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

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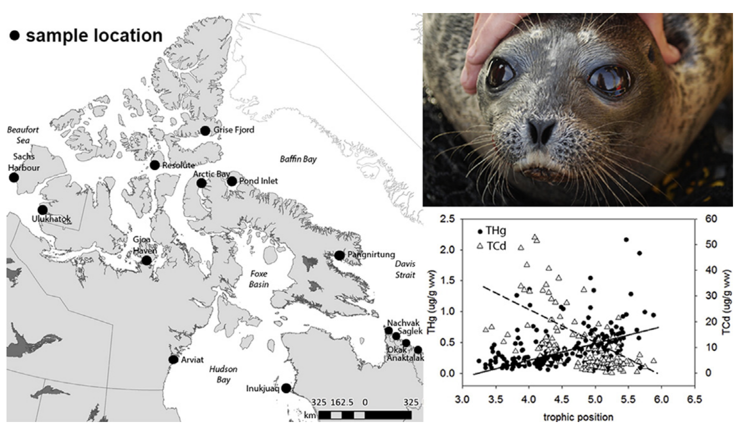
HIGHLIGHTS

- 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 ($\delta^{15}N$) and carbon ($\delta^{13}C$) have been used to understand diet and interpret contaminant trends in marine species. Based on relative isotopic fractionation processes $\delta^{15}N$ can be used to describe trophic level and $\delta^{13}C$ can be used to infer food web carbon sources. For example, enrichment of $\delta^{15}N$ 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 ($\delta^{15}N$ and $\delta^{13}C$), 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