. Hg values in LARS feed, which range between 0.3 ng/g and 11.5 ng/g, were lower than the values reported for plants from temperate regions (Gnamus et al. 2000), as well as values reported by Bailey and Gray (1997) for Alaska. In the Arctic, Ford et al. (1995) noted mean values of mercury for various lichens, mosses, and blueberries that range from about 2 to 50 ng/g. The Hg levels at LARS, when compared with data for the Teskekpuk herd, are consistent with the hypothesis of Larter and Nagy (2000) that caribou with a higher component of dietary lichen have higher contaminant levels of Hg.

Table 3. Total mercury in samples of feed, hay, and wild rice used at Large Animal Research Station (LARS), Fairbanks, Alaska, USA.

Sample	THg (ng/g)
"D" Ration[1]	1.9
"M" Ration	2.4
Textured Ration	0.3
Brome Hay	11.5
Wild Rice	2.1

[1] "D" Ration contained cereal grain, alfalfa, and molasses, while textured ration contained minerals, vitamins, and cereal grains with molasses (Alaska Pet and Garden, Anchorage, Alaska, USA; Barboza and Blake 2001).

Differences in regional geology, atmospheric deposition, and biogenic transport (dietary intake) have been cited to explain contaminant loading in the Arctic and sub-Arctic (Braune et al. 1999). For caribou, which are strict herbivores (Larter and Nagy 2000), amount of lichen in the diet may explain variation we observed between captive caribou at LARS and free-ranging caribou. Lichens are long–lived and readily accumulate atmospheric contaminants in a nonselective manner (Froslie et al. 1984, Crête et al. 1992, Thomas et al. 1992). Caribou that feed substantially on lichen would accumulate higher levels of mercury. The difference we observed between caribou at LARS and those at Teskekpuk can be explained by the higher component of dietary lichens in free-ranging caribou.

The relationship of Hg level with lichens in the diet has been suggested earlier. Froslie et al. (1984) reported reindeer to have higher mercury burdens than moose, roe deer (*Capreolus capreolus*), and red deer (*Cervus elaphus*) in Norway. In that study, reindeer foraged mainly on lichens, whereas moose, roe deer, and red deer foraged on grasses or herbaceous plants. Larter and Nagy (2000) presented extensive data showing higher Hg concentrations in Bluenose barren ground caribou that have a high component of lichens in winter diets; approximately 85%. Peary caribou on Banks Island with only a 1% lichen component in diets had low levels of Hg. The controlled diet of caribou at LARS will allow us to experimentally explore this relation in Arctic herbivores in the future.

Major routes of mercury exposure in terrestrial herbivores are inhalation and ingestion, which can be calculated (US EPA 1989, Gnamus et al. 2000). The estimate of intake of mercury by either ingestion or inhalation would give the relative magnitude of the importance of those 2 different routes

Table 4. Calculated minimum amount of mercury inhaled and ingested by caribou.

INHALATION

Inhalation (ng/kg/day) = CA x IR x ET x EF x ED/[BW x AT] = 3.2 ng/kg/day

Symbol	Description	Values
CA	Contaminant concentration in air (ng/m^3)	ca. 2.2 (Table 1)
IR	Inhalation rate (m^3/h)	0.6±.1 m^3/h
ET	Exposure duration (h/day)	24 h/day
EF	Exposure frequency (day/year)	365 day/year
ED	Exposure duration (years)	10 years
BW	Body weight (kg)	100kg
AT	Average exposure duration in the year (days)	365 days

INGESTION

Ingestion (ng/kg/day) = CC x IR x FI x EF x ED/[BW x AT] = ca 1,800 ng/kg/day

Symbol	Description	Values
CC	Contaminant concentration in food (ng/kg)	ca. 2.4 (Table 3)
IR	Feed ingestion rate (kg/meal)	5 kg/meal average dry foodstuffs consumed in intervals throughout the day.
FI	Fraction of contaminated food ingested (no units)	1
EF	Exposure frequency (meals/year)	365 meals/year (each meal lasts a day with intervals)

for mercury exposure to Alaskan herbivores. Using the equations of Gnamus et al. (2000), assuming metabolic similarities with the roe deer from Slovenia, we calculated inhalation and ingestion exposure for caribou. The rate of Hg inhalation of 3.2 ng/kg/day (Table 4) agrees well with the value for roe deer of 7 ng/kg/day. Ingestion exposure from mercury by LARS caribou was 1.8 µg/kg/day (Table 4). That result also supports the importance of lichen as a pathway of Hg to caribou and reindeer. The ingestion intake was 500 fold higher than exposure from inhalation.

Based on values for textured ration in Table 3 and mercury levels in caribou (Table 2), the concentration factor (CF) for mercury in LARS caribou was 12. The CF value for LARS muskox was 0.7, which is similar to roe deer (Gnamus et al. 2000). The proportion of Hg in inhalation intake for caribou is about 0.2% of the mercury intake through consumption of the grasses and feed. Similarly, river otters (*Lontra canadensis*) inhabiting coastal ecosystems (Ben-David et al. 2001) showed that the intake of mercury through ingestion of food is far more important than inhalation.

Despite small sample size, this study provides data to support the commonly accepted idea that mercury accumulation in Alaskan terrestrial herbivores is not very large. Nonetheless, differences between captive and wild populations suggest that it is essential to monitor the hair of large terrestrial herbivores as local and global conditions change.

Table 5. Studies of metal levels in large herbivores inhabiting Arctic and sub-Arctic environments.

Metal	Symbol	Animal	Reference
Cadminum	Cd	Moose	Froslie et al. (1984) Scanlon et al. (1988) Glooschenko et al. (1988) Crichton and Paquet (2000) Gustafson et al. (2000)
		Caribou	Braune et al. (1999) Elkin and Bethke (1995) Crête et al. (1992) Larter and Nagy (2000) Gamberg and Scheuhammer (1994)
		White-tailed Deer	Glooschenko et al. (1988) Brazil and Ferguson (1989)
Copper	Cu	Moose	Frank (1986) Flynn et al. (1977) Galgan and Petersson (1994)
		Caribou	Barboza and Blake (2001)
		Muskox	Barboza and Blake (2001) Blakeley et al. (2000)
Lead	Pb	Caribou	Crête et al. (1992) Elkin and Bethke (1995) Larter and Nagy (2000) Macdonald et al. (1996)
Mercury	Hg	Caribou	Larter and Nagy (2000) Crête et al. (1989) Froslie et al. (1986) Eriksson et al. (1990) Lodenius (1994) Elkin and Bethke (1995)
		Muskox	Braune et al. (1999)
Nickel	Ni	Caribou	Larter and Nagy (2000)

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