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Mercury in lichens and reindeer hair from Alaska: 2005–2007 pilot survey

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ABSTRACT. Reindeer and caribou are terrestrial herbivores, that feed on lichens and are used for commercial and subsistence food products. Caribou are a key component of the arctic food web and the bioaccumulation of toxic contaminants, such as mercury (Hg), needs to be monitored to establish a baseline as the arctic environment is impacted by both climate change and future industrial development. A changing climate in Alaska is influencing plant species composition, fire regime, melting and flooding events, and thus, impacting Hg bioavailability in the food chain. Industrial development in Asia is also projected to increase the atmospheric global pool of Hg from increased coal combustion. Reindeer, a domesticated representative of caribou, can be used as a terrestrial biomonitor for metal exposure. In this study total mercury concentrations were measured in lichens and in hair of grazing reindeer on defined ranges across Alaska to establish a baseline for future hypothesis development and testing regarding Hg deposition. The Hg mean level for Seward Peninsula lichens on the Davis Range was 37.4 ng g^{-1}, on the Gray Range 47.1 ng g^{-1}, on the Kakaruk Range 42.2 ng g^{-1}, and 41.7 ng g^{-1} on the Noyakuk Range. Lichen Hg levels on St. Lawrence Island was 46.6 ng g^{-1}. Methyl mercury levels in lichens were found to be below detection levels. Reindeer grazing on these ranges had mean Hg hair levels of 14.6 ng g^{-1} (Davis herd), 83.4 ng g^{-1} (Gray herd), and 40.3 ng g^{-1} (Noyakuk herd). Two reindeer on St. Lawrence Island had an average of Hg of 43.0 ng g^{-1}. Sample sizes ranged from $n = 2$ to $n = 11$. Hg mean levels in lichen on Seward Peninsula were higher than Hg means of two ranges in northern Mongolia. The Hg levels observed in this study indicate that Hg levels in Alaska are low at this time and pose no risk to the health of reindeer or human subsistence harvesters. A significant relationship between Hg in lichens on the ranges and the Hg in reindeer on those ranges has not been established. There are insufficient data on Hg levels in many areas of the north and more information is needed on location specific and time trends in Hg concentrations. Lichens and reindeer hair provide a good, non-invasive method of monitoring metal exposure changes in Alaskan ecosystems.

Contents

Introduction

Mercury (Hg) is a globally transported contaminant that can bioaccumulate in the food chain (AMAP 2003; Duffy and others 2005b; Dehn and others 2006; Sun and others 2006; Dunlap and others 2007). Globally, Hg is present in elemental, ionic and organic forms with a global distribution via long range transport (Fitzgerald and others 1998; Lindberg and others 2007). Hg is released by normal weathering processes, volcanic emissions and anthropogenic activities such as mining, coal combustion and waste incineration (Hylander and Meili 2003; Swain and others 2007). The transport of air masses from middle to high latitudes leads to the deposition of Hg in polar regions (Sun and others 2006); cooler climates also favour deposition (Martinez-Cortizas and others 1999). Since Hg can bioaccumulate in lichens (Conti 2008), atmospheric deposition can be monitored as Hg is passed up to higher trophic levels (Bhanera and Costa 1992; Scheuhammer and others 2007).

Currently in Alaska, especially on the Seward Peninsula, Hg exposure in the ecosystem is not well characterised but exists sporadically from gold mines, which once operated there. Hg exposure is expected to be minor since the peninsula is not part of Alaska's 'Hg ore belt', nor at this time is coal a major source of heat or power in the area. However, future natural resource development may lead to an increase in the local input of Hg so monitoring global and local inputs would be important to the community.

The biomagnification of Hg from plant and seed biomass to larger terrestrial herbivores is relatively low when compared with conversions in marine systems which have more trophic levels (Gamberg and others 2005), although high consumption rates of grasses and lichens among Arctic herbivores may intensify Hg uptake (Froslie and others 1984). Lichens are a symbiotic association of algae (phycobiont) and fungi (mycobiont), and are long lived, growing on rock, wood, or soil substrates. They extract water and labile nutrients from the air and are, therefore, sensitive to air pollution (Loppi and Bonini 2000; Riget and others 2000; Conti 2008). Among the many mammal and bird species having potential uses for biomonitoring, *Rangifer spp*. (caribou and reindeer) are particularly important because they continue to serve as a food staple across the circumpolar north. Reindeer are associated with both taiga and tundra biomes in which the major components of the summer diet are low growing species such as sedges, willows and lichens, while lichens are the main component in the winter diet (Ewing 1996; Aastrup and others 2000; Robillard and others 2001; Finstad and Kielland 2005; Inga 2007). These diet regimes can be impacted by climate change factors, such as temperature and precipitation by reducing the growth of lichens, which are replaced by sedges through competitive interactions. Arctic environs may be more impacted than temperate regions by melting of permafrost and by flooding (Finstad and Kielland 2005; Moen 2008). Warmer and wetter weather also increases the probabilities of ice crust formations, which decrease forage availability (Moen 2008).

In terrestrial settings, the concentration of mercury in reindeer hair offers a means of measuring variation in Hg bioaccumulation over time (Duffy and others 2001, 2005b). In selected samples, the methyl mercury (MeHg) levels in reindeer hair were approximately 79% of THg (range 58%–97%), (Duffy and others 2005a). In this pilot survey, our goals were 1) to compare Hg concentration in the hair of reindeer from four herds from western Alaska, 2) to compare Hg levels in the lichens from northwest Alaska and Mongolia and 3) to determine whether there was a relationship between Hg in reindeer herds and the Hg levels in lichens growing on reindeer ranges. Here, we report levels of THg in reindeer hair samples and lichens from the Seward Peninsula, St. Lawrence Island and Fairbanks in Alaska (Fig. 1), and from lichens collected from two reference ranges in Mongolia. These data will be an important baseline in testing the hypothesis that greater Hg deposition will accompany increased temperatures and rainfall runoff related to climate change (Munthe and others 2007; SNAP 2008) as well as any future industrial development on the Seward Peninsula.

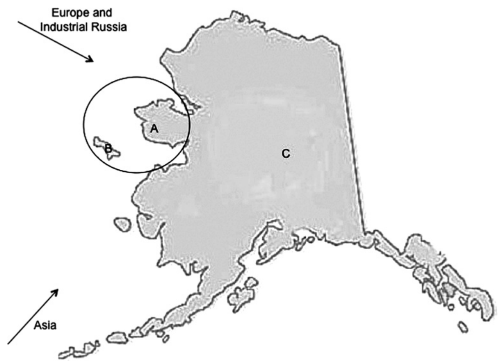

Fig. 1. Map of Alaska showing Seward Peninsula (A), St. Lawrence Island (B) and Murphy Dome (C) in Alaska. Wind patterns are indicated with arrows.

Material and methods

We collected full length winter coat guard hairs from the nape of the neck of domestic reindeer on Seward Peninsula and St. Lawrence Island ranges at the time of winter harvest in January and February. Animals were herded to the slaughter area. Sample sizes (*n*) were 10 where *n* represents an individual reindeer (or lichen plant), except for in the case of St Lawrence Island. The samples were collected in different years (see Table 1) from adult reindeer at harvest in January and February. Age and size of individual adult animals does not have a significant effect on Hg levels in hair (Duffy and others 2005a). Reindeer were adult castrated steers, 3 to 4 years old, with body weights of approximately 300 pounds (136 kg). New hair growth begins in the spring and continues to early winter. The Davis and Kakaruk ranges were coastal while the Noyakuk range was inland at a slightly higher elevation. The Gray range is also coastal, located on a peninsula projecting into Norton Sound. The general latitude and longitude for the herds are 65°N, 165°W with a range of 5,000 km^2 per herd (Fig 2). GPS locations of the sites sampled on the lichen transect of the range of each herd range are available by contacting the first author. These ranges are characterised by many isolated valleys in which reindeer feed high on the ridges during winter (Finstad 2008). Lichens (*Cladina* sp.) were sampled in July along a series of range transects on the Seward Peninsula and opportunistically from St. Lawrence Island. The percentages of ground cover of forage on the herd ranges were approximately 20% willow, 30% sedges and 40–50% lichens (Finstad 2008). Since we used an opportunistic sampling design over 2 years, individual characterisation of the Hg concentration relationship between lichens and sampled reindeer was not possible.

Northern Mongolia lichen samples were collected from the Tsateau community of reindeer herders, who occupy a geographically isolated area in the taiga and alpine tundra of Havsgol Province. The east taiga camp (ET1) 50°46’N, 99°22’E, is at a lower elevation than the west taiga camp (WT) 51°22’N 98°48’E. Since Asia including Mongolia is the direction from which the atmospheric circulation approaches Alaska (Fig. 1), it was interesting and potentially relevant to use this area as a reference region.

Hair samples or lichen samples where collected and placed in a plastic bag and transported by air to the laboratory. Hair samples were washed in dilute detergent and dried before analysis at Frontier Geosciences (Seattle, WA). Hair samples and lichens were digested whole without subsampling. Data are reported on a wet weight basis (ww) in ng/g.

Lichen THg analysis was conducted by Frontier Geosciences using a standard curve, spanning the entire analytical range of interest. Standard curves were

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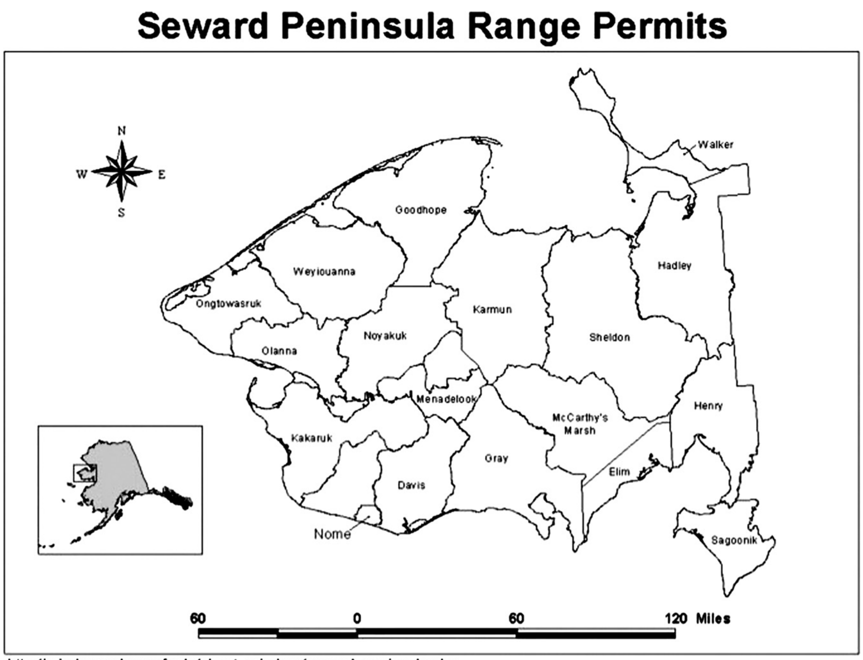
Table 1. Total Mercury (ng g^{-1}) in lichens and reindeer hair.

Year	Location	n*	THg ±SD	Range
A. Lichens				
2006	Davis	11	37.4 ± 21.2	23.1–99.5
2007	Gray	10	47.1 ± 19.8	26.1–87.8
2006	Kakaruk	8	42.2 ± 18.0	22.2–75.3
2007	Noyakuk	10	41.7 ± 21.9	22.1–96.0
2006	St. Lawrence	3	46.6 ± 15.0	34.7–63.4
2006	ETI-Mongolia	5	16.8 ± 4.5	12.1–22.5
2006	WT-Mongolia	5	25.6 ± 4.4	19.6–30.8
2007	Murphy Dome, FAI	6	32.4 ± 15.6	15.1–59.0
Year	Location	n**	THg ±SD	Range
B. Reindeer hair				
2006	Davis	10	14.6 ± 12.8	5.4–50.1
2005	Gray	10	83.4 ± 9.8	67.5–103
2005	Noyakuk	10	40.3 ± 6.3	29.8–49.8
2006	St. Lawrence	2	43.0 ± 5.3	39.3–46.8

n* is the number of individual plants analysed.
n** is the number of individual animals analysed.
Transects along which samples were collected:
Kakaruk (65°03' N, 166°07' W to 65°05' N, 165°51' W);
Davis (64°33' N, 165°51' W to 64°42' N, 165°17' W);
Gray (64°34' N, 164°44' W to 64°31' N, 164°21' W);
Noyakuk (65°34' N, 165°09' W to 65°09' N, 165°51' W).

calculated with blank corrected initial standards. For each analytical set of 20 samples, one matrix duplicate, two matrix spikes and several method blanks were co-processed and analysed in exactly the same manner as the samples. The blank THg mean was 0.20 ±.01 ng g^{-1}. The recovery of the DOLT −3 standard reference material averaged above 95% and the spike recovery averaged within 10% of the expected value. Variation of duplicate samples were also within the 10% range.

THg concentrations in the samples were measured at Frontier Geosciences using the cold vapour atomic fluorescence spectrometry (CVAFS) method. Results were reported on a wet weight (w/w) basis as ng g^{-1} (ppb). For total mercury in hair, approximately 0.25 g of



http://reindeer.salrm.uaf.edu/about_reindeer/seward_peninsula.php

Fig. 2. Map of Seward Peninsula with location of Reindeer herd ranges: Davis, Gray, Kakaruk, Noyakuk.

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each reindeer hair sample was digested with 10 mL of hot refluxing 70% HNO_3 : 30% H_2SO_4 for approximately 2 hours. The digests were then diluted to a final volume of 40 mL with a solution of 10% (v/v) 0.2N BrCl. Aliquots of each digest were reduced in pre-purged double-distilled water to Hg°, purged onto gold traps as a preconcentration step. The Hg contained on the gold traps was then analysed by thermal desorption into a cold vapor atomic fluorescence detector (CVAFS) with the dual amalgamation technique.

Methylmercury was also measured from select lichen samples for this study using the Frontier Geosciences standard method similar to the reported procedure for MeHg in hair described by Duffy and others (2005a). Ten samples from the Gray and Noyakuk ranges were below detectable levels (1ng/g MeHg). Since the lichens of these ranges were below detectable levels, only THg was measured on the other ranges. The limited literature on MeHg in lichens in non-polluted areas suggests that its concentration is very low.

Analysis of variance was used to analyse the data to evaluate the effects of sampling location on THg concentrations independently in hair and lichen from the different ranges. Data were analysed using SAS statistical software. Each site in Table 1 was treated as an individual site. Significant THg differences in hair and lichen samples from different ranges were tested using Tukey's Studentized Range test. Differences were considered significant at $p \leq 0.05$. A regression analysis was performed to evaluate a correlation between THg concentrations in hair and THg concentration in lichen on the ranges.

Results and discussion

THg in reindeer hair from the three Seward Peninsula herds ranged between 5–103 ng g^{-1} ww (Table 1). Means for the reindeer varied among the herds, with the Davis herd having the lowest mean (14.6 ng g^{-1}). The Gray herd had the highest mean THg (83.4 ng g^{-1}). The Noyakuk herd had a mean THg of 40.3 ng g^{-1} and the two reindeer from St. Lawrence Island off the coast of western Alaska had a mean THg average of 43.0 ng g^{-1}. The THg reindeer hair means from the Gray and Davis ranges were significantly different from each other and from the Noyakuk and St. Lawrence ($P<0.0001$, F value = 121). The Tukey's Studentized Range also showed significant differences between Davis, Gray and Noyakuk reindeer.

THg in lichens varied between herd ranges with the Davis Range being the lowest: Davis (37.4 ng g^{-1}); Gray (47.1 ng/g); Kakaruk (42.2 ng g^{-1}); Noyakuk (41.7 ng/g). Lichens from St. Lawrence Island had a mean THg level of 46.6 ng g^{-1}. As a non-Alaska site using an opportunistic sampling design, lichen samples from Mongolia showed the lowest levels of THg of any range (16.8 ng g^{-1}). Reindeer lichens from the interior of Alaska at Murphy Dome in Fairbanks showed a mean THg (32.4 ng g^{-1}) similar to lichens found on the Seward Peninsula. Ten

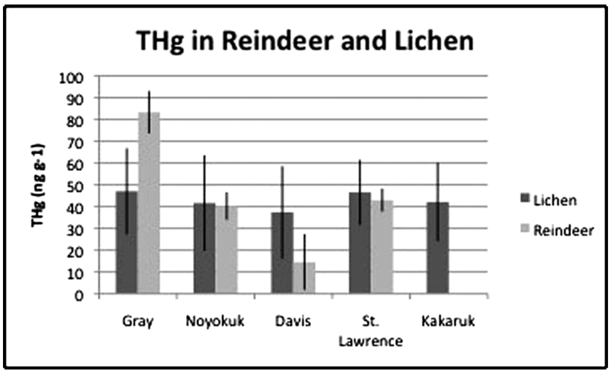

Fig. 3. Total Hg levels (ng g^{-1}) in reindeer and lichen on the Seward Peninsula.

lichen samples were analysed for methyl mercury (MeHg) and in all samples, MeHg was below detectable levels (≤ 1 ng g^{-1}). We found no significant relationship between the reindeer hair and lichen on these ranges. This may be due both to the different years and seasons when the reindeer and lichens were sampled. Small sample size is also a concern. As seen in Fig 3, the lichen THg levels on the ranges was similar to reindeer hair level with the exception of the Davis herd. The Davis herd hair THg was low in this survey, but the low number of samples or a year effect may have biased the mean. The Noyakuk and Gray herd values were similar to a previous screening (Duffy and others 2005b).

Environmental monitoring and health impact assessment of Hg exposure has increased worldwide over the last 20 years (Ebi and McGregor 2008). Coastal, highly populated communities have drawn the most attention, but rural and northern terrestrial systems are also important (Fitzgerald and others 1998) for monitoring changes in mercury distribution related to both local disturbance brought on by industrial activity (Swain and others 2007), or more general indirect effects such as global climate change (Wong and others 2006). For long term environmental monitoring of a region, two characteristics of a good indicator species would be wide distribution and accumulation of a contaminant (Burger and Gochfeld 2001). Lichens and reindeer have these characteristics and are good indicator species for terrestrial environments in many areas of Alaska (Horvat and others 2000; Ikingura and Akagi 2002; Robillard and others 2002; Burger and others 2001; Duffy and others 2005b) and across the circumpolar north (Gamberg and others 2006; Lindberg and others 2007). Lichens and reindeer have commonly been used in the past as biomonitors for the accumulation of environmental contaminants such as radioactive isotopes (O'Hara and others 1999) and metals (Robillard and others 2002; Riget and others 2002; Horvat and others 2000). Determining baseline levels for Hg in indicator species is important for communities on the Seward Peninsula since this region is projected to become warmer over the next 75 years. Warmer temperatures and a longer growing season are expected to increase evapotrasportation sufficient to counteract

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the projected regional increase in precipitation (SNAP 2008). Severe summer rains and runoff, despite drier annual climate predictions, may increase Hg mobilisation (Munthe and others 2007). Increased transport from Asia is also predicted (Wong 2006), so the Mongolian reference areas, which are impacted differently by climate change, will be important areas for comparison.

There are several factors other than lichens, that can contribute to THg in reindeer hair. These include THg in other browse (sedges, willows, grass) during the summer period of hair growth, and the methylation activity of rumen flora. Lichens play an important role in the diet of Alaskan reindeer during winter and early spring (Finstad and Kielland 2005). Reindeer that feed substantially on lichens accumulate mercury in tissues such as hair, muscle and liver in both inorganic and MeHg forms (Aastrup and others 2000; Ikingura and Akagi 2002). The relationship of Hg levels in reindeer with lichen biomass in the diet was suggested earlier by Froslie and others (1984) who reported that reindeer have higher mercury burdens than moose (*Alces alces*), roe deer *(Capreolus capreolus*) and red deer (*Cervus elaphus*) from Norway. In Froslie's study, reindeer foraged mainly on lichens, whereas moose, roe deer and red deer foraged on shrubs, grasses, or herbaceous plants. A study by Aastrup and others (2002) also suggested that winter forage rich in lichens explained increases in Hg levels. Lichen levels in summer diets of Seward Peninsula reindeer herds varied between 30 to 50% (Finstad and Kielland 2005), indicating that summer lichen consumption may differ from winter consumption across the ranges and between herds. While the data from this survey suggests that the THg in reindeer hair is similar to the THg in lichens on their ranges, we did not find a significant correlation between range lichen THg levels and the reindeer hair THg content of those that were harvested from the same (Fig. 3) range. Also the very low levels of THg in the hair of the Davis herd compared to the lichens needs to be repeated with a greater number of samples.

Our results suggest that further study is needed on the timing of lichen ingestion by northern ruminants. Our lichen samples were collected in the summer (July) while the hair samples were collected at harvest in January and February. Since hair growth occurs after shedding during the summer (April to September), the THg levels in summer sedges, willows and cotton grass may have a greater influence on the THg levels in the winter harvest. Also, changes in the composition and activity of intestinal flora could affect both the absorption and efficiency of the conversion of Hg to methyl mercury (Duffy and others 2005a). However, there is no laboratory data, at this time showing that anaerobic rumen flora can methylate Hg.

Several opportunistic samples of lichens from Mongolia, an area northeast of the sources of Chinese atmospheric Hg releases, showed that levels of THg in the Mongolian lichens were lower than levels detected from Alaska lichen samples. These lower levels may reflect Alaska being favoured by the global distillation mechanism of transport and deposition, or a difference in the natural background of mercury in the soil. These Mongolian ranges can serve as a reference area for future climate change trends in Alaska, with the expectation that Mongolian ranges will not be impacted in an identical way to western Alaska.

Reindeer are an important subsistence resource to northern people. At least twice in the last thirty years, various Alaskan herds declined quickly, but, through modern management techniques, both herds rebounded. Loss of habitat related to climate change can pose a risk to reindeer herd size, and increased metal deposition can impact their exposure by increases in lichen and other forage. Local mining development and road building can also impact metal exposure in reindeer ranges in Alaska. Increased local development, and increased deposition from the global Hg pool are potential sources of Hg (Wong and others 2005). Climate change, driven by release of 'greenhouse gases,' could make Hg more bioavailable through forest fires (Garcia and Carignan 2000; Turetsky and others 2006), flooding (Balogh and others 2006) and possibly permafrost melting. It is therefore important to develop both a baseline and a monitoring programme for metal exposure to the lichens on the different ranges utilised by Alaska herbivores (AMAP 2003; Burger and others 2005). Currently, there is only limited observational and retrospective data related to mercury accumulation in either plants or reindeer in the western Alaskan Arctic and Bering Sea islands (Gerlach and others 2006).

The Hg risk from the terrestrial food chain to Alaskan communities is low (Verbrugge 2007). Since the Alaskan subsistence diet in the Norton Sound region is a mixture of fish, marine mammal and caribou/reindeer (Verbrugge 2007; Dunlap and others 2007), there is no specific biomagnifications factor relating reindeer meat and human hair levels. Human women hair samples had a median level 530 ng g^{-1} with a maximum of 7,820 ng g^{-1} (Verbrugge 2007). Biomagnification, in this case, is defined as the process by which consumers accumulate higher contaminant than food sources at lower trophic levels, in contrast to bioaccumulation, which is the tendency for certain compounds to build up in a biological tissue. If only reindeer were consumed, the biomagnification factor from reindeer to man would be estimated around 10 based on the reindeer hair data of this study. However, since the Hg levels in reindeer are similar to the Hg levels in the lichen, reindeer do not significantly contribute to the biomagnification of Hg on the Seward Peninsula and the biomagnification factor would probably be closer to one.

Hg analysis of lichen and reindeer hair provides a good, non-invasive monitoring method in Alaska. While significant differences exist between lichen Hg levels and their ranges, a larger sample size and concurrent reindeer-lichen sampling is needed. Based on our limited data, Hg biomagnifications from the terrestrial food chain is not a significant factor in Hg exposure to communities in the Seward Peninsula of Alaska at this time.

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