

# Levels of Cadmium, Lead, Mercury and <sup>137</sup>Caesium in Caribou (*Rangifer tarandus*) Tissues from Northern Québec

S. ROBILLARD,<sup>1,2</sup> G. BEAUCHAMP,<sup>1</sup> G. PAILLARD<sup>3</sup> and D. BÉLANGER<sup>1,4</sup>

(Received 6 November 2000; accepted in revised form 8 May 2001)

**ABSTRACT.** Levels of cadmium (Cd), lead (Pb) and total mercury (Hg) were assessed in samples of muscle, kidney, and liver from caribou (*Rangifer tarandus*; n = 317) harvested in two regions of northern Québec between 1994 and 1996. Levels of <sup>137</sup>caesium (<sup>137</sup>Cs) were also examined in muscle samples. Log concentration of the three metals varied significantly among tissues and was lowest in diaphragm muscle and highest in kidneys and liver. Mean Cd (wet weight, w.w.) concentration was 0.01 µg·g<sup>-1</sup> in muscle, 7.69 µg·g<sup>-1</sup> in kidneys and 1.13 µg·g<sup>-1</sup> in liver. Levels of Cd exceeded tolerance thresholds for human consumption in nearly all kidney samples and in nearly half the liver samples. Mean Pb concentration (w.w.) was 0.05 µg·g<sup>-1</sup> in muscle, 0.26 µg·g<sup>-1</sup> in kidneys and 0.95 µg·g<sup>-1</sup> in liver, with few samples exceeding consumption thresholds. Mean total Hg concentration (w.w.) in muscle was 0.03 µg·g<sup>-1</sup>, 1.26 µg·g<sup>-1</sup> in kidneys and 0.67 µg·g<sup>-1</sup> in liver, with concentrations exceeding consumption thresholds in most kidney samples and nearly half the liver samples. Regional differences occurred in log concentration of the three metals for most tissues, with the western region consistently showing higher values. Mean log Cd and Pb concentrations increased with age in kidneys, but log Pb decreased with age in muscle samples. Interactions between month of collection and sex and region also occurred for some metals in some tissues. Mean level of <sup>137</sup>Cs in muscle samples was 94.7 Bq·kg<sup>-1</sup>, never exceeding the acceptable limit for human consumption.

**Key words:** caribou (*Rangifer tarandus*), cadmium, lead, mercury, <sup>137</sup>caesium, northern Québec, spatial variation

**RÉSUMÉ.** On a mesuré les niveaux de cadmium (Cd), de plomb (Pb) et de mercure total (Hg) dans des échantillons de muscle, de rein et de foie de caribous (*Rangifer tarandus*; n = 317) prélevés dans deux régions du Québec nordique entre 1994 et 1996. On a en outre étudié les niveaux de césium 137 (<sup>137</sup>Cs) dans des échantillons musculaires. Les concentrations enregistrées des trois métaux montraient d'importantes variations parmi les divers tissus et étaient les plus faibles dans le muscle du diaphragme et les plus élevées dans le rein et le foie. La concentration moyenne de Cd (poids frais, p. f.) était de 0,01 µg·g<sup>-1</sup> dans le muscle, de 7,69 µg·g<sup>-1</sup> dans le rein et de 1,13 µg·g<sup>-1</sup> dans le foie. Les niveaux de Cd dépassaient les seuils de tolérance pour la consommation humaine dans presque tous les échantillons de rein et dans près de la moitié des échantillons de foie. La concentration moyenne (p. f.) de Pb était de 0,05 µg·g<sup>-1</sup> dans le muscle, de 0,26 µg·g<sup>-1</sup> dans le rein et de 0,95 µg·g<sup>-1</sup> dans le foie, avec peu d'échantillons dépassant les seuils de consommation. La concentration moyenne de Hg total (p. f.) dans le muscle était de 0,03 µg·g<sup>-1</sup>, de 1,26 µg·g<sup>-1</sup> dans le rein et de 0,67 µg·g<sup>-1</sup> dans le foie, avec des concentrations qui dépassaient les seuils de consommation dans la plupart des échantillons de rein et presque la moitié des échantillons de foie. Des différences régionales sont apparues dans les concentrations enregistrées des trois métaux pour la plupart des tissus, la zone occidentale montrant constamment des valeurs plus élevées. Avec l'âge, les concentrations moyennes enregistrées pour le Cd et le Pb augmentaient dans le rein, alors que celles de Pb diminuaient dans le muscle. Des interactions entre le mois des prélèvements, le sexe et la région se produisaient aussi avec certains métaux et certains tissus. Le niveau moyen de <sup>137</sup>Cs dans les échantillons musculaires était de 94,7 Bq·kg<sup>-1</sup>, ne dépassant jamais la limite acceptable pour la consommation humaine.

**Mots clés:** caribou (*Rangifer tarandus*), cadmium, plomb, mercure, césium 137, Québec nordique, variation spatiale

Traduit pour la revue *Arctic* par Nésida Loyer.

## INTRODUCTION

Since the beginning of the industrial era, northern Québec (55°–62° N, 59°–80° W, Canada), like many northern ecosystems, has been exposed to large quantities of potentially toxic substances via long-range atmospheric transport (Barrie

et al., 1992). Moreover, the Chernobyl accident, which occurred in 1986, resulted in a 14–15% increase in concentrations of fission-related isotopes in Canadian Arctic lichens (Taylor et al., 1988). The presence of anthropogenic heavy metals, such as cadmium (Cd), lead (Pb) and mercury (Hg), and radioisotopes, such as <sup>137</sup>caesium (<sup>137</sup>Cs),

<sup>1</sup> Faculty of Veterinary Medicine, University of Montréal, CP 5000, St-Hyacinthe, Québec J2S 7C6, Canada

<sup>2</sup> Present address: Laboratoire de biologie animale, Université du Maine, 72085 Le Mans cedex France

<sup>3</sup> Ministère de l'Agriculture, des Pêcheries et de l'Alimentation, 2700 Einstein, Ste-Foy, Québec G1P 3W8, Canada

<sup>4</sup> Corresponding author: belande@medvet.umontreal.ca

has been documented in different wildlife species in these remote regions (Crête et al., 1987, 1989, 1990, 1993; Thomas et al., 1992; Gamber and Scheuhammer, 1994; Elkin and Bethke, 1995; Langlois and Langis, 1995; Kim et al., 1998; Braune et al., 1999; Aastrup et al., 2000; Larter and Nagy, 2000).

In northern ecosystems, the barren-ground caribou (*Rangifer tarandus*), as an herbivore, is exposed to toxic substances mainly through its plant diet. In northern Québec, lichens represent up to 77% of the caribou diet during the fall and winter seasons (Gauthier et al., 1989). Lack of a root system, slow growth, and large surface area in lichens are factors contributing to a large concentration of toxic substances, such as Cd, Pb, Hg, and  $^{137}\text{Cs}$  in these plants (Crête et al., 1989, 1992; Thomas et al., 1992; Nash and Gries, 1995; Macdonald et al., 1996). Several studies from Canada (Northwest Territories), Greenland, Norway, and Sweden report variation in tissue levels among different populations of caribou and reindeer (Froslic et al., 1986; Eriksson et al., 1990; Aastrup et al., 2000; Larter and Nagy, 2000). Both spatial and temporal heterogeneity in diet may account for the differences across studies. For instance, populations of caribou with a larger lichen component in their diet may show greater levels of heavy metal contamination. Variation in the availability of lichens as winter forage has also been linked to seasonal differences in contamination in caribou and reindeer (Hanson, 1967; Aastrup et al., 2000). Therefore, factors that influence the contaminant burden in lichens and other plant foods are expected to affect contamination levels in caribou. In addition, variation in underlying geology was suggested to account for some of the differences across studies (Larter and Nagy, 2000).

Once in the body, Cd, Pb, and Hg accumulate in different organs, such as the liver and kidneys. They also target other tissues, including muscles, but to a lesser degree (Goyer, 1991; Stansley et al., 1991; Medvedev, 1995). In contrast, absorbed  $^{137}\text{Cs}$  concentrates in different tissues but mainly in muscle (Holleman et al., 1971). Several factors may influence the fate of toxic substances once they have entered the body. For instance, chemical form, diet type, and age can influence the proportion of Pb that will be absorbed through the intestines (Humphreys, 1991).

In Québec, Native people such as the Inuit, as well as sport hunters, consume caribou on a more or less regular basis (Rapport Santé Québec, 1992). Moreover, a project of commercialization of caribou meat was undertaken in 1993. Following a study in northern Québec in the late 1980s, Crête et al. (1989) advised limiting the consumption of caribou liver and kidneys, basing their recommendations on those of the World Health Organization. To our knowledge, no surveillance system related to toxic substances is currently in place in northern Québec. Hence, the current levels of potentially toxic substances in caribou from this area are unknown. Although caribou are harvested from different regions in northern Québec, regional trends in toxic compound levels have not been investigated in

the area. Crête et al. (1989, 1990) focused primarily on the caribou herd belonging to the George River region. Harvested caribou may also originate from the more westerly Leaf River region.

We investigated spatial trends in Cd, Pb, Hg, and  $^{137}\text{Cs}$  levels in different tissues sampled from caribou belonging to two different, spatially segregated populations. We also evaluated the influence of age, sex, body mass, and time of year on tissue levels. Recent studies, often with limited sample sizes, have reported inconsistent effects of age and sex on metal levels in different tissues (Froslic et al., 1986; Crête et al., 1989; Eriksson et al., 1990; Aastrup et al., 2000; Larter and Nagy, 2000). Finally, we discuss current findings with respect to the risk associated with human consumption and the toxicological significance for these animals.

## METHODS

### *Study Area and Sampling Procedure*

The area studied is in northern Québec, more specifically in the central Ungava peninsula (Fig. 1). Granites and gneisses characterize soils in the area. Interspersed among small bodies of water in the area are two plant communities: shrub-tundra and stands of small trees. Lichens, and to a lesser extent moss- or shrub-dominated vegetation, dominate the plant communities. The lichen biomass available in the Leaf River basin is twice that found in the eastern area. The George River herd, in the middle of the eastern region, consists of approximately 800 000 individuals. The herd resides in the George River basin during calving and migrates south and west in the winter, reaching as far as Hudson Bay. The smaller Leaf River herd contains approximately 100 000 individuals (Crête and Doucet, 1998). The Leaf River herd resides to the west of Ungava Bay in the summer and migrates south to the Leaf River Basin and below in winter, overlapping with the George River herd. A third herd, more to the east and probably segregated spatially from the George River herd, resides in the Torngat Mountains during calving and summer and migrates to the coastal plain of eastern Ungava Bay in the winter (Schaefer and Luttich, 1998). Local industry and mining activity are limited, and many contaminants in the area represent fallout from long-range atmospheric transport.

Samples of diaphragm muscle, kidney, liver, and teeth (first incisors) were collected on caribou brought by Inuit hunters to four different abattoirs during the periods December 1994–May 1995 and December 1995–May 1996 (Fig. 1). In the first season, carcasses from only one abattoir (Umiujjaq) were sampled. Every second animal brought to the facilities was selected for analysis on sampling days. Animals collected at the Umiujjaq, Quaqtaq, and Kangiqsujuaq facilities are known to originate from the Leaf River region. Animals collected at the Kangiqsualajuaq facility originate from either the more

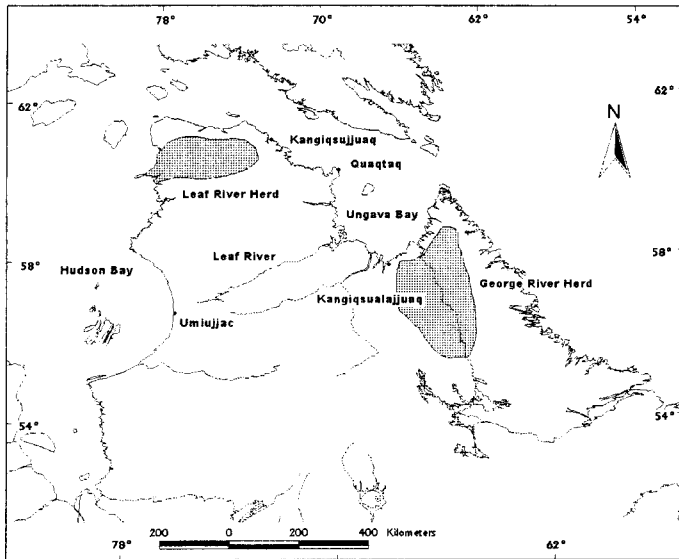


FIG. 1. Map of northern Québec, showing the location of the four abattoirs where caribou samples were collected between 1994 and 1996 and the boundaries of the summer residence areas of the two caribou herds.

easterly George River region or the Torngat Mountains region. For statistical purposes, caribou were assigned to one of two regions of origin: 1) Leaf River (LR) and 2) George River-Torngat Mountains (GRTM).

#### Analytical Procedure

Approximately 200 g of tissue (or occasionally one whole kidney) made up each sample. Specimens were frozen at  $-20^{\circ}\text{C}$  in separate plastic bags and shipped to the laboratories. The heavy metal analyses were conducted at the Ministère de l'Agriculture, des Pêcheries et de l'Alimentation in Québec City, Québec, Canada. Thirty diaphragm-muscle samples of 500 g from the Umiujjaq, Kangiqsujuaq, or Kangiqsualujuaq facilities were frozen at  $-20^{\circ}\text{C}$  in separate plastic bags and shipped to the Whiteshell Laboratories near Pinawa, Manitoba, Canada, for  $^{137}\text{Cs}$  analysis. Analytical methods and quality control for the  $^{137}\text{Cs}$  analysis are described in Macdonald et al. (1996).

For the heavy metal analyses, sample handling was conducted under clean-room, class 100 conditions. All tissue samples were homogenized using a blade mixer. For Cd and Pb determinations, 2 g aliquots of homogenized samples were first digested in 6 ml of nitric acid mixed with 2 ml of hydrogen peroxide 50% (w/w). Samples were then dried and microwave-digested, using 3 ml of concentrated nitric acid in a Prolabo Microdigest A300 system. Blanks containing 3 ml of nitric acid were included every 15 samples for quality control. Digests were diluted to 25 ml. Sample digestion for Hg analysis was conducted separately, using 5 g aliquots of homogenized samples. Samples were wet-digested by heating, using nitric acid and sulphuric acid consecutively in a Tecator digesting system.

Cd and Pb determination ( $\mu\text{g}\cdot\text{g}^{-1}$ , wet weight) was done with a Perkin Elmer 4100ZL furnace atomic absorption

spectrophotometer, which uses transverse heating of graphite cuvettes and longitudinal Zeeman effect background correction. Cd and Pb were measured at a wavelength of 228.8 nm and 283.8 nm, respectively. Metal determination was made on pyrocoated platforms, using ammonium phosphate and magnesium nitrate as matrix modifiers. The precision of the spectrophotometer was verified with a certified reference product (Setpoint from Analytical Products Group Inc #7878) at the beginning of each session. A quality-control sample was run every five samples. Calibration of the Cd and Pb analysis was continually checked using certified standard samples (Seigniory Chemical Products #SC 5213151-Cd; #SC 5305430-Pb). The detection limits were 1 ppb w.w. for Cd and 5 ppb w.w. for Pb. All the muscle samples were analyzed without further dilution. Liver samples could be diluted at 1/20 and kidney samples at 1/100 for Cd determination. For Pb determination, all the kidney samples were analyzed without dilution, and liver samples were diluted by one-third.

Total Hg ( $\mu\text{g}\cdot\text{g}^{-1}$ , wet weight) was determined by cold vapour technique atomic absorption spectrophotometry (Varian AA-1475), using tin chloride as a reducing agent. The wavelength used for Hg was 253.7 nm. Calibration of the analysis was continually checked every four samples, using certified standard samples (Seigniory Chemical Products #SC 5318457). The detection limit was 10 ppb w.w. To prepare samples for analyses, 5 g of tissue sample was digested in a solution containing 20 ml of nitric acid (69%) and 10 ml of sulphuric acid (99%). Final temperature during digestion was maintained between  $115^{\circ}$  and  $120^{\circ}\text{C}$ . Temperature was not increased above the boiling point of nitric acid. The procedure was stopped when the nitric acid in the solution was almost completely evaporated. These conditions insured permanent oxidizing conditions and protected samples from any possible Hg loss by evaporation, caused by the presence of organic compounds in the sulphuric acid solution.

Age was determined by counting cementum annuli of the central incisors. The error associated with aging has been estimated at  $\pm 1$  year (Matson's Laboratory, Milltown, Montana, U.S.A.). Body mass (kg) was obtained from noneviscerated animals upon their arrival at the abattoir.

#### Statistical Analyses

Concentration data were  $\log_{10}$  transformed to normalize distributions. Separate repeated-measure ANOVAs for the three metals were used to examine differences in log concentrations in the three different tissues of the same individual carcass. Tukey's post-hoc tests on log means were used to examine differences between tissues.

Separate general linear models were performed to test the effects of age category (immature = 0–2 years; adult = 3 years or older), sex, region of origin (LR, GRTM), time of year (month of collection treated as a class variable), and body mass on log Cd, Pb, and total Hg concentrations in the three different tissues. Interactions between region

and age, region and time of year, age and sex, and finally sex and time of year were also examined. Nonsignificant interaction terms were dropped from the final models. Directional trends with respect to time of year were assessed using regression parameters. Because of the small sample size, the analysis for log  $^{137}\text{Cs}$  was restricted to the effect of region, time of year, and age category. Data on metal levels were available in both study years for one abattoir only. In the models for this subsample, year of study failed to influence log concentration of all three metals in kidneys and liver; however, log concentration for some metals did vary between years in muscle samples. We thus pooled data from the two field seasons for kidney and liver analyses and used data from the second field season only in muscle analyses. In all analyses, the level of statistical significance was set at 0.05.

## RESULTS

A total of 317 carcasses were examined over the two-year study. A large proportion of caribou carcasses ( $n = 264$ , 83.3%) originated from the LR region. Sex was determined in 303 animals, and 80% were males. Males were on average  $5.3 \pm 2.4$  years old and weighed  $88.0 \pm 20.7$  kg. Females were on average  $3.9 \pm 2.8$  years old and weighed  $62.2 \pm 16.5$  kg.

Log concentration varied among tissues for the three metals ( $p < 0.0001$ ) and was lowest in muscle (Fig. 2). Highest levels occurred in kidneys for log total Hg and log Cd and in liver for log Pb (all pair-wise comparisons:  $p < 0.0001$ ).

Region of origin influenced log concentrations of Cd and Hg in muscle, kidneys, and liver, with samples from the LR region showing higher levels (Tables 1 and 2). The same effect of region on log Pb concentration was found in kidneys, but the trend was not significant in muscle and liver samples (Table 2).

Log concentration increased with the age of the animal for Cd and Pb in kidneys, but log Pb concentration decreased with age in muscle (Tables 1 and 2). Effects of sex proved limited and marginal: in liver samples, females showed higher log levels of Cd than males, and in kidneys, a pattern of decrease in log Hg concentration was documented in males, but not in females (Table 2). Variation between months of collection occurred in nearly all metal-tissue combinations, and time of year occasionally interacted with region or sex (Table 2). With respect to region, log concentration of Pb in kidneys and log Hg in kidneys and muscle decreased from December to May in the LR region, but it increased or remained stable in the GRTM region (Table 2). With respect to sex, log Hg concentration decreased with time of year in males, but not in females (Table 2). Effects of body mass proved limited and marginal: in kidneys, log Cd concentration increased with body mass, while log Pb concentration decreased with body mass (Table 2). All interaction terms

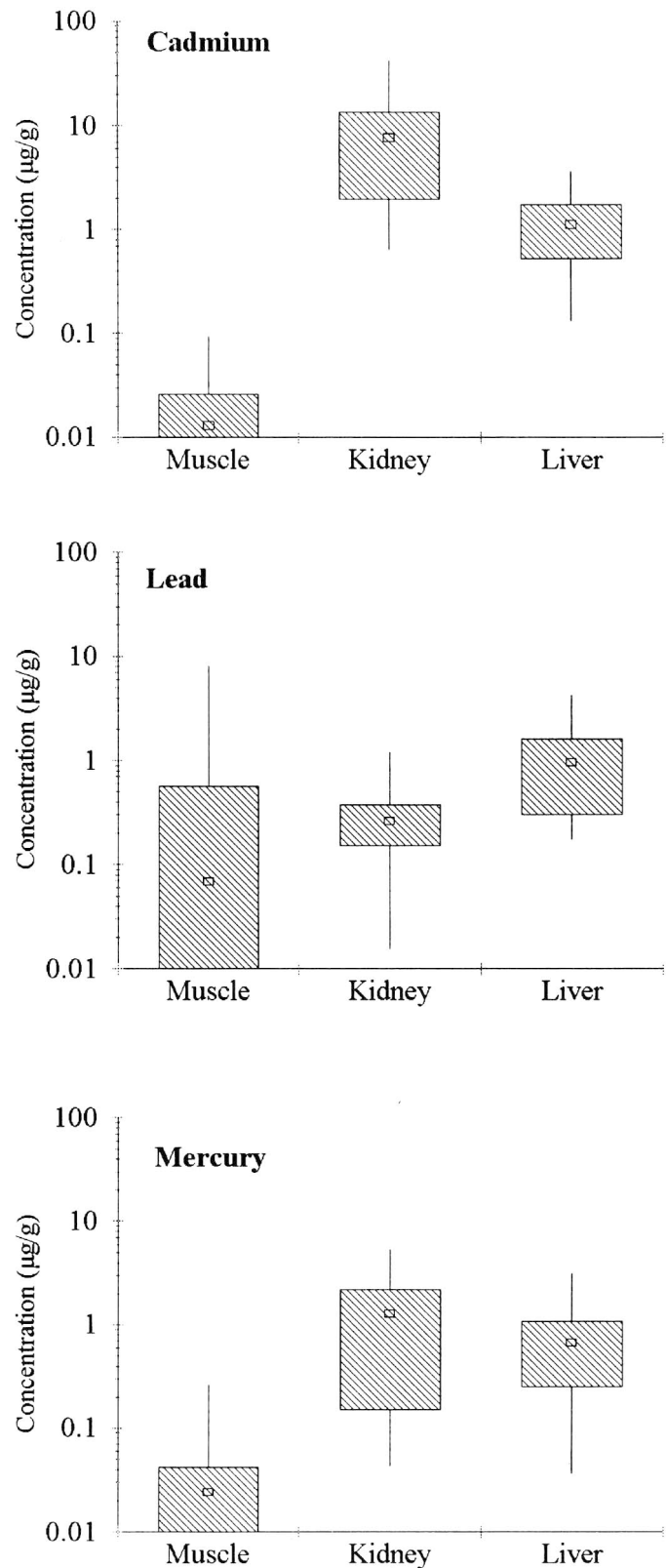


FIG. 2. Mean cadmium, lead, and mercury concentrations ( $\mu\text{g}\cdot\text{g}^{-1}$ , wet weight) in samples of caribou muscle ( $n = 293$ ), liver ( $n = 307$ ) and kidneys ( $n = 309$ ) collected between 1994 and 1996 in two regions of northern Québec. Concentration values are shown on a log scale. Boxes include mean values and range across two standard deviations. Lines connect minimum and maximum values.

TABLE 1. Mean (SD, n) cadmium, lead, and mercury concentrations ( $\mu\text{g} \cdot \text{g}^{-1}$ , wet weight) as a function of age (immature: 0–2 years; adult: 3 years and older) in samples of caribou muscle, liver, and kidney collected between 1994 and 1996 in two regions of northern Québec.

Tissue	Leaf River Region		George River-Tornat Mountains Region	
	Immature	Adult	Immature	Adult
<b>Cadmium</b>				
Muscle	0.013 (0.012,57)	0.017 (0.017,104)	0.013 (0.020,19)	0.008 (0.008,28)
Kidney	6.73 (4.41,86)	8.93 (6.24,177)	3.93 (2.80,19)	5.23 (5.12,27)
Liver	1.16 (0.64,84)	1.18 (0.61,176)	0.84 (0.48,19)	0.94 (0.62,28)
<b>Lead</b>				
Muscle	0.078 (0.21,57)	0.033 (0.16,104)	0.094 (0.32,19)	0.014 (0.017,28)
Kidney	0.27 (0.13,86)	0.28 (0.090,177)	0.15 (0.032,19)	0.20 (0.051,27)
Liver	1.06 (0.77,84)	0.89 (0.57,176)	1.19 (0.86,19)	0.89 (0.53,28)
<b>Mercury</b>				
Muscle	0.030 (0.033,57)	0.027 (0.012,104)	0.021 (0.008,19)	0.019 (0.008,28)
Kidney	1.37 (0.95,86)	1.39 (0.91,178)	0.62 (0.23,19)	0.56 (0.19,26)
Liver	0.70 (0.46,84)	0.70 (0.41,176)	0.45 (0.23,19)	0.38 (0.15,28)

between region and age and between age and sex proved nonsignificant.

$^{137}\text{Caesium}$  in muscle ranged from 51.0 to 154.8  $\text{Bq} \cdot \text{kg}^{-1}$ , with a mean value of  $94.7 \pm 28.0 \text{ Bq} \cdot \text{kg}^{-1}$ . We failed to detect an influence of region ( $p > 0.7$ ), time of year ( $p > 0.8$ ), or age ( $p > 0.3$ ).

## DISCUSSION

### *Tissue Metal Concentrations within Animals*

With respect to type of tissue, log levels were lowest in muscle and highest in kidneys (for Cd and total Hg) and liver (for Pb). A similar relative order of tissue concentrations has been reported in other recent studies (Braune et al., 1999; Jorhem, 1999; Medvedev, 1999).

### *Spatial Variation*

Regional differences occurred in log concentration of the three metals for most tissues, with the western region consistently showing higher values. Spatial variation in Cd concentrations has been documented on a much larger scale in the Canadian Arctic, with western herds of caribou showing higher levels than eastern herds (Gamberg and Scheuhammer, 1994; Elkin and Bethke, 1995; Braune et al., 1999). On a smaller scale, Crête et al. (1992) have shown that lichens in the western part of northern Québec exhibit higher Cd, Pb, and Hg concentrations than lichens in the middle-eastern regions. In recent years, caribou from the LR region have occupied the more contaminated northwestern regions, while caribou from the GRTM region have ranged mostly within the less contaminated eastern regions of northern Québec (Crête et al., 1992). The lichen biomass of the northwestern part is also twice that found in the eastern area. Therefore, the LR region probably provides a larger supply of more contaminated lichens to browsing caribou.

Results indicate that contaminant concentrations often vary from month to month and can interact with

region of origin. In particular, concentration of several metals decreased from December to May in the LR region, but increased during the same period in the GRTM region. Details about the foraging ecology and physiology of caribou during this period in the two regions would be needed to pinpoint the causes of these fluctuations. Such interactions indicate that the scope of differences across regions in general can vary as a function of time of year. This suggests a potential source of bias when comparing results from studies undertaken at different times of the year.

To facilitate comparison with our wet-weight concentrations, dry-weight concentration estimates available from the literature were transformed to a wet-weight basis using the mean water content of different organs in caribou from northern Québec (Crête et al., 1989). Mean Cd concentrations measured in kidney, liver, and muscle of caribou from the GRTM region were lower than those reported a few years earlier in the same region (liver: 1.2–2.9  $\mu\text{g} \cdot \text{g}^{-1}$ ; kidney: 11.7–40.7  $\mu\text{g} \cdot \text{g}^{-1}$ ; muscle: 0–0.20  $\mu\text{g} \cdot \text{g}^{-1}$ ; Crête et al., 1989). Mean Pb concentrations in liver were also lower than those reported in the earlier study from the GRTM region (1.1  $\mu\text{g} \cdot \text{g}^{-1}$ ; Crête et al., 1990). Mean Pb concentrations in our kidney samples were similar to those found in caribou from the Northwest Territories (0.20–2.7  $\mu\text{g} \cdot \text{g}^{-1}$ ), but concentrations in our liver samples were generally higher than those from caribou in the Northwest Territories (0.07–0.34  $\mu\text{g} \cdot \text{g}^{-1}$ ; Elkin and Bethke, 1995).

Total mercury liver concentrations from the LR region were smaller than earlier levels found in the same region (0.5  $\mu\text{g} \cdot \text{g}^{-1}$ ; Crête et al., 1990). Levels found in the GRTM regions must therefore have decreased to an even greater extent. Liver and kidney concentrations in our samples were generally similar to those found in caribou from the Northwest Territories (kidney: 0.41–2.32  $\mu\text{g} \cdot \text{g}^{-1}$ , liver: 0.12–0.66  $\mu\text{g} \cdot \text{g}^{-1}$ ; Elkin and Bethke, 1995).

### *Effect of Age, Sex, and Mass*

Not surprisingly, age was associated with kidney log Cd levels. In other cervids, older animals usually exhibit higher

TABLE 2. Linear models of log cadmium, lead, and mercury concentrations in different caribou tissues as a function of the region of origin, age, sex, month of collection, and body mass of the carcass.

Variable	Region <sup>1</sup>	Age <sup>2</sup>	Sex <sup>3</sup>	Month <sup>4</sup>	Mass <sup>4</sup>	Region*month <sup>4</sup>	Sex*month <sup>4</sup>
<b>Cadmium</b>							
Muscle	L > G < 0.0001	ns 0.35	ns 0.54	+ 0.0004	ns 0.32	ns > 0.05	ns > 0.05
Kidney	L > G < 0.0001	A > I 0.02	ns 0.12	± 0.0002	+ + 0.03	ns > 0.05	ns > 0.05
Liver	L > G 0.01	ns 0.66	F > M 0.05	± 0.0002	ns 0.40	ns > 0.05	ns > 0.05
<b>Lead</b>							
Muscle	ns 0.06	I > A 0.02	ns 0.31	ns 0.06	ns 0.37	ns > 0.05	ns > 0.05
Kidney	L > G < 0.0001	A > I 0.04	ns 0.82	- < 0.0001	- 0.03	L - G+ 0.03	ns > 0.05
Liver	ns 0.99	ns 0.15	ns 0.19	± 0.003	ns 0.17	ns > 0.05	ns > 0.05
<b>Mercury</b>							
Muscle	L > G 0.004	ns 0.27	ns 0.57	- 0.002	ns 0.37	L - G+ < 0.0001	ns > 0.05
Kidney	L > G < 0.0001	ns 0.56	ns 0.91	- < 0.0001	ns 0.41	L - G0 0.0008	M- F0 0.03
Liver	L > G < 0.0001	ns 0.97	ns 0.58	± < 0.0001	ns 0.66	ns > 0.05	ns > 0.05

<sup>1</sup> Region of origin: Leaf River (L), George River-Torngat Mountains (G).

<sup>2</sup> Age: Immature (I), Adult (A).

<sup>3</sup> Sex: Male (M), Female (F).

<sup>4</sup> Sign of effect: + = significant increase, - = significant decrease, 0 = no effect, ± = increase or decrease.

levels (Wolf et al., 1982; Froslic et al., 1986; Crête et al., 1989, 1990; Eriksson et al., 1990; Stansley et al., 1991; Rintala et al., 1995; Borch-Johnsen et al., 1996). Continued retention is known to induce a progressive accumulation of Cd in soft tissues, particularly in kidneys (Goyer, 1991). The effect of age was not documented for log total Hg.

Log Pb concentration increased with age in kidneys, but decreased with age in muscle. In most animal species, skeletal tissue is the main site of Pb accumulation following chronic exposure to low doses. Bone Pb concentrations tend to be higher in older individuals. When the level of bone Pb reaches a threshold value, Pb is then also deposited in other tissues, especially kidneys, where it has a low turnover rate (National Academy of Sciences, 1980). In humans, concentration in kidneys increases with age. The decrease with age in caribou muscle was therefore unexpected. As uptake and storage of Pb vary in many species with age, Pb kinetics may be influenced by physiological stress on immature, growing animals, in relation to the metabolism of calcium (Humphreys, 1991).

We documented few effects of sex. Sex-related differences in metal concentrations have been reported in other big game species (Khan and Forester, 1995; Crête et al., 1989). In caribou, kidney mass is known to fluctuate through time as a function of reproductive status. Kidney mass thus increases in males from the stressful rut period in early winter to the summer months (Crête et al., 1989). The reverse trend occurs in females, presumably as a response to the demands of gestation and lactation in the early summer months. Consequently, metal concentra-

tions in kidneys are expected to vary seasonally, in response to organ mass fluctuations, in the opposite direction for males and females. Data were collected from early winter to the beginning of summer in the present study. We documented only one interaction between sex and time of year: males showed lower values of log Hg in kidneys with time, while females retained similar levels. This result provides only partial support for the stress hypothesis. Females showed higher levels of Cd in liver than males, but data on seasonal mass fluctuations in liver are not available to assess the stress hypothesis for this organ. We documented few effects of body mass, presumably because body mass and age are correlated, and age is a better predictor of contaminant burden.

### <sup>137</sup>Caesium

Higher levels of <sup>137</sup>Cs in lichens have been documented in the middle-eastern part of northern Québec, a region associated with caribou from the GRTM region (Crête et al., 1992). However, spatial variation in lichen burden failed to influence <sup>137</sup>Cs levels in our limited muscle samples. Muscle samples, in general, contain the lowest levels of contaminants (e.g., Jorhem, 1999; Medvedev, 1999; this study). It remains to be seen whether the lack of spatial heterogeneity would also be documented in tissues such as liver and kidneys, which contain higher concentrations of contaminants.

Average concentrations in muscle samples were higher than levels found in various other regions of the Canadian

North (Macdonald et al., 1996). However, current levels in muscle are lower than those found in caribou from the GRTM region in 1986 and 1988 (Crête et al., 1990, 1993). The decrease in concentration continues to the present day (Braune et al., 1999) and probably follows the moratorium on atmospheric weapons testing reached in 1963 (Elkin and Bethke, 1995).

#### *Food Safety Issues and Caribou Health*

Cd levels exceeded the 1 ppm consumption tolerance threshold in nearly all kidney samples and in 45% of liver samples. Pb concentrations never exceeded the 2 ppm consumption threshold in the kidneys, but 6% of liver samples showed unacceptable levels. Total Hg concentrations exceeded the 0.5 ppm consumption threshold in 54% of liver samples and 81% of kidney samples. Thus current restrictions on the consumption of liver and kidneys of caribou from northern Québec should still be maintained. Apparent temporal changes in metal concentrations suggest that regular monitoring is important to provide realistic advice.

Concentrations of <sup>137</sup>Cs in samples of caribou muscle are known to follow an annual cycle; the highest levels are reached in winter and the lowest in summer (Crête et al., 1993; Macdonald et al., 1996). Values reported in the present study should thus be considered as upper estimates. The International Commission on Radiological Protection recommends that the public should not be exposed to more than 1 mSv of radiation from a regulated human activity in a given year (ICRP, 1991). Using data from the GRTM region, more than 275 muscle meals from caribou killed in winter must be consumed to achieve the annual dose of 1 mSv. Similarly, more than 600 muscle meals from caribou killed in summer would be needed to achieve the target value (Crête et al., 1993). The <sup>137</sup>Cs levels measured in the present study appear safe in view of the ICRP recommendations.

Caribou in the present study were selected by Inuit hunters, which probably biased sampling toward more desirable prey, such as large and vigorous animals. While the present results are representative of the hunted population of caribou, it remains to be seen to what extent these findings apply to the populations of northern Québec caribou as a whole.

Despite large individual variation in metal concentrations among sampled caribou, no particular ill-health events were reported at the abattoirs. If exposure levels led to clinical signs in certain animals, selective hunting would not likely present a valid picture at the herd level. Subclinical signs may have been present. However, neither NOELs (no observed effect levels) nor LOELs (lowest observed effect levels) are provided for cervids in the literature. Moreover, the toxicological significance of Cd residue levels in certain species, including cervids, needs to be evaluated (Stansley et al., 1991, Wren et al., 1995). Biomarkers of subclinical effect would be necessary in order to evaluate such a level of pathology. More studies are definitely needed.

Contaminants measured in this study biomagnify along the food chain and could have an impact at the ecosystem level, which remains to be investigated. However, in contrast to the levels found in aquatic ecosystems, bioaccumulation of Hg in terrestrial ecosystems is relatively small, as shown in the relatively low levels of Hg in tissues compared to those found in piscivorous mammals (Wren, 1986; Fortin et al., 2001). In addition, Native populations are exposed to more than one source of these contaminants through their traditional way of life.

#### ACKNOWLEDGEMENTS

The authors gratefully acknowledge veterinary inspectors at the abattoirs and Dr. Geneviève Rousseau, who supervised this project at the Ministère de l'Agriculture, des Pêcheries et de l'Alimentation du Québec. We also thank the personnel of Nunavik Arctic Food, who provided the samples, and the local hunters who participated in the study. We thank C.R. Macdonald and M. Laverock (AECL Research, Whiteshell Laboratories, Pinawa, Manitoba) for the <sup>137</sup>Cs analyses and the Makivik Research Center staff for their helpful collaboration. A traveling scholarship was provided by the Ministère des Affaires Indiennes et du Nord Canadien. We thank three anonymous referees for useful comments.

#### REFERENCES

- AASTRUP, P., RIGET, F., DIETZ, R., and ASMUND, G. 2000. Lead, zinc, cadmium, mercury, selenium and copper in Greenland caribou and reindeer (*Rangifer tarandus*). *Science of the Total Environment* 245:149–159.
- BARRIE, L.A., GREGOR, D., HARGRAVE, B., LAKE, R., MUIR, D., SHEARER, R., TRACEY, B., and BIDLEMAN, T. 1992. Arctic contaminants: Sources, occurrences and pathways. *Science of the Total Environment* 122:1–74.
- BORCH-JOHNSEN, B., NISSEN, K.J., and NORHEIM, G. 1996. Influence of season and diet on liver and kidney content of essential elements and heavy metals in Svalbard reindeer. *Biology of Trace Element Research* 51:235–247.
- BRAUNE, B., MUIR, D., DeMARCH, B., GAMBERG, M., POOLE, K., CURRIE, R., DODD, M., DUSCHENKO, W., EAMER, J., ELKIN, B., EVANS, M., GRUNDY, S., HEBERT, C., JOHNSTONE, R., KIDD, K., KOWNIG, B., LOCKHART, L., MARSHALL, H., REIMER, K., SANDERSON, J., and SHUTT, L. 1999. Spatial and temporal trends of contaminants in Canadian arctic freshwater and terrestrial ecosystems. *Science of the Total Environment* 230:145–207.
- CRÊTE, M., and DOUCET, G.J. 1998. Persistent suppression in dwarf birch after release from heavy summer grazing by caribou. *Arctic and Alpine Research* 30:126–132.
- CRÊTE, M., POTVIN, F., WALSH, P., BENEDETTI, J.L., LEFEBVRE, M.A., WEBER, J.P., PAILLARD, G., and GAGNON, J. 1987. Pattern of cadmium contamination in the liver and kidneys of moose and white-tailed deer in Quebec. *Science of the Total Environment* 66:45–53.

- CRÊTE, M., NAULT, R., WALSH, P., BENEDETTI, J.L., LEFEBVRE, M.A., WEBER, J.P., GAGNON, J. 1989. Variation in cadmium content of caribou tissues from northern Quebec. *Science of the Total Environment* 80:103–112.
- CRÊTE, M., LEFEBVRE, M.A., COOPER, M.B., MARSHALL, H., BENEDETTI, J.L., CARRIÈRE, P.E., and NAULT, R. 1990. Contaminations in caribou tissues from northern Quebec. *Rangifer, Special Issue #3*:289.
- CRÊTE, M., LEFEBVRE, M.A., ZIKOVSKY, L., and WALSH, P. 1992. Cadmium, lead, mercury and <sup>137</sup>caesium in fruticose lichens of northern Quebec. *Science of the Total Environment* 121:217–230.
- CRÊTE, M., LÉGARÉ, J.M., DAVELUY, A., and GAUTHIER, J. 1993. Concentrations saisonnières des radioéléments les plus importants (<sup>210</sup>Pb, <sup>210</sup>Po et <sup>137</sup>Cs) présents dans les tissus de caribous du nord québécois après l'accident de Tchernobyl. Report from the Ministère de l'Environnement et de la Faune du Québec. 29 p.
- ELKIN, B.T., and BETHKE, R.W. 1995. Environmental contaminants in caribou in the Northwest Territories, Canada. *Science of the Total Environment* 160/161:307–321.
- ERIKSSON, O., FRANK, A., NORKSVISK, M., and PETERSON, L.R. 1990. Heavy metals in reindeer and their forage plants. *Rangifer, Special Issue #3*:315–331.
- FORTIN, C., BEAUCHAMP, G., DANSEREAU, M., LARIVIÈRE, N., and BÉLANGER, D. 2001. Spatial variation in mercury concentrations in wild mink and river otter carcasses from the James Bay territory, Quebec, Canada. *Archives of Environmental Contamination and Toxicology* 40:121–127.
- FROSLIE, A., HAUGEN, A., HOLT, G., and NORHEIM, G. 1986. Levels of cadmium in liver and kidneys from Norwegian cervids. *Bulletin of Environmental Contamination and Toxicology* 37:453–460.
- GAMBERG, M., and SCHEUHAMMER, A.M. 1994. Cadmium in caribou and muskoxen from the Canadian Yukon and Northwest Territories. *Science of the Total Environment* 143:221–234.
- GAUTHIER, L., NAULT, R., and CRÊTE, M. 1989. Variations saisonnières du régime alimentaire des caribous du troupeau de la rivière George, Quebec nordique. *Naturaliste Canadien* 116:101–112.
- GOYER, R.A. 1991. Toxic effects of metal. In: Amdur, M.O., Doull, J., and Klaassen, C.D., eds. *Casarett and Doull's toxicology: The basic science of poisons*, 4th ed. Montréal: McGraw-Hill. 633–638.
- HANSON, W.G. 1967. Cesium-137 in Alaskan lichens, caribou, and Eskimos. *Health Physics* 13:383–389.
- HOLLEMAN, D.F., LUICK, J.R., and WHICKER, F.W. 1971. Transfer of radiocaesium from lichen to reindeer. *Health Physics* 21:657–666.
- HUMPHREYS, D.J. 1991. Effects of exposure to excessive quantities of lead on animals. *British Veterinary Journal* 147: 18–30.
- ICRP (INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION). 1990. Recommendations on the International Commission on radiological protection. ICRP Publication 60. New York: Pergamon Press.
- JORHEM, L. 1999. Lead and cadmium in tissues from horse, sheep, lamb and reindeer in Sweden. *Zeitschrift für Lebensmittel-Untersuchung und -Forschung A-Food Research & Technology* 208:106–109.
- KHAN, A.T., and FORESTER, D.M. 1995. Mercury in white-tailed deer forage in Russell plantation, Macon County, Alabama. *Veterinary Human Toxicology* 37:45–46.
- KIM, C., CHAN, H.M., and RECEVEUR, O. 1998. Risk assessment of cadmium exposure in Fort Resolution, Northwest Territories, Canada. *Food Additives Contamination* 15:307–317.
- LANGLOIS, C., and LANGIS, R. 1995. Presence of airborne contaminants in the wildlife of northern Quebec. *Science of the Total Environment* 160/161:391–402.
- LARTER, N.C., and NAGY, J.A. 2000. A comparison of heavy metal levels in the kidneys of High Arctic and mainland caribou populations in the Northwest Territories of Canada. *Science of the Total Environment* 246:109–119.
- MACDONALD, C.R., EWING, L.L., ELKIN, B.T., and WIEWEL, A.M. 1996. Regional variation in radionuclide concentrations and radiation dose in caribou (*Rangifer tarandus*) in the Canadian Arctic, 1992–1994. *Science of the Total Environment* 182:53–73.
- MEDVEDEV, N. 1995. Concentrations of cadmium, lead and sulphur in tissues of wild, forest reindeer from north-west Russia. *Environmental Pollution* 90:1–5.
- . 1999. Levels of heavy metals in Karelian wildlife, 1989–91. *Environmental Monitoring and Assessment* 56:177–193.
- NASH, T.H., and GRIES, C. 1995. The response of lichens to atmospheric deposition with an emphasis on the Arctic. *Science of the Total Environment* 161:737–747.
- NATIONAL ACADEMY OF SCIENCES. 1980. Mineral tolerance of domestic animals. Washington, D.C.: National Academy of Sciences.
- RAPPORT SANTÉ QUEBEC. 1992. Et la santé des Inuits, ça va? Rapport de l'enquête Santé Québec auprès des Inuits de Nunavik, Tome 3. Quebec. 179 p.
- RINTALA, R., VENALANINEN, E.R., and HIRVI, T. 1995. Heavy metals in muscle, liver, and kidney from Finnish reindeer in 1990–91 and 1991–1992. *Bulletin of Environmental Contamination and Toxicology* 54:158–165.
- SCHAEFER, J.A., and LUTTICH, S.N. 1998. Movements and activity of caribou, *Rangifer tarandus caribou*, of the Torngat Mountains, northern Labrador and Quebec. *Canadian-Field Naturalist* 112:486–490.
- STANLEY, W., ROSCOE, D.E., and HAZEN, R.E. 1991. Cadmium contamination of deer livers in New Jersey: Human health risk assessment. *Science of the Total Environment* 107: 71–82.
- TAYLOR, H.W., SVOBODA, J., HENRY, G.H.R., and WEIN, R.W. 1988. Post-Chernobyl <sup>134</sup>Cs and <sup>137</sup>Cs levels at some localities in northern Canada. *Arctic* 41:293–296.
- THOMAS, D.J., TRACEY, B., MARSHALL, H., and NORSTROM, R.J. 1992. Arctic terrestrial ecosystem contamination. *Science of the Total Environment* 122:135–164.



WOLF, A., SMITH, J.R., and SMALL, L. 1982. Metals in livers of white-tailed deer in Illinois. *Bulletin of Environmental Contamination and Toxicology* 28:189–194.

WREN, C.D. 1986. A review of metal accumulation and toxicity in wild mammals. I. Mercury. *Environmental Research* 40: 210–244.

WREN, C.D., HARRIS, S., and HATTRUO, N. 1995. Ecotoxicology of mercury and cadmium. In: Hoffman, D.J., Rattner, B.A., Burton, G.A., Jr., and Cairns, J., Jr., eds. *Handbook of ecotoxicology*. Boca Raton, Florida: Lewis Publisher. 392–423.