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Mercury and cadmium in ringed seals in the Canadian Arctic: Influence of location and diet

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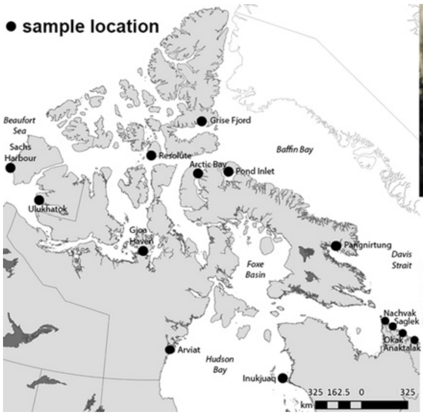
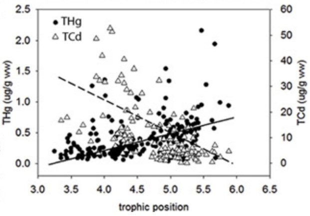
HIGHLIGHTS

- Diet and location influenced THg and Cd in ringed seals across the Canadian Arctic.
- Biomagnification processes contribute to elevated THg levels in the western Arctic.
- Consuming low-trophic position prey explains high Cd levels in the eastern Arctic.

GRAPHICAL ABSTRACT





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ABSTRACT

Concentrations of total mercury (THg) and total cadmium (TCd) were determined in muscle and liver of ringed seals (*Pusa hispida*) from up to 14 locations across the Canadian Arctic. Location, trophic position (TP) and relative carbon source best predicted the THg and TCd concentrations in ringed seals. THg concentrations in ringed seals were highest in the western Canadian Arctic (Beaufort Sea), whereas TCd was highest in the eastern Canadian Arctic (Hudson Bay and Labrador). A positive relationship between THg and TP and a negative relationship between THg and relative carbon source contributed to the geographical patterns observed and elevated THg levels at certain sites. In contrast, a negative relationship between TCd and TP was found, indicating that high TCd concentrations are related to seals feeding more on invertebrates than fish. Feeding ecology appears to play an important role in THg and TCd levels in ringed seals, with biomagnification driving elevated THg levels and a dependence on low-trophic position prey resulting in high TCd concentrations. The present study shows that both natural geological differences and diet variability among regions explain the spatial patterns for THg and TCd concentrations in ringed seals.

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1. Introduction

The presence of mercury (Hg) and cadmium (Cd) in the Arctic is from both natural and anthropogenic sources (Macdonald et al., 2000). Atmospheric, terrestrial, and oceanic pathways deliver Hg and Cd to Arctic marine waters, where they are taken up by algae and bacteria and transferred through the Arctic marine food web (Atwell et al., 1998; Campbell et al., 2005). Hg and Cd are known to bioaccumulate in the tissues of organisms (Morel et al., 1998; Woshner et al., 2001a), and Hg in the form of methyl mercury (MeHg), is known to biomagnify to relatively high concentrations in upper trophic level marine mammals (Braune et al., 2015). In contrast, Cd accumulation in marine mammals is driven more by dietary selection (Dehn et al., 2005). For example, Bowhead whales (*Balaena mysticetus*) feed at a low trophic position and yet have higher Cd concentrations than top-level Arctic consumers (e.g. polar bear (*Ursus maritimus*) and Arctic fox (*Alopex lagopus*)) (Ballard et al., 2003; Bratton et al., 1997; Woshner et al., 2001a; Woshner et al., 2001b). The investigation of total mercury (THg) and total cadmium (TCd) levels in fish, birds and marine mammals in the Arctic has been of concern for decades due to their dietary and cultural importance for Inuit and because of reports of elevated levels of these contaminants in some organisms (Braune et al., 2015; Gaden et al., 2009). Furthermore, it has been shown that elevated levels of THg and TCd can have adverse health effects (e.g. neurotoxicity, immunotoxicity, liver and kidney lesions) on marine mammals (AMAP, 2005; Basu et al., 2009; Frouin et al., 2012; Rawson et al., 1993).

The ringed seal (*Pusa hispida*) was chosen as sentinel species for monitoring contaminants in the Arctic because it is the most abundant Arctic pinniped, has a circumpolar distribution, plays an important role in the Arctic marine food web, and is an important part of the Inuit diet (Fisk et al., 2002; Laird et al., 2013; Rigét et al., 2005). Ringed seals typically feed on a variety of fish, amphipods, euphosiids, mysids, shrimp, bivalves and cephalopods (Holst et al., 2001; Lowry et al., 1980; McLaren, 1958; Smith, 1987). Spatial differences have been detected in the diet of ringed seals (Yurkowski et al., 2015a) and some studies have shown diet variability due to age, sex, and season (Holst et al., 2001; Lowry et al., 1980; Thiemann et al., 2007). Contaminant levels among ringed seals can differ due to differences in trophic positions and foraging strategies, along with other biological factors (Dehn et al., 2005).

Elevated THg and TCd concentrations were first reported in ringed seal liver and kidney in the Canadian Arctic in the 1970s (Muir et al., 1992; Smith and Armstrong, 1975; Wagemann and Muir, 1984). THg concentrations increased from the early-1970s to mid-1980s, and continued to increase to the mid- and late-1990s (AMAP, 2005; Wagemann et al., 1996). While present day THg levels in ringed seals exceed historical concentrations, no significant changes have been reported from 1999 to 2009 (Braune et al., 2015). A lack of data in the Canadian Arctic from the early 1970s onwards has precluded a temporal comparison for TCd in ringed seals (Muir et al., 1999; Wagemann et al., 1996). In Greenland, however, temporal comparisons have been reported and show an increase in TCd concentrations in ringed seal liver from the late 1970s to the mid-1980s, and then a decrease from the mid-1980s to the mid-1990s (Rigét and Dietz, 2000). The difference in temporal trends from the mid-1980s to mid-1990s for THg and TCd concentrations in ringed seals are thought to reflect changes in diet rather than a change in environmental levels (Rigét and Dietz, 2000).

Stable isotopes of nitrogen (δ^{15}N) and carbon (δ^{13}C) have been used to understand diet and interpret contaminant trends in marine species. Based on relative isotopic fractionation processes δ^{15}N can be used to describe trophic level and δ^{13}C can be used to infer food web carbon sources. For example, enrichment of δ^{15}N ratios increases with trophic position in marine food webs providing a continuous variable with which to assess both trophic level and food web transfer of contaminants (Fisk et al., 2001). Carbon isotope ratios can evaluate the relative contributions of inshore/benthic versus offshore/pelagic feeding preferences (France and Peters, 1997), and can provide insights in contaminant exposure (Fisk et al., 2003a; Fisk et al., 2002).

Previous spatial studies observed that THg levels were higher in ringed seals from the western Canadian Arctic compared with the eastern Canadian Arctic, whereas TCd concentrations increased from west to east (Muir et al., 1992; Rigét et al., 2005). The contrasting spatial pattern of THg and TCd concentrations across the Canadian Arctic observed by some has been attributed strictly to geological influences, but these study designs did not enable an evaluation of the potential role that diet played in these geographic patterns. The objectives of the present study were (i) to assess spatial patterns across the Canadian Arctic in THg and TCd concentrations in liver and muscle tissue of ringed seals, and (ii) to evaluate the influence of diet and location on THg and TCd concentrations. While earlier studies (Dehn et al., 2005; Rigét et al., 2005; Wagemann et al., 1996) incorporated several of the same sites as the present study, this is the most comprehensive survey of THg and TCd in ringed seals from across the Canadian Arctic.

2. Materials and methods

2.1. Samples collection

Muscle and liver samples were collected from 14 and 12 locations, respectively in the Canadian Arctic for a total of 506 (282 sub adults; 224 adults) ringed seals (Tables 1 and 2). Samples were collected in the summer months between 2007 and 2011. All samples were collected from local subsistence hunts. Sex, length, girth, and blubber thickness were determined in the field (Tables 1 and 2). Ages were determined by Matson's Laboratory, USA, by longitudinally thin sectioning a lower canine tooth and counting annual growth layers in the cementum using a compound microscope and transmitted light. All samples were stored at −20 °C until analyzed for stable isotopes (δ^{15}N and δ^{13}C), THg, and TCd within 1 year of sample collection. For all samples collected, appropriate permits and community approval were obtained from local government agencies and Department of Fisheries and Oceans Canada.

2.2. Chemical analysis

Seal muscle tissue (0.2 g wet) was analyzed for THg using a Direct Mercury Analyzer (DMA; Milestone Inc., Shelton, CT, USA). TCd and THg concentrations in liver were determined by multielement analyses following Environment Canada, National Laboratory for Environmental Testing (NLET) inorganics laboratory procedures (Environment Canada, 2015). Briefly, subsampled liver (0.5 g) was digested in a 120 mL TFM digestion vessel using an 8:1 ratio of nitric acid and hydrogen peroxide. Cd was determined using inductively coupled plasma mass spectrometry while THg was determined by cold vapor-atomic absorption spectroscopy. Quality assurance steps included the analysis of reference materials for THg and TCd and reagent blanks with each batch of samples.

Duplicate sample runs ($n = 4$) had coefficients of variation ranging from 4.6% to 19.4%. Seven standard reference materials (SRM; DORM-2, DOLT-3, NIST-1566B, 2976 mussel, DOLT-2, TORT-2) were run for THg and 95.9 ± 2.11% ($n = 97$) of the standard reference material was recovered. Three standard reference materials (DOLT-2, DOLT-4, TORT-2) were run for TCd and 100.0 ± 3.21 ($n = 14$) of the standard reference material was recovered.

2.3. Stable isotope analysis and trophic position and relative carbon source calculations

Muscle tissue was freeze-dried and homogenized. Lipid was extracted from samples from Nachvak, Saglek, Okak and Anaktalak by agitating the dried powdered muscle tissue in a 2:1 chloroform–methanol

Table 1

Mean (±SD) morphometric data and THg and TCd concentrations (μg/g wet wt) for sub adult (<6 year; male and female combined) ringed seals collected from the Canadian Arctic. Locations are ordered according to longitude from west to east. n.a., not available.

Location	Age (range)	Year	*n*	Length cm	Girth cm	Blubber thickness cm	THg muscle μg/g ww	THg liver μg/g ww	TCd liver μg/g ww
Sachs Harbour	0.2–3	2007–11	31	90 ± 2.8	65 ± 2.1	3 ± 0.1	0.507 ± 0.054	10.1 ± 4.79	2.37 ± 0.513
Ulukhaktok	1–5	2010	4	98 ± 3.8	84 ± 3.8	2.1 ± 0.1	0.393 ± 0.038	12.6 ± 2.56	5.61 ± 2.90
Gjoa Haven	0.2–5	2008–09	12	115 ± 8.2	81 ± 5.4	3.5 ± 0.3	0.254 ± 0.094	2.89 ± 0.982	3.10 ± 2.52
Resolute	0.2–5	2007–11	61	113 ± 3.0	90 ± 3.0	6.6 ± 2.4	0.457 ± 0.043	5.89 ± 1.34	3.89 ± 0.609
Arviat	0.2–5	2007–11	58	103 ± 2.8	81 ± 1.9	4.2 ± 0.2	0.377 ± 0.070	8.94 ± 1.33	13.8 ± 1.64
Inukjuaq	0.2–5	2007	13	94 ± 3.7	70 ± 4.0	3.3 ± 0.3	0.118 ± 0.017	1.35 ± 0.293	13.8 ± 5.87
Arctic Bay	0.2–3	2009	9	115 ± 7.7	82 ± 5.0	4.0 ± 0.3	0.206 ± 0.028	1.93 ± 0.854	2.75 ± 1.08
Grise Fjord	0.2–3	2008	13	104 ± 4.0	86 ± 4.1	4.0 ± 0.3	0.157 ± 0.034	2.74 ± 1.10	3.25 ± 1.28
Pond Inlet	0.2–4	2009	12	98 ± 3.4	71 ± 4.4	5.9 ± 2.3	0.203 ± 0.030	5.81 ± 2.33	4.11 ± 1.48
Pangnirtung	0.2–4	2011	14	117 ± 3.1	94 ± 5.9	n.a.	0.256 ± 0.030	4.73 ± 0.811	7.38 ± 1.20
Nachvak Fjord	0.2–4	2008–11	6	107 ± 8.0	89 ± 5.6	3.9 ± 0.5	0.155 ± 0.028	n.a.	8.64 ± 1.20
Saglek Fjord	0.2–4	2008–09	16	104 ± 5.8	82 ± 7.8	3.1 ± 0.3	0.150 ± 0.021	n.a.	n.a.
Okak Bay	0.2–3	2008–09	14	94 ± 2.6	81 ± 1.9	4.2 ± 0.2	0.134 ± 0.012	n.a.	n.a.
Anaktalak Bay	0.2–2	2008–09	7	100 ± 4.0	80 ± 3.9	3.5 ± 0.4	0.100 ± 0.016	n.a.	2.76 ± 0.82

solution for 24 h. The tissue and solvent were then filtered, and the resulting residue-filter paper dried at 60 °C for 48 h to evaporate the remaining solvent. Next, 500 μg of lipid-extracted tissue was weighted into tin capsules, and stable carbon and nitrogen isotope ratios were analyzed by continuous flow ion ratio mass spectrometer (Finnigan MAT Delta^{Plus}; Thermo Finnigan). Muscle samples from all remaining locations had a C:N < 3.5 and subsequently were not pre-extracted to remove lipid (Post et al., 2007). Further, lipid extraction of ringed seal muscle following the method outlined in Yurkowski et al. (2015b) has a very minor influence that is within the measurement error of the machine. Stable isotope abundances are expressed in delta (δ) values as the deviation from standards in parts per thousand (‰) using the equation

$$\delta_{sample}‰ = \left[\left(R_{sample}/R_{standard}\right)-1\right] \times 1000 \tag{1}$$

where R is the ratio of heavy to light isotope ($^{15}N/^{14}N$ or $^{13}C/^{12}C$) in the sample and standard. The nitrogen and carbon stable isotope standards were atmospheric nitrogen and Pee Dee Belemnite limestone formation, respectively. Estimates of precision, based on 2 standards (bovine muscle [NIST 8414] and an internal laboratory standard [tilapia fish muscle], $n = 209$ for each), were <0.17‰ and <0.09‰ for $\delta^{15}N$ and $\delta^{13}C$, respectively. Accuracy of isotope analysis, based on NIST standards sucrose (NIST 8542) and ammonia sulfate (NIST 8547) analyzed during the present study ($n = 3$ for each) were within <0.1‰ of certified $\delta^{15}N$ and $\delta^{13}C$ values.

Trophic position (TP) relative to the copepod *Calanus hyperboreus*, which we assumed occupied trohpic level 2 (i.e. primary herbivore), were determined using equations modified from Post (2002).

$$TP_{CONSUMER} = TP_{BASELINE} + \frac{\delta^{15}N_{CONSUMER} - \delta^{15}N_{BASELINE}}{\Delta^{15}N} \tag{2}$$

Diet-tissue discrimination factors ($\Delta^{15}N$) have not been reported for ringed seals, so we used previously published values of 2.4‰ for $\delta^{15}N$ in harp seal muscle, respectively fed a high-protein diet ($\delta^{15}N = 13.0‰$; $\delta^{13}C = -20.3‰$) (Hobson et al., 1996). Under a scaled trophic level framework in the Canadian Arctic, Hussey et al. (2014) estimated a $\Delta^{15}N$ of 2.1‰ for species at a TP of 4, which agrees with our TP approach for ringed seals. The mean $\delta^{15}N$ value for *C. hyperboreus* for the study locations was obtained from Loseto et al. (2008a) for Ulukhatok (9.4‰), from Chambellant et al. (2013) for Chesterfield Inlet (10.6‰), from Morris, 2015 for Gjoa Haven (8.8‰), from Hobson and Welch (1992) for Resolute (9.2‰), from McMeans et al. (2015) for Pangnirtung (9.9‰) and from Brown et al. (2010) for Saglek Bay (9.8‰). For the rest of the locations, mean values were applied as follows based on geographic proximity: 1) the mean value in Resolute was applied to Arctic Bay, Grise Fjord, and Pond Inlet, 2) the mean value in Ulukhaktok was applied to Sachs Harbour, 3) the mean value in Chesterfield Inlet was applied to Arviat and Inukjuaq, and 4) the mean value for Saglek Bay was appled to Nachvak, Okak, and Anaktalak.

$\delta^{13}C$ values for individual ringed seals relative to the copepod *C. hyperboreus* (baseline organism) was calculated to correct for

Table 2

Mean (±SD) morphometric data and THg and TCd concentrations (μg/g wet wt) for adult (≥6 year; male and female combined) ringed seals collected from the Canadian Arctic. Locations are ordered according to longitude from west to east. n.a., not available.

Location	Age (range)	Year	*n*	Length (cm)	Girth (cm)	Blubber thickness (cm)	THg muscle μg/g ww	THg liver μg/g ww	TCD liver μg/g ww
Sachs Harbour	6–30	2007–11	9	127 ± 2.4	90 ± 2.4	3.3 ± 0.5	1.07 ± 0.222	70.4 ± 34.7	4.31 ± 0.788
Ulukhaktok	6–37	2010	16	121 ± 1.6	88 ± 2.0	2.0 ± 0.1	0.558 ± 0.034	23.9 ± 3.38	6.24 ± 0.850
Gjoa Haven	6–27	2008–09	9	146 ± 7.4	96 ± 1.9	5.2 ± 0.6	0.380 ± 0.073	10.6 ± 1.97	1.97 ± 0.717
Resolute	6–42	2007–11	33	137 ± 2.2	104 ± 3.7	4.2 ± 0.2	0.573 ± 0.038	23.2 ± 7.76	6.91 ± 0.979
Arviat	6–33	2007–11	66	116 ± 1.2	96 ± 1.9	5.0 ± 0.2	0.318 ± 0.032	23.2 ± 2.89	21.5 ± 2.68
Inukjuaq	11–21	2007	5	124 ± 2.6	92 ± 2.0	3.0 ± 0.3	0.162 ± 0.055	6.08 ± 2.38	13.1 ± 2.49
Arctic Bay	8–38	2009	9	129 ± 9.4	99 ± 9.0	3.4 ± 0.6	0.409 ± 0.056	8.69 ± 2.06	4.86 ± 1.24
Grise Fjord	6–30	2008	7	144 ± 7.2	108 ± 4.8	3.7 ± 0.5	0.107 ± 0.021	19.0 ± 3.46	6.76 ± 3.01
Pond Inlet	7–32	2009	4	130 ± 8.2	95 ± 2.2	3.8 ± 0.5	0.533 ± 0.133	10.8 ± 3.37	9.06 ± 2.48
Nachvak Fjord	6–25	2008–09	19	126 ± 2.4	109 ± 3.9	5.5 ± 0.2	0.176 ± 0.027	n.a.	17.6 ± 2.83
Saglek Fjord	8–28	2008	12	117 ± 1.8	100 ± 9.4	5.2 ± 0.5	0.191 ± 0.013	n.a.	n.a.
Okak Bay	6–23	2008–09	14	123 ± 2.2	105 ± 2.0	5.7 ± 0.5	0.291 ± 0.040	n.a.	n.a.
Anaktalak Bay	7–15	2008	4	129 ± 4.8	108 ± 2.8	5.8 ± 0.4	0.220 ± 0.030	n.a.	14.3 ± 3.94

differences across locations and was determined using the equation modified from Fisk et al. (2003b).

$$\text{Relative carbon source} = \frac{\delta^{13}C_{\text{CONSUMER}}}{\delta^{13}C_{\text{BASELINE}}} \tag{3}$$

C. hyperboreus is a pelagic-filter feeding herbivore that has a pelagic δ^{13}C signal, values close to 1 or below are pelagic and values greater than 1 represent a more benthic carbon source. The mean δ^{13}C value for *C. hyperboreus* for the study locations was obtained from Loseto et al. (2008a) for Ulukhatok (−25.6‰), from Morris, 2015 for Gjoa Haven (−28.4‰), from Chambellant et al. (2013) for Chesterfield Inlet (−20.5‰), from Hobson and Welch (1992) for Resolute (−20.4‰), from McMeans et al. (2015) for Pangnirtung (−20.4‰) and from Brown et al. (2010) for Saglek Bay (−20.1‰). For the rest of the locations, mean values were applied as follows based on geographic proximity: 1) the mean value in Resolute was applied to Arctic Bay, Grise Fjord, and Pond Inlet, 2) the mean value in Ulukhaktok was applied to Sachs Harbour, 3) the mean value in Chesterfield Inlet was applied to Arviat and Inukjuaq, and 4) the mean value for Saglek Bay was applied to Nachvak, Okak, and Anaktalak. See Table 3 for mean δ^{15}N, TP, δ^{13}C and relative carbon source values for sub adult and adult ringed seals for each location.

2.4. Data analysis

THg concentrations in phocids increase with age (Dietz et al., 1996; Nyman et al., 2002; Rigét et al., 2005). To control for these confounding factors, we separated the data into two groups for statistical exploration: sub adults (<6 year, male and females combined) and adult (≥6 year, male and females combined). Seals 6 year and older were considered adults (McLaren, 1958; Smith, 1987; Smith et al., 1973) and those under 6 year were considered sub adults. Before analyzing the data separately, a one-way analysis of variance (ANOVA) confirmed differences ($p < 0.001$; Table S1) between sub adult and adult seals in mean THg and TCd concentrations in muscle and liver tissues. Data for males and females were combined since THg and TCd concentrations in ringed seal tissues are not influenced by sex (Dehn et al., 2005). Further, in the present study no differences ($p > 0.05$; Table S2) in THg and TCd concentrations were found between sexes for sub adult and adult ringed seals. In order to ensure that spatial trends were not confounded by inter-annual variation (2007 to 2011) of THg or TCd concentrations within seal populations, a one-way ANOVA using year was used to test for differences in muscle and liver for each location. Where inter-annual differences were detected using the ANOVA, a Tukey's HSD post hoc test was performed to identify the year that differed. With the exception of THg in muscle in seals ($n = 29$) from the four Labrador fiords (Nachvak, Saglek, Okak, and Anaktalak) in 2010 (which were excluded from further analysis), there existed no inter-annual differences in either sub adult or adult seals ($p > 0.05$; Table S3 and Table S4) and data across years were combined.

Univariate statistical analyses were performed in SPSS 22.0 for Windows. Data were log transformed when necessary to meet the normality assumptions for parametric analyses. One-way ANOVA using location was used to test for differences in mean THg and TCd concentrations in muscle and liver. Tukey's post hoc test was performed to test for differences in mean concentrations among locations.

The best variable or combination of variables (location, length, girth, blubber thickness, age, TP and relative carbon source) to describe THg and TCd was selected using the lowest Akaike information criteria (AIC) (SYSTAT Version 13, Systat Software Inc., San Jose, CA, USA). The Akaike differences (Δi) and normalized Akaike weights (w_i) were calculated to select the best variable or variables. The models with a ΔAIC of zero and up to two were considered to have the most support (Ferguson et al., 2006). Linear regression analysis was used to further assess the relationships between THg and TCd and TP and relative carbon source.

3. Results and discussion

3.1. THg in ringed seal muscle and liver

The average for condition indices (length, girth, blubber thickness) and the average concentrations of THg measured in ringed seal muscle and liver in the present study (Tables 1 and 2) generally fell within the range reported previously for ringed seals in the Canadian Arctic (Muir et al., 1999; Muir et al., 2000; Rigét et al., 2005; Wagemann et al., 1996). On average, THg concentrations in seal muscle were 37-fold lower than in liver for tissues collected from the same location. Similar ratios in ringed seals have been reported elsewhere in the Arctic for liver and muscle (Braune et al., 2015; Dehn et al., 2005; Wagemann et al., 1996).

As expected THg concentrations in muscle and liver were higher ($p < 0.001$; Table S5) in adults than in sub adults for most (69% and 83%, respectively) of the locations sampled across the Arctic (Fig. 1, Tables 1 and 2). This observation is consistent with previous findings which showed THg concentrations in liver and kidney tissues in ringed seals being higher in adults than sub adults across the Arctic (Rigét et al., 2005). No differences ($p \geq 0.05$; Table S5) were found between adults and sub adults in muscle tissue for Arviat, Grise Fjord, Inukjuak, and

Table 3
Mean δ^{15}N, trophic position (TP), δ^{13}C and relative carbon source values for sub adult and adult ringed seals. n.a., not available. Relative carbon source values close to 1 or below are pelagic and values greater than 1 represent a more benthic carbon source.

Location	δ^{15}N[a]		TP		δ^{13}C[a]		Relative carbon source	
	Sub adult	Adult	Sub adult	Adult	Sub adult	Adult	Sub adult	Adult
Sachs Harbour	16.9 ± 0.1	17.3 ± 0.2	5.1	5.3	−21.1 ± 0.2	−20.6 ± 0.2	0.82	0.81
Ulukhaktok	15.9 ± 0.3	16.9 ± 0.1	4.2	5.1	−21.3 ± 0.2	−20.9 ± 0.1	0.83	0.82
Gjoa Haven	17.5 ± 0.3	16.6 ± 0.2	5.6	5.2	−22.2 ± 0.3	−22.3 ± 0.4	0.78	0.78
Resolute	16.4 ± 0.2	16.9 ± 0.1	5.0	5.2	−19.1 ± 0.1	−18.7 ± 0.1	0.94	0.92
Arviat	15.7 ± 0.2	16.2 ± 0.1	4.1	4.3	−20.5 ± 0.1	−20.5 ± 0.2	1.00	1.00
Inukjuaq	14.2 ± 0.2	14.4 ± 0.2	3.5	3.6	−20.1 ± 0.2	−20.0 ± 0.2	0.98	0.97
Arctic Bay	16.4 ± 0.3	16.6 ± 0.2	5.0	5.1	−18.9 ± 0.2	−18.6 ± 0.1	0.93	0.91
Grise Fjord	15.9 ± 0.3	16.3 ± 0.3	4.8	5.0	−18.9 ± 0.2	−18.4 ± 0.1	0.93	0.90
Pond Inlet	16.5 ± 0.3	16.5 ± 0.4	5.0	5.0	−18.8 ± 0.1	−18.7 ± 0.2	0.92	0.92
Nachvak Fjord	13.5 ± 0.3	14.8 ± 0.2	3.5	4.1	−18.1 ± 0.2	−18.0 ± 0.1	0.90	0.89
Saglek Fjord	15.1 ± 0.6	14.7 ± 0.2	4.2	3.7	−15.0 ± 2.5	−18.1 ± 0.1	0.74	0.85
Okak Bay	13.4 ± 0.1	13.9 ± 0.2	3.5	3.7	−18.5 ± 01	−17.9 ± 0.1	0.92	0.89
Anaktalak Bay	14.0 ± 0.2	15.2 ± 0.8	3.7	4.3	−19.1 ± 0.1	−18.1 ± 0.5	0.95	0.90
Pangnirtung	15.6 ± 0.1	n.a.	4.4	n.a.	−19.8 ± 0.2	n.a.	0.97	n.a.

[a] Values are mean ± SE.

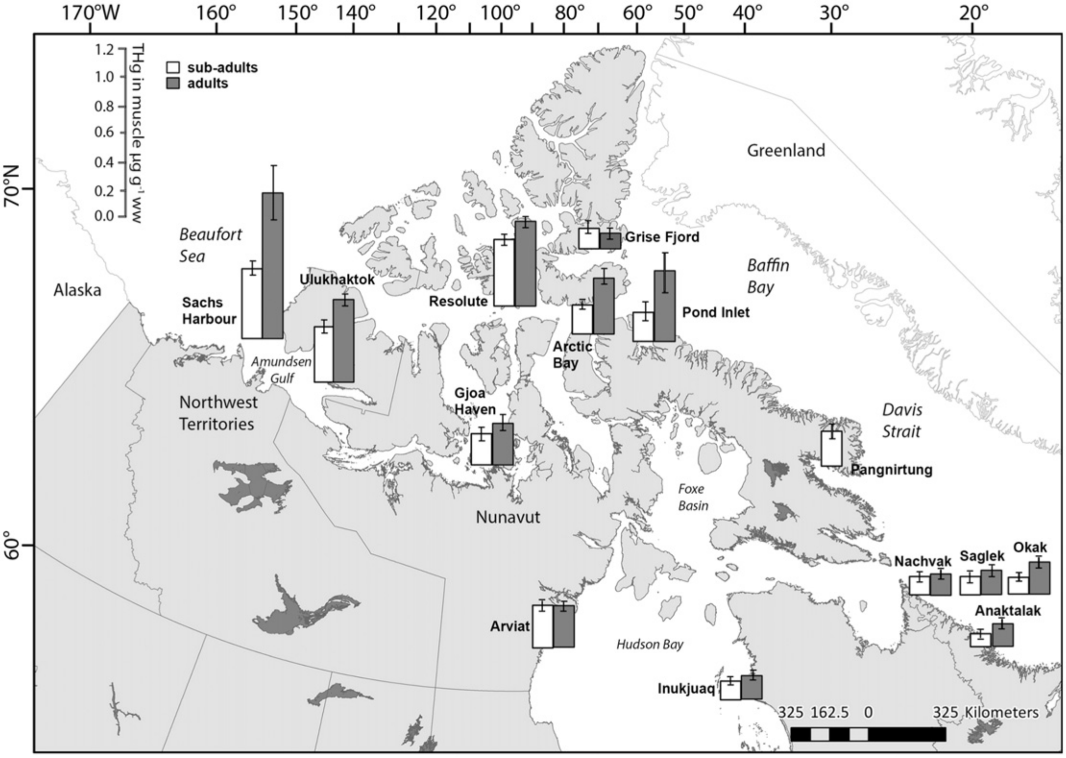

Fig. 1. Average THg concentrations ± SD in sub adult and adult ringed seal muscle from 14 communities across the Canadian Arctic sampled between 2007 and 2011.

Nachvak. No differences ($p \geq 0.05$; Table S5) were found between adults and sub adults in liver tissue for Pond Inlet and Ulukhaktok.

Concentrations of THg in muscle and liver varied by location ($p < 0.001$; Table S6) for both sub adults and adults. The highest concentrations of THg in muscle and liver for both sub adult and adult ringed seals was observed in Sachs Harbour and Ulukhaktok (Fig. 1, Tables 1 and 2). Elevated THg concentrations in muscle and liver were also observed in sub adult and adult Resolute seals, adult Pond Inlet seals and in sub adult and adult (liver only) Arviat seals (Tables 1 and 2; Fig. 1). Elevated levels observed in ringed seals sampled from the southern Beaufort Sea region (Sachs Harbour and Ulukhaktok) is consistent with previous studies which showed THg concentrations to be 2–3 fold higher in ringed seals sampled from the Beaufort Sea compared with the eastern Canadian Arctic (Braune et al., 2015; Gaden et al., 2009; Muir et al., 1992; Rigét et al., 2005; Wagemann et al., 1996). Our results are also consistent with higher relative THg concentrations in western Canadian Arctic polar bears, beluga whales (*Delphinapterus leucas*), and narwhals (*Monodon monoceros*) (Braune et al., 2015; Rigét et al., 2005; St. Louis et al., 2011).

The higher THg concentrations in seals from the Beaufort Sea region may be partly due to a higher natural geological background relative to other areas and the influence of the Mackenzie River (Leitch et al., 2007). Wagemann et al. (1995) described the geological settings in the western Arctic to have higher environmental background concentrations of Hg relative to the eastern Arctic. For example, the concentration range of Hg in nearshore surficial sediments in the western Arctic is 68–243 ng/g dry wt. (Thomas et al., 1982) and 40–60 ng/g dry wt. in the eastern Arctic (Loring, 1984). In Arctic marine waters methylated Hg concentrations were about two- to three-fold greater at the chlorophyll *a* maximum and oxycline in the Beaufort Sea than in the Canadian Arctic Archipelago (Kirk et al., 2012). Further, in the rocks the range of mercury is approximately 10-fold greater in the western Arctic than in the eastern Arctic (Cannon et al., 1978; Wedephol et al., 1978).

Natural geological variation, however, does not appear to explain the relatively high levels of THg in seals from Resolute and Arviat, and adult seals in Pond Inlet. Braune et al. (2015) reviewed earlier THg data (2000–2008), also reporting relatively high levels in ringed seals sampled in Resolute and Arviat. These authors (2015) were unable to attribute these findings to a particular factor, but one possible explanation could be that seals are feeding at a higher trophic level in these locations compared with those from other areas.

The lowest concentrations of THg in muscle were observed in Anaktalak and Inukjuaq for sub adult ringed seals and in Grise Fjord and Inukjuaq for adult seals (Tables 1 and 2). This observation is consistent with a previous study which showed the lowest THg concentrations in muscle in adult ringed seals sampled from Grise Fjord and Inukjuaq between 1999 and 2008 (Braune et al., 2015). The lowest concentrations of THg in liver were observed in Inukjuaq and Arctic Bay for sub adult and adult ringed seals (Tables 1 and 2). While univariate statistical approaches in the present study provided evidence for location influencing THg levels in muscle and liver, best-fit models using AIC (Table 4) confirmed that location, which is likely related to natural geological differences, explains the variation in THg levels across the Arctic. In addition, TP and relative carbon source contributed to the final model for sub adult and adult ringed seals (Table 4). These statistical results build on the observations above, which indicate that feeding ecology plays an important role in influencing THg levels in ringed seals across the Arctic.

3.2. TCd in ringed seal liver

The average concentrations of TCd found in ringed seal liver in the present study (Tables 1 and 2) generally fell within the range reported previously for ringed seals in the Canadian Arctic (Dehn et al., 2005; Rigét et al., 2005). TCd concentrations were higher ($p < 0.05$; Table S6) in adults than in sub adults for most (67%) of the locations sampled across the Arctic (Fig. 2, Tables 1 and 2). This observation is consistent with previous findings which showed TCd concentrations in liver and kidney tissues in ringed seals being higher in adults than sub adults across the Arctic (Rigét et al., 2005). No differences ($p \geq 0.05$;

Table 4
Akaike information criterion (AIC) in combination with backwards stepwise regression confirmed that location, trophic position and relative carbon source (R-C13) were the best variables to explain the variations of THg muscle and liver levels and TCd liver levels in ringed seals from across the Canadian Arctic.

Element	Age class	Tissue	Predictors	r^2	*p*-Value	AIC	AIC_c	ΔAIC_c	w_i
THg	Sub adults	Muscle	Location, TP	0.36	0.000	202.1	202.4	0	0.36
			Location, TP, R-C13	0.37	0.000	202.4	202.9	0.49	0.28
THg		Liver	Location	0.30	0.000	1232.3	1232.6	0	0.50
			Location, R-C13, age	0.31	0.001	1233.3	1233.7	1.38	0.28
TCd		Liver	Location, TP	0.55	0.000	1106.2	1106.4	0	0.41
			Location, TP, R-C13	0.56	0.000	1106.5	1106.9	0.46	0.32
THg	Adults	Muscle	Location, TP	0.35	0.000	0.179	0.507	0	0.45
			Location, TP, R-C13	0.36	0.000	0.360	0.821	0.31	0.38
THg		Liver	TP	0.30	0.03	170.2	170.4	0	0.41
			TP, location	0.32	0.03	170.3	170.6	0.19	0.37
TCd		Liver	R-C13, TP	0.42	0.000	123.8	124.4	0	0.45
			R-C13, TP, location	0.64	0.000	124.4	125.3	0.83	0.30

Table S7) were found between adults and sub adults for Gjoa Haven, Inukjuaq and Ulukhaktok.

Concentrations of TCd in liver varied by location ($p < 0.001$; Table S6) for both sub adults and adults. The highest concentrations of TCd in liver for sub adults was in Hudson Bay (Arviat and Inukjuaq) and in Nachvak Fjord (Fig. 2, Tables 1 and 2). The highest concentrations of TCd in liver for adults was in Hudson Bay (Arviat and Inukjuaq) and in the two northern Labrador fjords (Nachvak and Anaktalak). Previous studies have reported that invertebrate prey species are associated with higher levels of TCd than fish (Bustamante et al., 2003; Bustamante et al., 1998; Macdonald, 1988; Macdonald and Sprague, 1988). Ringed seals from Inukjuaq and the Labrador fjords have been reported to consume mainly pelagic forage fish and amphipods (Young and Ferguson, 2013; Yurkowski et al., 2015a), indicating that the high amphipod consumption in these areas may explain the elevated TCd concentrations reported in seals from these areas. While, ringed seals from western Hudson Bay (Arviat) have been reported to consume mostly Sand lance (*Ammodytes* sp.), Arctic cod (*Boreogadus saida*), and capelin (*Mallotus villosus*) (Chambellant et al., 2013; Yurkowski et al., 2015a), they have also been reported to have high niche overlap with eastern Hudson Bay ringed seals (Yurkowski et al., 2015a) and have been found to have increased consumption of invertebrates during the open-water period (Chambellant et al., 2013), which may be contributing to their elevated Cd levels.

The higher TCd concentrations in seals from the eastern Arctic may be partly due to different environmental background concentrations of Cd in the two regions. While no published measurements of Cd concentrations are currently available for marine sediments across the Arctic, our results (Fig. 2) are consistent with higher relative TCd concentrations in eastern Canadian Arctic ringed seals, beluga whales and polar bears (Braune et al., 1991; Norstrom et al., 1986; Wagemann et al., 1996).

Best-fit models using AIC (Table 4) confirmed that location explains the variation in Cd levels across the Arctic. In addition, TP and relative

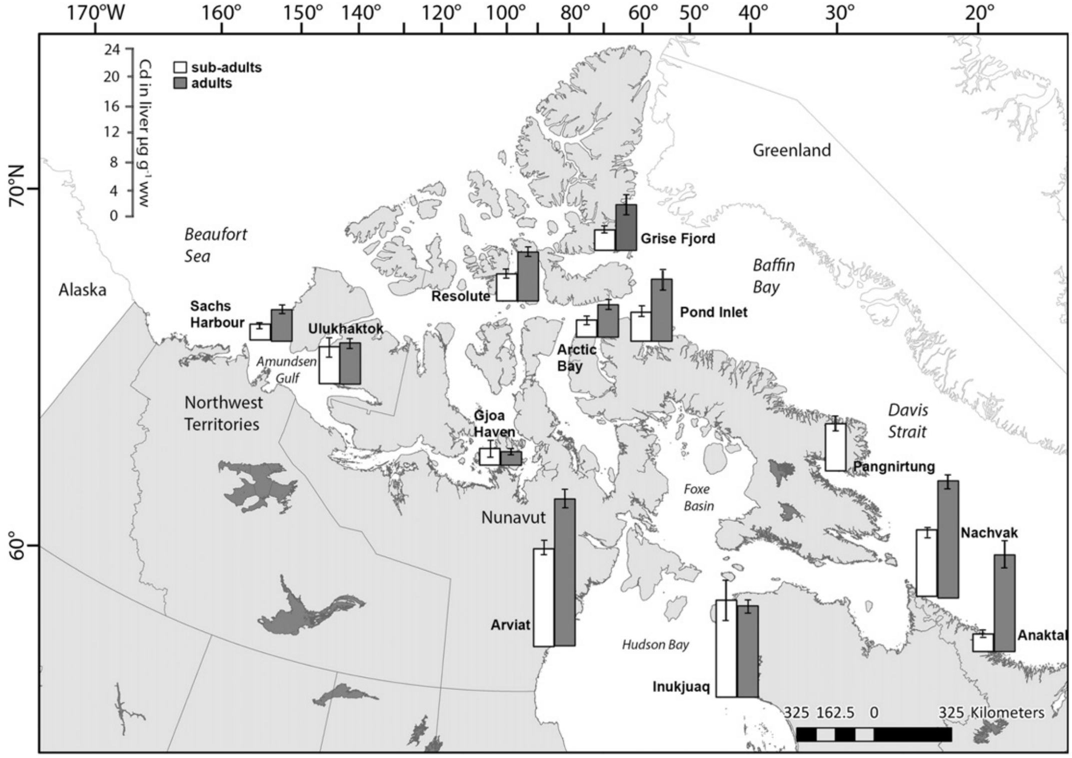

Fig. 2. Average TCd concentrations ± SD in sub adult and adult ringed seal liver from 10 communities across the Canadian Arctic sampled between 2007 and 2011.

carbon source contributed to the final model for sub adult and adult ringed seals (Table 4), which indicates that feeding ecology plays an important role in influencing TCd levels in ringed seals across the Arctic.

3.3. Feeding ecology and THg and TCd levels

THg increased with trophic position (TP) in adult (muscle: $r^2 = 0.32$, $p < 0.001$, Fig. S1; liver: $r^2 = 0.17$, $p = 0.04$) and sub adult (muscle: $r^2 = 0.36$, $p < 0.001$, Fig. S1) ringed seals. No relationship was found between THg and TP in liver tissue ($r^2 = 0.10$, $p = 0.16$) of sub adult ringed seals. As expected, THg levels in ringed seals reflect biomagnification processes. This observation is consistent with previous findings for Arctic beluga whales, where THg concentrations in muscle and liver positively correlated with δ^{15}N (Loseto et al., 2008b). The highest TP was observed in Gjoa Haven (5.6) and Sachs Harbour (5.1) for sub adult ringed seals and in Sachs Harbour (5.3), Resolute (5.2) and Gjoa Haven (5.2) for adult ringed seals (Fig. 3; Table 3). Sachs Harbour, Ulukhaktok and Resolute ringed seals had the highest THg concentrations (Tables 1 and 2) for both sub adult and adults. Levels of TP for ringed seals from these locations were high and exceeded average levels of TP (4.5) for sub adult and adult ringed seals from across the Canadian Arctic (Table 3). These results suggest that feeding on higher trophic level prey (e.g. Arctic cod) is contributing to the elevated THg levels in seals from these locations. In contrast, low THg concentrations were observed in sub adult and adult ringed seals from Inukjuaq and the four fjords in northern Labrador and in sub adults from Arctic Bay. These low THg concentrations (Tables 1 and 2) are consistent with the relatively low TP levels (averages were <4.3) reported in these seals (Table 3).

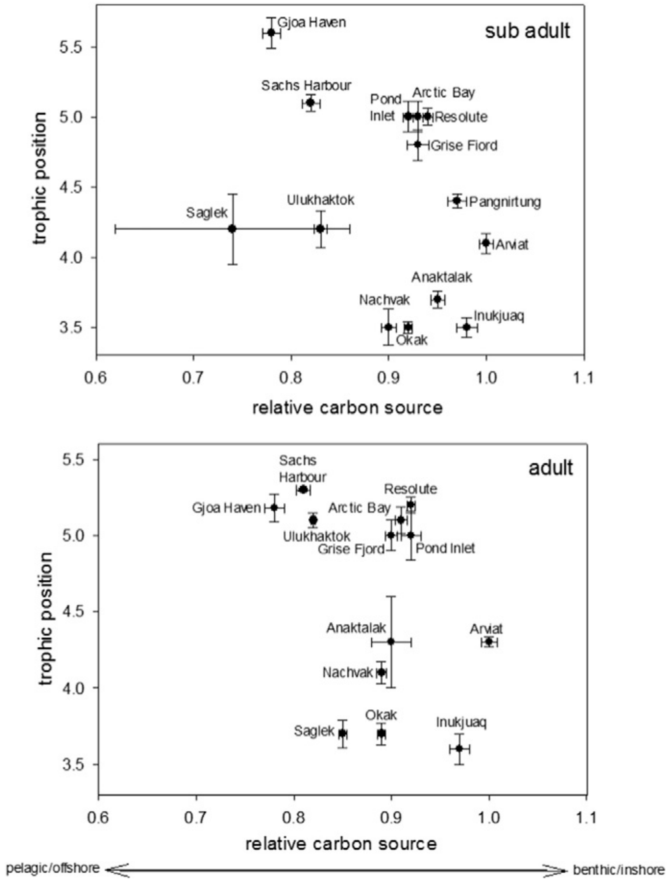

Fig. 3. Mean TP and relative carbon source ± SE for ringed seal muscle tissues collected from the Canadian Arctic. Relative carbon source values close to 1 or below are pelagic and values greater than 1 represent a more benthic carbon source.

Sub adult ringed seals from Gjoa Haven and Grise Fjord and adult ringed seals from Grise Fjord and Arctic Bay had relatively low levels of THg and high TP levels (averages were >4.8) (Tables 1, 2 and 4). Gaden et al. (2009) reported a four-month lag between when THg from prey is deposited in ringed seal muscle and when stable isotope signatures of prey are incorporated into muscle. Ringed seals can experience dietary shifts seasonally (Dehn et al., 2005), thus the inconsistencies between THg and TP reported at these three locations could be due to a change in their foraging ecology during the four-month lag period. Other possible explanations for the low levels of THg and high TP in ringed seals from Gjoa Haven is that these seals feed predominately on Arctic char (*Salvelinus alpinus*) which have been reported to have extremely low THg concentrations across the Arctic (Braune et al., 2015; Evans et al., 2015) or that there are naturally low levels of Hg in the marine system. Consistent with our findings, Gjoa Haven ringed seals have been found to have elevated δ^{15}N values and low δ^{13}C, which were attributed to seals feeding predominately on anadromous Arctic char (Butt et al., 2008; Morris, 2015).

THg in muscle decreased with relative carbon source in adult ($r^2 = 0.23$, $p < 0.001$) and sub adult ($r^2 = 0.20$, $p = 0.004$) ringed seals (Fig. S2). No univariate relationships ($p \geq 0.05$) were found between THg in liver and relative carbon source in sub adult and adult ringed seals. Best-fit models using the AIC did however suggest that relative carbon source contributed to the THg levels in liver in sub adult ringed seals (Table 4). Sub adult and adult ringed seals from all locations, except Arviat (1.0), had a relative carbon source value that was close to 1 or below (Table 3). Based on the relative carbon source values, sub adult and adult ringed seals from Sachs Harbour, Ulukhaktok, Gjoa Haven and Saglek appear to feed more in the pelagic food web than ringed seals from the rest of the locations sampled (Fig. 3; Table 3). Based on Fig. 3, TP increased in sub adult ($r^2 = 0.26$, $p < 0.001$) and adult ($r^2 = 0.20$, $p = 0.005$) ringed seals feeding more in the pelagic compared with the benthic food web. These results are consistent with stable isotope profiles reported for ringed seal populations across the Arctic, where a higher trophic position was associated with a greater proportion of pelagic or offshore food sources (Butt et al., 2008). Further, these results are consistent with adult ringed seals from Ulukhaktok and Sachs Harbour having the highest THg levels and with McMeans et al. (2015) who showed that THg increased at a faster rate in the pelagic compared with the benthic food webs of Greenland sharks (*Somniosus microcephalus*). However, prior to drawing further conclusions on the importance of ringed seals feeding pelagically versus benthically in influencing their δ^{15}N and THg levels, more research is needed on stable isotope ratios and THg concentrations in ringed seal prey across habitats and ecosystems.

TCd in liver decreased across all sites with TP in adult ($r^2 = 0.39$, $p < 0.001$) and sub adult ($r^2 = 0.40$, $p < 0.001$) ringed seals (Fig. S1). The lowest TP was observed in sub adult and adult seals from Inukjuak and the four Labrador fjords (Nachvak, Saglek, Okak, Anaktalak) (Fig. 3; Table 3). These results are consistent with previous studies which reported that ringed seals from Inukjuaq consume mainly pelagic forage fish and amphipods (Young and Ferguson, 2013; Yurkowski et al., 2015a) and that ringed seals from the Labrador fjords consume predominately lower trophic level prey (Yurkowski et al., 2015a). Inukjuak, Arviat and the Labrador fjord ringed seals had the highest TCd concentrations (Tables 1 and 2) for both sub adult and adults. The high amphipod consumption likely explain the elevated TCd concentrations found in the Inukjuaq and Labrador fjord ringed seals compared with seals sampled elsewhere. TP in Arviat seals (sub adult: 4.10 ± 0.07; adult: 4.31 ± 0.03) were below average for both sub adult (4.49 ± 0.04) and adult (4.49 ± 0.05) ringed seals from across the Canadian Arctic (Table 3). These results suggest that seals from Arviat are consuming more lower trophic level prey compared with other ringed seal populations in the Canadian Arctic. Further, ringed seals from Arviat display a niche overlap with eastern Hudson Bay (Inukjuak) seals (Yurkowski et al., 2015a) and have been found to have increased consumption of invertebrates during the open-water period (Chambellant et al., 2013), which may be exposing them to higher Cd levels via increased consumption of amphipods. High levels of Cd have been reported for hyperiid amphipods (*Parathemisto libellula*) and copepods from the Canadian Arctic, averaging 6.3 and 5.0 μg/g, respectively (Bohn and McElroy, 1976; Hamanaka and Ogi, 1984; Macdonald and Sprague, 1988; Ritterhoff and Zauke, 1997). In contrast, concentrations in mysids and whole Arctic cod were 10-fold lower with 0.17 and 0.62 μg/g, respectively (Bohn and McElroy, 1976; Macdonald and Sprague, 1988).

TCd in liver increased with relative carbon source in adult ($r^2 = 0.31$, $p < 0.001$) and sub adult ($r^2 = 0.20$, $p = 0.006$) ringed seals (Fig. S2). High TCd levels in marine mammals can also be attributed to feeding heavily on benthic invertebrates (Dehn et al., 2005). Adult and sub adult ringed seals from Arviat had a carbon source value of 1.0 which suggests that benthic feeding could be playing a role in the elevated TCd levels in these seals. The average relative carbon source values in ringed seals from all other locations was less than 1.0 which suggests that these seals are feeding more on pelagic species than on benthic prey (Table 3; Fig. 3).

3.4. Health risks to Canadian Arctic ringed seals

Previous field studies have shown associations between THg and TCd and adverse health effects (e.g. neurotoxicity, immunotoxicity, liver and kidney lesions) in marine mammals (AMAP, 2005; Basu et al., 2009; Frouin et al., 2012; Rawson et al., 1993). However, with the confounding factor of complex mixtures inherent in field studies, evidence of a cause–effect relationship between an adverse health effect and a single contaminant has been difficult to achieve.

Effects thresholds for marine mammal species are virtually nonexistent. To our knowledge, only two hepatic toxicity thresholds for THg and TCd exist for marine mammals. These include the THg toxicity threshold (61 μg/g ww, (Rawson et al., 1993)) for liver abnormalities which was determined in Atlantic bottlenose dolphins (*Tursiops truncates*) and the TCd toxicity threshold of 200 μg/g ww (AMAP, 2005) for liver abnormalities. The only ringed seals in which the mean value exceeded the THg threshold value (61 μg/g ww) for toxic effects in marine mammals was for adult seals from Sachs Harbour (70 μg/g ww). Adult ringed seals from Ulukhaktok, Resolute and Arviat had mean THg concentrations that approached the terrestrial mammal toxic threshold value (30 μg/g ww, (Thompson, 1996)). None of the ringed seals exceeded or approached the TCd toxicity threshold (200 μg/g ww) for toxic effects in marine mammals or the terrestrial mammal toxic threshold (40 μg/g ww). These results collectively suggest that among Arctic ringed seals, adults from Sachs Harbour may be those considered to be most at risk for toxic effects from THg exposure. It is important to note that the proportion of methyl mercury, which is the toxic form of Hg, will vary with species and organ type and potentially other factors (e.g., age) (Dehn et al., 2005), and as such could confound these comparisons.

4. Conclusions

The present study illustrates that location due to natural geological differences across the Arctic and feeding ecology can influence THg and TCd concentrations in tissues of ringed seals. THg concentrations were highest in the western Canadian Arctic, while TCd concentrations were highest in the eastern Canadian Arctic (Hudson Bay and Labrador). THg increased with increasing TP and decreasing relative carbon source, suggesting increased THg exposure for seals feeding on pelagic high-trophic position prey. In contrast, TCd concentrations increased with decreasing TP, suggesting increased TCd exposure for seals feeding on invertebrates. Further, our study shows that THg concentrations in adult ringed seals from Sachs Harbour exceeded an adverse health effects threshold for marine mammals. This study is not only important in light of the potential risks to the health of ringed seals, but also to those that rely on ringed seals as a food item, such as polar bears or Inuit (Dietz et al., 2009; Donaldson et al., 2010).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.12.030.

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