



Contaminants in two West Greenland caribou populations



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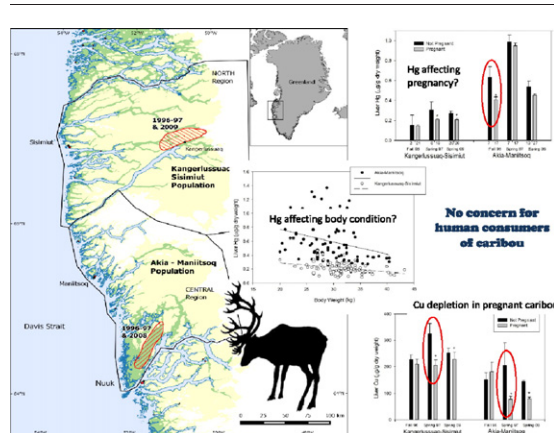
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HIGHLIGHTS

- Caribou tissue contaminant profiles may reflect different diets.
- Low hepatic copper may result in copper depletion in pregnant caribou.
- High hepatic mercury may negatively affect fertility in caribou cows.
- Hepatic mercury is negatively correlated with body condition in caribou cows.
- Metal levels in tissues are not a health concern to people consuming caribou.

GRAPHICAL ABSTRACT



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ABSTRACT

Two caribou populations in West Greenland were sampled and the kidneys, liver and muscle analyzed for contaminants, including aluminum, arsenic, cadmium, copper, lead, mercury, selenium and zinc. Although close in proximity, the two populations are topographically separated by an ice cap, which creates different climates and vegetation types in each region. Contaminant levels reflected the differing diets of the two caribou populations. To the south in the wetter lichen-rich region, caribou had significantly more aluminum, arsenic, cadmium, lead, mercury, selenium and zinc, likely due to atmospheric deposition on lichens. To the north in the dry desert steppe where grasses predominate, caribou had higher levels of copper. Cows collected in late winter had significantly less hepatic copper, lead and mercury if pregnant, indicating placental transfer of these elements. Our results suggest that hepatic copper levels $<200 \mu\text{g g}^{-1}$ dry weight may result in copper depletion in pregnant cows and hepatic mercury concentrations above $0.5 \mu\text{g g}^{-1}$ dry weight may negatively affect fertility in caribou cows. Hepatic mercury levels were negatively correlated with cow body weight, suggesting an adverse effect on body condition. Element concentrations found in tissues from these caribou are not considered to be of a health concern for those consuming this traditional food.

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1. Introduction

Caribou (*Rangifer tarandus groenlandicus*) are significant to Greenland culture and economy. Not only does caribou meat (muscle)

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constitute a substantial portion of the diet, but liver consumption is popular. Hence there is concern about the presence of contaminants and the potential effect on human health. For example, in northern Quebec, Canada, levels of cadmium (Cd) exceeded tolerance thresholds for human consumption in many caribou livers and kidneys (Robillard et al., 2002). The Akia-Maniitsoq (AM) and Kangerlussuaq-Sisimiut (KS) caribou populations are the largest in West Greenland (Cuyler et al., 2005, 2011). Although geographically close, these two populations are physically separated by the Sukkertoppen Ice Cap, with KS to the north and AM to the south (Fig. 1). On its north side this ice cap creates a dry climate with desert steppe vegetation poor in lichen abundance while rich in grasses and sedges for the KS caribou. In contrast, to the south, the AM range receives regular precipitation and lichens are common (Tamstorf, 2004). Rumen analysis (Lund et al., 2000)

revealed dietary differences between the two populations. In general, to the south the AM diet was dominated by lichens, which were notable by their absence in the northern KS diet of grasses and dwarf-shrub (Lund et al., 2000). Further, AM evidenced an early to late winter diet shift from lichens to more dwarf-shrub while for KS the same seasonal shift was from dwarf-shrub to more grasses/sedges (Lund et al., 2000).

Aastrup et al. (2000) analysed contaminants in muscle and liver tissue collected 1996–97 from these same two caribou populations, and reported significantly more lead (Pb), Cd, selenium (Se) and mercury (Hg) in liver samples from AM caribou than from KS, while copper (Cu) was greater in KS. The dissimilar AM and KS diets described by Lund et al. (2000), explain some of the differences, e.g., grasses are known for higher tolerance of Cu accumulation from soil (Plenderleith and Bell, 1990). Further, with the exception of Hg and Se, which were



Fig. 1. Locations of the two West Greenland caribou populations, Akia-Maniitsoq and Kangerlussuaq-Sisimiut, in the North and Central regions. Sample collection sub-areas indicated in red. Elevations of 0–200 m above sea level are green; elevations >200 m are yellow; ice caps are white.

higher, most element levels were similar to those in caribou elsewhere in the Arctic and were greatest in late winter relative to early winter (Aastrup et al., 2000).

Although assessing levels of contaminants in wildlife is important, it is equally important to determine whether concentrations are increasing or decreasing over time. If concentrations are increasing, then it is important to determine whether the concentrations may pose health issues for people consuming the food, or the animals themselves. Decreasing concentrations could provide supportive evidence for the efficacy of local and/or international environmental controls. Most contaminant time series for Greenland biota involve marine or freshwater systems, e.g. Hg has increased while Cd decreased over time in ringed seals (*Phoca hispida*) (Riget et al., 2004). With more than a decade since the first terrestrial study, this project adds a second series of data points to the terrestrial time series for West Greenland caribou.

If there is a difference in element concentrations in caribou collected in the two time periods, then we can also address the question of the cause of those differences. Any difference seen would necessarily be a difference in environmentally available quantities of these elements, either from naturally occurring cycles or from anthropogenically-produced contaminants in the atmosphere. Greenland is considered to be relatively free of local anthropogenic sources of pollution, the main source of pollution coming from Eurasia (Dietz et al., 1998). In the terrestrial environment, the mode of transport of this pollution must necessarily be atmospheric. Lichens are particularly effective at absorbing airborne nutrients as well as contaminants, whereas graminoids absorb their nutrients and contaminants more from the substrate (Gamberg, 2009; Garty, 2001). If changes in local naturally occurring concentrations of these elements are responsible for the differences we see between time periods, it should be more apparent in the KS herd, where lichens are not a primary food source. Alternatively, if changes in anthropogenically produced and atmospherically carried contaminants are responsible for the differences, it should be more apparent in the AM herd, where lichens constitute a major winter food source.

1.1. Present study

To examine possible temporal changes in element concentrations, we did late winter (March/April) collections of AM and KS caribou cows in 2008 and 2009 respectively. We examined and compared the contaminant content of the kidney, liver and muscle tissues of these two populations, accounting for cow age. Changes in contaminant levels over time within the same herds were assessed using data from Aastrup et al. (2000). Although Cd and Hg are of primary concern for human health (Robillard et al., 2002), we also examined 34 elements including the following: aluminum (Al), As, Cu, Pb, Se and Zn. We discuss whether element concentrations have changed in these populations since the initial collection in 1996–97 and possible relationships between cow contaminant levels to diet, reproductive state, and body condition.

2. Materials and methods

2.1. Study area

The west coast of Greenland has a complex topography with deep fjords, high mountains, and several glaciers that cut from the Greenland Ice Cap down to the sea. Winter sea ice is poorly developed or non-existent. Aside from valley bottoms and AM's Akia coastal low lands, topography is rugged uplands or mountainous. The underlying geology of the AM and KS study areas both consist of metamorphic orthogneisses from the Archaean Eon (c. 3 billion years ago) (Hollis et al., 2005). While the gneisses in the KS study area have been more intensely deformed and reworked due to a mountain building event in the Proterozoic Eon (c. 1.9 billion years ago) (Engström and Klint, 2014) when compared to the AM study area, the mineralogical content of the gneisses in the two areas are fairly similar. Sporadic cover of glacial

moraine and boulders are common in both areas. Within the KS study area there are many closed basin salt/saline lakes where pH can exceed 7.5 (Williams, 1991). These are caused by the dry continental climate with its high negative ratio of precipitation to evaporation during the summer coupled with poor ground water inputs and lack of surface outflow (Anderson and Bennike, 1997; Anderson et al., 1999; Aebly and Fritz, 2009). Generally, vegetation and climate are low-arctic in both areas, but precipitation is very different (Tamstorf, 2004). A wet maritime climate characterizes AM habitat, which is sandwiched between a dominating high pressure over the Greenland Ice Cap to the east and low-pressure systems that regularly sweep over it from the southwest. At nearby Nuuk (Fig. 1), the annual precipitation is 752 mm and the mean is July temperature 6.5 °C (DMI, 2014). In contrast the KS inland habitat is dry continental desert steppe. The Sukkertoppen Ice Cap blocks southwest storms appropriating the precipitation creating a rain shadow, so that annual KS precipitation is <150 mm and mean July temperature 10.7 °C (DMI, 2014). Lichens are common on the AM range, but KS inland range is dominated by dwarf shrub heath and grasslands with almost no lichens. Plant communities vary with elevation, aspect, and proximity to the maritime influences of the sea coast or the Greenland Ice Cap's continental climate. For detailed descriptions of the study area see Lund et al. (2004); Tamstorf (2004) and Tøttrup (2009).

2.2. Sampling

In March/April 2008, 41 adult female caribou were culled from the AM herd and in March 2009, 40 adult female caribou were culled from the KS herd. This was part of a body condition study being conducted by the Greenland Institute of Natural Resources and the Circum Arctic Rangifer Monitoring and Assessment (CARMA) network (CARMA, 2012). Entire kidneys, and portions of liver and skeletal muscle (gastrocnemius) were taken for contaminant analysis. Sampling protocols for tissue samples and body condition indices followed standardized monitoring techniques described in Kutz et al. (2013). All meat resulting from collections was owned by the Greenland Government, who distributed carcasses among hospitals, schools, old age homes and local commercial hunters for sale at their market.

Tissue samples were analyzed individually for a suite of 34 elements using the inductively coupled plasma technique with mass spectroscopy at Environment Canada's National Laboratory for Environmental Testing (Ontario, Canada). Prior to analysis, each kidney was individually homogenized. Briefly, subsampled kidney (0.5 g) was digested using an 8:1 ratio of nitric acid and hydrogen peroxide. A suite of 33 elements was determined using inductively coupled plasma mass spectrometry (ICP-MS) while mercury was determined by cold vapor-atomic absorption spectroscopy. Three standard reference materials (DOLT-2, DORM-2, TORT-2) were run for each batch of 20 samples. Incisors were extracted from each caribou and used to age the caribou using the cementum technique for adults (Gasaway et al., 1978) and tooth eruption for animals under three years of age.

Kidney, liver and muscle data were analyzed separately. Although 34 elements were analyzed, only eight elements of concern were explored in detail (Al, As, Cd, Cu, Pb, Hg, Se, Zn). If more than 50% of the samples were below detection limits for a particular element, those data were not analyzed. Otherwise half the detection limit was used in calculations. Nine data points (specific element concentrations) were considered outliers (greater than two standard deviations away from the mean) and were removed from the dataset. These outliers were from both herds and all three tissues and are considered true outliers rather than errors. Where the data were not normal, transformations ($\ln(x)$) were made to normalize the data before analysis. If that was unsuccessful, non-parametric statistics were used to analyze the data.

Linear regression or Spearman's rank order correlation was used to test the effect of age and body condition indices on element concentrations. Liver and muscle data collected for the same herds in March/April

of 1997 (Aastrup et al., 2000) were compared with data from this study to determine whether element concentrations have changed over time. The 2008 AM and 2009 KS foraging and collection areas were identical to those used in the previous study. Similarly the 2008 and 2009 spring collection period, March–April, was the same as in 1997. Although the 1997 samples (as well as the 1996 samples) were analyzed using atomic absorption spectrophotometry while the more recent samples were tested using inductively coupled plasma spectroscopy, the results should be comparable (results for both studies were generally <12.5% of certified values for standard reference materials [Asmund and Cleemann, 2000 and this study]). Either a t-test or a Mann-Whitney rank sum test was used to test differences between herds and time periods and to test the effect of pregnancy and extended lactation on element concentrations. Neither the two herds nor the two sampling times differed significantly in ages, so no adjustment was used when comparing element concentrations. Data from Aastrup et al. (2000) were also used when testing the effect of pregnancy on element concentrations. Statistical significance was considered when $\alpha > 0.05$.

3. Results and Discussion

Arithmetic mean concentrations of elements in kidney, liver and muscle from the AM and KS herds are presented in Table 1. Values reported as ‘<’ are below the detection limits for that element. Element concentrations were similar to those reported for other caribou and reindeer (Table 2), with the exception of hepatic Cu concentrations in the KS herd, which will be discussed in more detail further in the discussion. Cd was positively correlated with animal age in kidney ($r = 0.47$, $p < 0.0001$) and muscle ($r = 0.29$, $p < 0.0001$) but not in liver, while Se was negatively correlated with age in kidneys ($r = -0.24$, $p = 0.03$), but not liver or muscle. It is generally important to consider age as a potential cofactor when comparing element concentrations among herds, seasons or year of collection or between genders, since it has been shown to significantly affect some element concentrations in caribou (Gamberg and Scheuhammer, 1994). However, since ages did not differ between groups being compared in this study, age was not considered in the between herd and year of collection analyses. It is however, still important to consider when comparing data from this study to published literature on other caribou herds.

Element concentrations were generally higher in the AM herd than the KS herd with a few exceptions (Table 1). Notably, Cu was markedly higher in KS livers. Lichens tend to have higher concentrations of most elements (except Cu) than graminoids (Gamberg, 2009), which are often used in phytoremediation at sites contaminated by mining activity owing to their ability to accumulate Cu (Van der Lelie et al., 2001; Kabata-Pendias, 2011; Gutierrez and Arteaga, 2013). Given that the KS herd had virtually no lichens in its range (Tamstorf, 2004), we would expect the AM herd to have higher concentrations of most elements.

Al, As, Pb, Hg, and Se concentrations were higher in all three tissues (muscle, liver and kidney) in the AM herd. Cd levels were higher in liver and kidney in the AM herd but not significantly higher in muscle, while Zn was higher in kidney from the AM herd but not liver or muscle. Conversely, Cu concentrations were higher in liver in the KS herd, as would be expected from caribou consuming graminoids all year rather than switching to a winter diet of lichens (Lund et al., 2000). It is of note that although Cu concentrations were higher in the muscle tissue from the AM herd, liver concentrations are the best indicator of Cu status in mammals (Puls, 1994). These results confirm those of Aastrup et al. (2000) who found higher Cd, Pb, Hg and Se levels in muscle and liver from the AM herd as compared to the KS herd but higher Cu levels in the KS herd. Although diet may be a significant factor in determining different contaminant profiles in the two caribou herds, ingestion of soil may also play a role. MacDonald and Gunn (2004) found a soil ingestion rate of 20% of the diet for caribou in Arctic Canada.

The observed differences in Cu between the KS and AM herds cannot be attributed to underlying geology, as both areas have similar mineralogical content (Engström and Klint, 2014). However, the characteristics of the saline lakes at KS may be partially responsible for the higher Cu values in KS caribou. Cu is more soluble in saline than in fresh water and solubility increases above pH 7.5 (Zeitoun, 1969). Further, if the saline water is lacking or low in biotic and organic compounds then Cu will also remain soluble (Richey and Roseboom, 1978; Gerringa et al., 1998). These attributes describe the saline lakes in the KS area (Williams, 1991; Anderson et al., 1999). We postulate that KS caribou could be accumulating higher levels of Cu through drinking saline lake water and foraging on Cu tolerant grasses and sedges along the saline lake shores. Investigations testing the Cu content in the lakes, grasses and caribou rumen contents are needed to test this speculation.

Generally, most elements were higher in 1997 than in 2008/09, although there were some exceptions (Table 2). Hepatic Se was higher in the more recent time period in both herds, and Cu did not differ between time periods for either tissue in either herd. Although we have data for both muscle and liver, element concentration in muscle reflects recent forage consumption while liver is more indicative of the long-term status of the animal as these elements tend to be sequestered there (Loseto et al., 2008). The difference between 1997 liver concentration and 2008/09 concentrations of Al, As and Pb are all greatest for the KS herd, indicating natural fluctuations of these elements. However, while in 1997 concentrations of both Cd and Hg were higher in the AM herd (similar to the other elements), in 2008/09 they were actually higher in the livers from the KS herd (Cd was significantly higher while Hg was not statistically different). This suggests that the main source of Cd and Hg for the AM caribou is through atmospheric deposition on lichens, while naturally occurring variation is affecting hepatic Cd and to a lesser degree, Hg, in the KS herd.

Table 1
Age, organ weights (g fresh weight) and element concentrations ($\mu\text{g g dry weight}$) in tissues from female caribou collected from two herds from western Greenland in 2008/09 (arithmetic mean \pm SE).

Herd	Kidney		Liver		Muscle	
	Akia-Maniitsoq	Kangerlussuaq–Sisimiut	Akia-Maniitsoq	Kangerlussuaq–Sisimiut	Akia-Maniitsoq	Kangerlussuaq–Sisimiut
N	41	40	40	40	20	20
Age (years)	4.9 \pm 2.6	6.4 \pm 3.2	4.9 \pm 2.6	6.6 \pm 3.3	5.9 \pm 2.8	6.3 \pm 3.6
Organ weight (g)	65.3 \pm 1.5*	72.9 \pm 1.9	665 \pm 12.2*	798 \pm 17.1		
Moisture (%)	79.1 \pm 1.4	76.5 \pm 1.6	67 \pm 1.5	66.6 \pm 2.8	70.8 \pm 1.1	70.8 \pm 1.1
Aluminum	5.34 \pm 5.42*	0.87 \pm 0.38	2.2 \pm 0.60*	0.83 \pm 0.29	0.79 \pm 0.86*	0.20 \pm 0.16
Arsenic	0.03 \pm 0.01*	<0.004	0.01 \pm 0.00*	<0.008	0.03 \pm 0.01*	<0.008
Cadmium	14.6 \pm 8.7*	10.5 \pm 8.1	1.8 \pm 0.8*	1.2 \pm 0.8	0.006 \pm 0.003	0.005 \pm 0.003
Copper	20 \pm 6	19 \pm 2	100 \pm 48*	240 \pm 104	13 \pm 1*	12 \pm 2
Lead	0.63 \pm 0.24*	0.07 \pm 0.06	1.83 \pm 1.26*	0.09 \pm 0.09	0.01 \pm 0.01*	<0.004
Mercury	2.22 \pm 0.78*	1.41 \pm 0.39	0.46 \pm 0.12*	0.24 \pm 0.07	0.05 \pm 0.02*	0.02 \pm 0.01
Selenium	4.76 \pm 0.56*	2.72 \pm 0.52	1.86 \pm 0.45*	0.44 \pm 0.19	0.77 \pm 0.08*	0.17 \pm 0.10
Zinc	101 \pm 6*	88 \pm 7	96 \pm 22	102 \pm 19	82 \pm 9	90 \pm 26

* Indicates a significant difference between herds ($p < 0.05$).

Table 2A comparison of mean element concentrations ($\mu\text{g g dry weight}$) in caribou and reindeer tissues from the circumpolar north.

	Herd	N	Moisture	Al	As	Cd	Cu	Pb	Hg	Se	Zn	Ref
Kidney	Akia-Maniitsoq	41	79.1	5.34	0.03	14.60	20.0	0.63	2.22	4.76	101.0	This study
	Kangerlussuaq-Sisimiut	40	76.5	0.87	<0.004	10.50	19.0	0.07	1.41	2.72	88.0	This study
	Reindeer Sweden	64				14.36*	21.7*	1.36*			105.9*	Eriksson et al. (1991)
	Reindeer Norway	248				25.91*						Froslie et al. (1986)
	Canada											
	Lake Harbour (NU)	10		11.72		31.98		0.47	11.63*			Elkin and Bethke (1995)
	Cape Dorset (NU)	10		6.58		14.06		0.42	5.68*			Elkin and Bethke (1995)
	George River (QC)	27				23.77*		0.91*	2.55*			Robillard et al. (2002)
	Leaf River (QC)	177				40.59*		1.27*	6.32*			Robillard et al. (2002)
	Southampton Island (NU)	10		6.36		18.79		0.33	10.09*			Elkin and Bethke (1995)
	Qamanirjuaq (NU)	78	78.0	1.39	0.05	27.23	22.2	0.40	5.67	4.26	107.4	Gamberg, 2013
	Bathurst (NT)	10		5.64		9.68		0.11	2.36*			Elkin and Bethke (1995)
	Bluenose (NT)	20		1.48		42.60		0.21	10.45			Larter and Nagy (2000)
	Banks Island (NT)	20		1.56		12.23		0.98	5.43			Larter and Nagy (2000)
	Porcupine (YT)	218	79.0	2.21	0.14	30.72	24.7	0.14	1.49	4.75	120.2	Gamberg, 2013
	Akia-Maniitsoq (2008)	40	67.0	2.20	0.01	1.80	100.0	1.83	0.46	1.86	96.0	This study
	Akia-Maniitsoq (1997)	24	72.9	2.63	0.03	3.07	114.5	3.94	0.96	1.13	110.9	Astrup et al. (2000)
	Kangerlussuaq-Sisimiut (2009)	40	66.6	0.83	<0.008	1.20	240.0	0.09	0.24	0.44	102.0	This study
	Kangerlussuaq-Sisimiut (1997)	23	72.6	2.97	0.03	0.76	235.9	0.23	0.23	0.35	101.4	Astrup et al. (2000)
Liver	Reindeer Sweden	64				1.21*	78.1*	1.30*	0.12*		89.4	Eriksson et al. (1991)
	Reindeer Norway	57			0.03*	1.27*	129.1*	1.54*	0.23*		98.5	Froslie et al. (1984)
	George River (QC)	28				2.85*		2.70*	1.15*			Robillard et al. (2002)
	Leaf River (QC)	176				3.58*		2.70*	2.12*			Robillard et al., 2002
	Akia-Maniitsoq (2008)	20	70.8	0.79	0.03	0.01	13.0	0.01	0.05	0.77	82.0	This study
	Akia-Maniitsoq (1997)	24	75.6	2.17	0.10	0.01	12.9	0.02	0.12	0.72	72.7	Astrup et al. (2000)
	Kangerlussuaq-Sisimiut (2009)	20	70.8	0.20	<0.008	0.01	12.0	<0.004	0.02	0.17	90.0	This study
	Kangerlussuaq-Sisimiut (1997)	24	74.5	2.23	0.02	0.01	11.6	0.03	0.04	0.18	89.2	Astrup et al. (2000)
	George River (QC)	28				0.03*		0.03*	0.07*			Robillard et al. (2002)
	Leaf River (QC)	104				0.06*		0.11*	0.09*			Robillard et al. (2002)
Muscle	Akia-Maniitsoq (2008)	20	70.8	0.79	0.03	0.01	13.0	0.01	0.05	0.77	82.0	This study
	Akia-Maniitsoq (1997)	24	75.6	2.17	0.10	0.01	12.9	0.02	0.12	0.72	72.7	Astrup et al. (2000)
	Kangerlussuaq-Sisimiut (2009)	20	70.8	0.20	<0.008	0.01	12.0	<0.004	0.02	0.17	90.0	This study
	Kangerlussuaq-Sisimiut (1997)	24	74.5	2.23	0.02	0.01	11.6	0.03	0.04	0.18	89.2	Astrup et al. (2000)
	George River (QC)	28				0.03*		0.03*	0.07*			Robillard et al. (2002)
	Leaf River (QC)	104				0.06*		0.11*	0.09*			Robillard et al. (2002)

* Wet weight was converted to dry weight using the following % moistures: kidney 78%, liver 67%, muscle 79% (based on data from this table).

Hepatic Cu was significantly higher in non-pregnant than in pregnant caribou in all spring-collected animals, but not in fall-collected animals (Fig. 2). This was expected since the fetus claims Cu at the expense of the dam, reducing Cu reserves in the dam as pregnancy progresses (Rombach et al., 2003). In the fall, early pregnancy would not have begun to drain the dam's Cu reserves whereas in spring, just before birth, Cu would be lowest in the dams. In both herds, as expected, non-pregnant animals had similar hepatic Cu concentrations in spring and fall, when comparing fall 1996 and spring 1997 data. In the AM herd, pregnant females had lower hepatic Cu in the spring than the fall, as anticipated, because the fetus preferentially takes up Cu at the expense of the dam. However, in the KS herd, although hepatic concentrations in pregnant females were higher in the fall than the spring, the difference was slight and not statistically significant. This difference may be due to differences in the absolute amount of Cu found in each herd. Concentrations in the KS herd were significantly higher than the AM herd and may be adequate to nourish the fetus and at the same time allow the dam to maintain adequate Cu reserves. Concentrations in the AM herd were much lower and the dams may have to experience Cu depletion to adequately provide for the fetus. This assessment may provide a reasonable indication of Cu requirements in wild caribou, suggesting that hepatic concentrations of less than 200 ^{-1} dry weight may result in Cu depletion in pregnant females. However, at least for the AM herd, this depletion appears to be a temporary one, Cu stores being replenished over the summer through the consumption of Cu-rich graminoids. As an aside, hepatic Cu concentration in both herds was generally negatively correlated to kidney fat ($r = -0.48$, $p < 0.0001$). This was expected since Cu stores deplete during pregnancy and pregnant animals consistently had more kidney fat and greater body weight.

Hepatic Hg was also significantly higher in non-pregnant than in pregnant caribou in spring-collected animals from the KS herd, but not in fall-collected animals from the same herd (Fig. 2). This is expected since Hg may be transferred through the placenta from the dam to the fetus (World Health Organization, 2003), thereby creating an elimination pathway for this toxic element, which will be more apparent in spring at the end of gestation, than in the fall at the beginning of

pregnancy. The situation in the AM herd is not as clear. While Hg is higher in non-pregnant than pregnant females, which is normal, the only significant difference was seen in fall-collected animals, not in spring-collected caribou (Fig. 2). It appears that although there is some loss of Hg through placental transfer, Hg concentrations are so much higher in this herd that the reduction in levels due to pregnancy is relatively small and therefore not statistically significant. The difference seen in pregnant and non-pregnant females in the fall is puzzling since placental transfer cannot be the reason. These data were re-analyzed to see if Hg concentrations were different in fall-collected females that had a calf-at-heel (from the previous year) or none, to determine if differences could be attributed to the transfer of Hg through the placenta and through lactation from the previous spring, but no difference in those two groups was apparent. One possible explanation might be that higher hepatic Hg concentrations may reduce fertility so that those individuals do not become pregnant. Adverse reproductive effects have been associated with elevated levels of mercury in a number of wildlife species (Scheuhammer et al., 2012). In this case the Hg concentrations would be affecting pregnancy rather than pregnancy affecting Hg concentrations as was seen in spring-collected animals. It is of note that in the KS herd where Hg concentrations were significantly lower, there was no difference between hepatic concentrations of Hg in pregnant and non-pregnant females in the fall, indirectly supporting this hypothesis. If this is the case, then Hg hepatic concentrations of about $0.5 \mu\text{g g}^{-1}$ dry weight or above may be negatively affecting fertility in these caribou. Our conjecture regarding Hg impacting fertility is supported by herd abundance data. The AM population has declined steadily since 2001 while the KS caribou has remained relatively stable (Cuyler et al., 2011).

Hepatic Pb concentrations were also generally lower in pregnant females than in non-pregnant female caribou in spring-collected animals, but not in those collected in the fall (Fig. 2). As with Hg, this is expected since Pb may also be transferred through the placenta from the dam to the fetus (World Health Organization, 2001), thereby reducing concentrations in the adult pregnant females. However, in the AM herd, Pb was actually higher in pregnant than non-pregnant caribou in animals

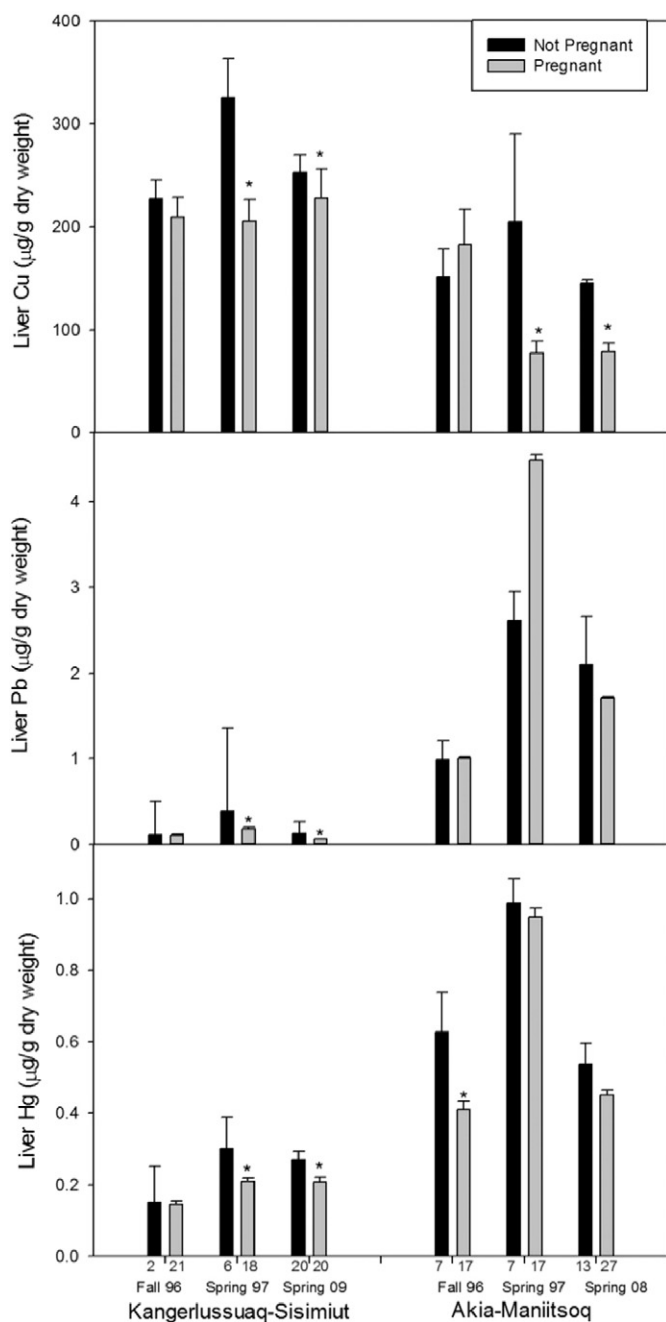


Fig. 2. Element concentrations in caribou livers from the Akia-Maniitsoq (AM) and Kangerlussuaq-Sisimiut (KS) herds of western Greenland collected from 2008/09 (this study) and 1996–1997 (Aastrup et al., 2000). * indicates a significant difference between pregnant and non-pregnant caribou ($p < 0.05$). Sample sizes are indicated below the x-axis.

collected in the spring of 1997. This may be related to an unusual strongly negative spike in the North Atlantic Oscillation Index (NAO) in 1996, which represents the flow of warm, wet air masses from the mid-latitude Atlantic Ocean to the Greenland/Labrador Sea region. Accordingly, air temperature anomalies indicated unusual warming during the month of February on the east coast of Greenland (Stein, 1997) and precipitation in the AM region was well above normal in 1996 almost doubling depending on whether rain or snow is considered (DMI, 2014). This NAO spike may have indirectly increased long-range transport of Pb by increasing the carrying capacity of the atmosphere. Pregnant caribou would need to consume more forage (and in this case, perhaps significantly more Pb) than non-pregnant caribou to support the growth of the fetus. Although a similar effect was not apparent

in hepatic Hg in these animals, the same situation may have occurred, but if Hg was more efficiently transferred (than Pb) to the fetus in the pregnant animals, there could be no net effect.

Hepatic Hg concentrations were negatively correlated with body weight in both herds (Fig. 3). The correlation was statistically significant for both herds and the slopes of the lines similar, although the fit was not particularly high in either herd ($r = -0.34$, $p = 0.001$ in the AM herd; $r = -0.31$, $p = 0.003$ in the KS herd). Still, this does suggest that higher Hg concentrations may contribute to a lower body weight and hence may negatively affect body condition.

As and Cd concentrations in both herds were below the level that would be considered toxic for cattle, whereas concentrations of Al and Se in the AM herd and Se in the KS herd were higher than would be considered normal for cattle, particularly in liver (Puls, 1994). Whether these are toxic levels for caribou is not known. Generally Pb and Hg concentrations in these herds are not near toxic levels, but some individuals have concentrations that would be considered 'high' to 'toxic' for domestic cattle. Cu and Zn are essential elements. Since they are homeostatically controlled, excess Cu and Zn is excreted in the urine, and toxicity is rare under normal conditions. Cu deficiency is more likely in the natural environment and has been noted in Alaskan moose with faulty hoof keratinization and reduced reproductive rates (Flynn et al., 1977). Cu and Zn concentrations in both herds in this study exceeded what would be considered normal for cattle; whether these levels are toxic for caribou is not known, but considered unlikely.

Element concentrations found in tissues from these caribou are not considered to be of concern for those consuming this traditional food. According to maximum recommended intake by the World Health Organization (2010), the most restrictive limits would be a 70 kg adult consuming a maximum of 132g (AM herd) to 164 g (KS herd) of kidney each and every week (or two kidneys every week) (Table 3). It is of note that the most restrictive recommendation for muscle tissue is a maximum of 5.6 kg/week or 800g/day, every day for the lifetime of that individual. Therefore, we do not consider these element concentrations to be of concern for human health and do not recommend limiting consumption of liver, kidney or muscle from these caribou for health reasons.

4. Conclusions

Although in close proximity, these two West Greenland caribou populations had divergent contaminant profiles, which may reflect their differing diets, the result of the intervening ice cap. The AM herd,

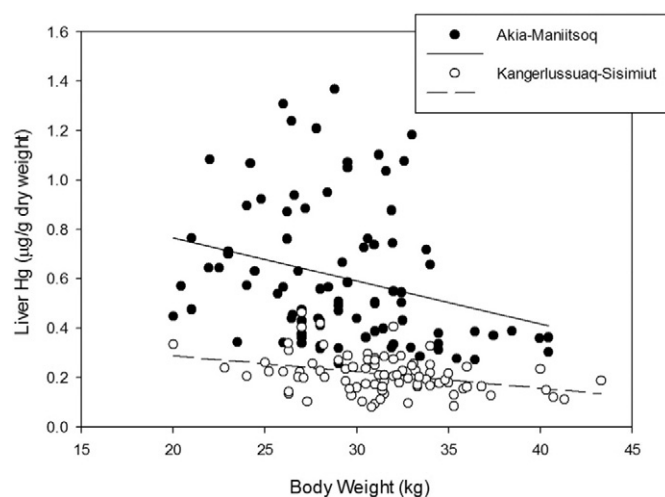


Fig. 3. Liver Hg concentrations relative to body weight in the Akia-Maniitsoq ($r = -0.34$, $p = 0.001$) and Kangerlussuaq-Sisimiut caribou herds ($r = -0.31$, $p = 0.003$) from western Greenland collected in 2008–2009 (this study) and 1996–1997 (Aastrup et al., 2000).

Table 3

Maximum recommended weekly intake of caribou tissue (kg wet weight) based on FAO/WHO recommendations (World Health Organization, 2010). PTWI = provisional tolerable weekly intake; PTMI = provisional tolerable monthly intake; PMTDI = provisional maximum tolerable daily intake; RfD = oral reference dose. Note that Pb was not included because no PTMI or RfD currently exists for Pb.

FAO/WHO recommendation	Maximum recommended weekly intake of caribou tissue for a 70 kg adult (kg wet weight)					
	Kidney		Liver		Muscle	
	AM	KS	AM	KS	AM	KS
Al PTWI 1 mg/kg bw	63	340	94	253	305	1198
As RfD 0.0003 mg/kg bw/day ¹	27	156	41	55	19	63
Cd PTMI 25 µg/kg bw/month	0.132	0.164	0.7	1	226	258
Cu PMTDI 0.5 mg/kg bw/day	58	56	7	3	67	73
Hg PTWI 4 µg/kg bw	0.6	0.8	2	3	21	49
Zn PMTDI 0.3–1 mg/kg bw/day ²	7	7	5	4	6.2	5.6

¹ US EPA (1991); FAO/WHO 1988 recommendation, was withdrawn in 2011 and has not been replaced.

² Used 0.3 mg/kg bw/day for calculations

south of the ice cap, consumes more lichens, which are subject to contamination from atmospheric deposition. This herd exhibits significantly higher levels of Al, As, Cd, Pb, Hg, Se and Zn than the KS herd. To the north, in the dry desert steppe, the KS herd consumes more grasses, which are less affected by atmospheric deposition as much as natural cycling of the elements. This herd exhibits generally higher Cu concentrations which may be associated with physical characteristics of the area. Advanced pregnancy was associated with significantly less hepatic Cu, Pb and Hg, indicating placental transfer from dam to foetus. Hepatic copper levels <200 µg g⁻¹ dry weight may result in copper depletion in pregnant cows. Hepatic mercury concentrations higher than 0.5 µg g⁻¹ dry weight may negatively affect fertility in caribou cows. In both populations, cow body weight was negatively correlated with hepatic mercury concentration, which suggests an adverse effect of mercury on body condition. Element concentrations found in tissues from these caribou are not considered to be of a health concern for those consuming this traditional food.

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