Temporal Trends in Beluga, Narwhal and Walrus Mercury Levels: Links to Climate Change

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Abstract The exposure of Arctic marine mammals to contaminants may change via ecological dynamics in response to climate change. For example, changes to the structure of the food web or shifts in regional foraging could affect dietary exposure. We examined the temporal variation of total mercury (THg) concentrations in Hudson Bay beluga (*Delphinapterus leucas*) and Foxe Basin walrus (*Odobenus rosmarus rosmarus*) and narwhal (*Monodon monoceros*) with $\delta^{15}N$ and $\delta^{13}C$ signatures (beluga only) and the North Atlantic Oscillation. We found THg concentrations in female Arviat beluga muscle tissue decreased significantly from the early 1980s to 2008. Similarly $\delta^{13}C$ signatures in beluga sampled from Arviat declined over the same time period. $\delta^{15}N$ and the NAO index did not appear to significantly change over time nor strongly influence THg concentrations. Results suggest beluga summering in Arviat may forage in more offshore areas upon less contaminated prey in response to the increasing ice-free season over the last couple of decades. As sea ice continues to recede, dietary mercury exposure may continually decrease in beluga and other marine mammals.

Keywords Marine mammals • Contaminants • $\delta^{15}N$ • $\delta^{13}C$ • NAO index • Diet

Introduction

Monitoring contaminant concentrations in marine mammals is necessary to regulate consumption guidelines for the safety, health, and well-being of northern communities and to protect marine mammal health. In a time of accelerating warming in the Arctic (Overpeck et al. 1997), these monitoring efforts are particularly important to document what types of changes, if any, marine mammals are experiencing.

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Mercury (Hg) is both a natural element and an anthropogenic pollutant that infiltrates all biotic and abiotic components in aquatic ecosystems. Sources of Hg to Hudson Bay in order of decreasing magnitude are rivers, resuspension from glacial till on the seafloor, the atmosphere, the Arctic Ocean and coastal erosion (Hare et al. 2008). A mass balance model for Hudson Bay demonstrates these influxes are near equilibrium with exportation mechanisms (sedimentation and transport to the North Atlantic Ocean, Hare et al. 2008). A net of ~0.1 tonne Hg is added to the waterbody annually. Like the Arctic Ocean, Hudson Bay seawater is saturated with Hg (Outridge et al. 2008), so imports or exports to the overall Hg pool would not noticeably affect the 1 tonne (~1%) of Hg bound up in the biota (Hare et al. 2008).

Of the various species of Hg that exist in the marine environment, it is the methylated species (methyl mercury; MeHg) which is both bioavailable and toxic to organisms. Methylation, the process of converting inorganic Hg to an organic form, is mediated by sulfate-reducing bacteria in estuaries and coasts (Sunderland et al. 2004). Recently, high MeHg concentrations have been measured in the lower depths and in the sub-surface thermocline portion of the water column (Monperrus et al. 2007; Kirk et al. 2008; Cossa et al. 2009; Sunderland et al. 2009). MeHg provides no biological use to organisms (World Health Organization 2002), yet in marine ecosystems it is readily adsorbed from the water column by microorganisms and organic matter (Morel et al. 1998; Ravichandran 2004) and is then consumed by (and correlated to) higher trophic levels (Atwell et al. 1998; Stern and Macdonald 2005; Dehn et al. 2006; Pazerniuk 2007; Loseto et al. 2008a). Therefore, due to different diets (Stewart and Lockhart 2005), marine mammals at higher trophic levels such as beluga (*Delphinapterus leucas*) and narwhal (*Monodon monoceros*) tend to have higher contaminant concentrations than lower trophic level marine mammals like walrus (*Odobenus rosmarus rosmarus*; i.e. Wagemann et al. 1995).

The elimination rate of MeHg in animals is slower compared to its rate of intake and thus bioaccumulates in the body (Wagemann et al. 1996, 1998; Carrier et al. 2001; Nigro et al. 2002). Because of their high trophic level and long life spans, marine mammals accrue large quantities of MeHg. In mammals, MeHg accumulates primarily in the liver where it undergoes demethylation in the presence of selenium, forming a less toxic, inorganic compound (mercuric selenide (HgSe), Koeman et al. 1973; Ikemoto et al. 2004). This demethylation process leaves about only 15% MeHg in the liver (Wagemann et al. 1998). Nonetheless, marine mammals face ongoing exposure to MeHg from feeding on MeHg-contaminated prey (Loseto et al. 2008a, b), so MeHg is continually entering into their tissues. In muscle tissue, for example, MeHg is the exclusive species of Hg (Wagemann et al. 1998) and its concentration in muscle reflects relatively recent dietary exposure (Loseto et al. 2008b; Gaden et al. 2009). In this text we refer to total mercury (THg) which was analyzed in both liver and muscle tissues, but the composition of the term is different for each tissue as noted above: liver THg is a product of a majority of inorganic mercury and some MeHg; muscle THg is made up almost entirely of MeHg.

The dominant effect resulting from high MeHg exposure to mammals ranging from mink (*Mustela vison*) to polar bear (*Ursus maritimus*) is hindered functioning of the brain and central nervous system (Ronald et al. 1977; Basu et al. 2005, 2007, 2009). The immune systems of beluga whales (De Guise et al. 1996) and harbour porpoises (*Phocoena phocoena*; Bennett et al. 2001) have also been observed to function abnormally from MeHg-contaminated diets. Few studies have determined threshold concentrations for health effects in marine mammals, although Law (1996) reported 60 µg/g THg (wet weight) in the liver of marine mammals was damaging to hepatic processes. In humans, health effects from high MeHg exposure include subdued neurological functions in young children (via prenatal exposure) and high blood pressure (Grandjean et al. 1992, 1997, 2003; Sørensen et al. 1999).

Few studies have found clear temporal trends in THg among Canadian Arctic marine biota from the 1980s to 2000s. Lockhart et al. (2005) documented a strong trend in Mackenzie Delta beluga in which age-adjusted liver THg concentrations increased throughout the 1980s to mid-1990s. THg content in the eggs of northern fulmars (*Fulmarus glacialis*) and thick-billed murres (*Uria Iomvia*) significantly increased from 1975 to 2003 (Braune 2007) as well. Using a global scale model with a time period of 100 years, Booth and Zeller (2005) noted an increase in MeHg levels in all marine organisms, including marine mammals. However, the majority of studies investigating THg in Arctic marine mammal populations have shown considerable variation in concentrations over time (Muir and Kwan 2003; Muir et al. 2001, 2006; Lockhart et al. 2005; Gaden et al. 2009).

Contaminant concentrations can also vary spatially among marine mammals. Spatial variation can result from differences in geography and ambient environmental concentrations. For example, in the late 1980s to early 1990s western Canadian Arctic marine mammals were observed to have higher concentrations of mercury compared to their counterparts in the eastern Canadian Arctic (Wagemann et al. 1996). At a smaller scale, Hare et al. (2008) found seawater THg concentrations were higher in Hudson Bay compared to Foxe Basin. Contaminant concentrations among spatially segregated populations may also emanate from differences in food-web structure or length (Hoekstra et al. 2003; Braune 2007; Pazerniuk 2007).

How contaminant exposure to marine mammals will change over time in association to climate change is uncertain although some progress had been made (Gaden et al. 2009; McKinney et al. 2009). Hudson Bay has experienced earlier break-up and later freeze-up over the last few decades (Gagnon and Gough 2005; Ford et al. 2009), and the length of the ice-free season in Hudson Bay and Foxe Basin has increased by 1.3 and 2.1 days/year, respectively, from 1979 to 2006 (Rodrigues 2009). Such large-scale environmental change could trigger ecological responses including shifts in the availability, abundance and types of prey species, influencing MeHg exposure to marine mammals (Tynan and DeMaster 1997; Macdonald et al. 2005; Learmonth et al. 2006; Simmonds and Isaac 2007; Burek et al. 2008).

We investigate the THg concentrations in Hudson Bay beluga and Foxe Basin walrus and narwhal harvested throughout the 1980s–2000s. THg concentrations are analyzed both over time, with length of the ice-free season, and the North Atlantic Oscillation (NAO). We include stable isotope ratios of nitrogen and carbon in our investigation to determine changes in trophic level ($\delta^{15}N$) and regional foraging ($\delta^{13}C$) (Hobson and Welch 1992; Cherel and Hobson 2007) which may influence contaminant exposure.

Methods

Field Sampling

Each year hunters from communities in the Canadian Arctic lend invaluable skills and expertise in harvest-based monitoring programs. Most marine mammal samples analyzed in this study were harvested in the open-water season. Liver and muscle tissues were collected from beluga at Arviat (1984–2008) and Sanikiluaq (1994–2008). From Sanikiluaq there were no female beluga harvested in 2002, nor were there any male muscle samples from 2004. No liver was available from 2005 female Arviat beluga. There were fewer beluga muscle samples available than liver, particularly for Arviat. Liver samples were harvested from walrus at Hall Beach (1988–2008) and Igloolik (1982–2008) and narwhal from Repulse Bay (1993–2001). Specific years and sample sizes are presented in Tables 1 and 2.

Sex and length were recorded from the samples, and the lower jaws (excluding narwhal) were collected for determining age estimates. Tissues were frozen soon

Table 1 Annual mean ages, lengths and THg (in liver, µg/g wet weight) ± standard errors in Hudson Bay beluga (separated by sex), walrus and narwhal (Liver THg values by Wagemann et al. 1983, 1995, 1996, 1998; Wagemann and Stewart 1994; Lockhart et al. 2005; Stern and Lockhart 2009)

Species, location	Year	N	Age	Length (cm)	THg in liver
Beluga, Arviat (females)	1984	18	23.3 ± 3.0	337 ± 14	6.8 ± 1.5
	1986	10	21.8 ± 5.7	306 ± 17	7.5 ± 3.0
	1997	5	33.2 ± 4.0	367 ± 19	20.6 ± 6.3
	1999	17	21.2 ± 2.4	321 ± 10	12.8 ± 2.1
	2003	13	19.7 ± 2.4	324 ± 7	17.7 ± 4.5
	2007	3	31.0 ± 11.6	230 ± 14	14.9 ± 5.0
	2008	6	23.7 ± 10.1	363 ± 16	11.6 ± 4.4
Beluga, Arviat (males)	1984	3	24.0 ± 9.9	299 ± 40	10.2 ± 6.0
	1986	5	25.4 ± 5.5	343 ± 29	7.6 ± 3.2
	1997	4	27.8 ± 5.1	371 ± 16	12.0 ± 6.9
	1999	15	22.8 ± 2.5	350 ± 13	12.4 ± 3.2
	2003	19	19.3 ± 2.6	343 ± 12	5.8 ± 1.4
	2005	9	21.3 ± 3.9		15.1 ± 5.9
	2007	9	7.9 ± 3.3	329 ± 20	4.7 ± 1.4
	2008	6	17.3 ± 6.5	363 ± 15	8.9 ± 2.9

(continued)

Table 1 (continued)

Species, location	Year	N	Age	Length (cm)	THg in liver
Beluga, Sanikiluaq (females)	1994	12	26.4 ± 3.4	307 ± 12	8.1 ± 1.5
	1995	7	24 ± 5.5	383 ± 23	11.3 ± 2.1
	1998	9	28.7 ± 3.9		21.1 ± 4.4
	2003	4	17.8 ± 8.0	273 ± 16	7.6 ± 3.6
	2004	8	40.9 ± 7.2	375 ± 15	13.3 ± 1.5
	2005	4	27.8 ± 11.6		18.2 ± 11.0
	2007	4	16.3 ± 5.1	305 ± 14	8.4 ± 4.1
	2008	5	18.5 ± 3.7	328 ± 2.6	10.2 ± 2.9
Beluga, Sanikiluaq (males)	1994	15	29.9 ± 2.8	376 ± 14	17.2 ± 22.8
	1995	7	38.1 ± 6.1	386 ± 49	42.9 ± 15.8
	1998	10	23.1 ± 3.3		24.1 ± 11.3
	2002	6	22.5 ± 3.9	372 ± 32	10.2 ± 4.3
	2003	7	17.9 ± 3.3	353 ± 33	5.8 ± 1.2
	2005	8	22.5 ± 2.8		15.9 ± 4.1
	2007	9	18.3 ± 5.2	366 ± 12	10.1 ± 4.0
	2008	7	16.0 ± 2.7	362 ± 10	8.2 ± 2.1
Walrus, Hall Beach and Igloolik	1982	11	12.7 ± 1.1	287 ± 15	1.1 ± 0.26
	1983	23	13.0 ± 0.9	284 ± 5	1.2 ± 0.18
	1987	15	9.4 ± 1.2	268 ± 12	1.2 ± 0.26
	1988	28	9.1 ± 0.9	256 ± 11	1.4 ± 0.21
	1992	20	11.1 ± 1.5	257 ± 14	1.2 ± 0.29
	1993	5	18.6 ± 1.4	320 ± 5	2.5 ± 0.43
	1996	29	15.7 ± 1.5	287 ± 11	1.9 ± 0.30
	2004	15	14.0 ± 0.9	302 ± 5	1.5 ± 0.2
	2007	7		294 ± 29	4.9 ± 1.5
	2008	7		300 ± 9	1.8 ± 1.1
Narwhal, Repulse Bay	1993	4		398 ± 7	8.9 ± 4.6
	1999	16		365 ± 16	12.0 ± 1.8
	2001	10		432 ± 11	9.8 ± 1.3

upon retrieval and shipped to the Freshwater Institute (Winnipeg, MB) for contaminant analysis. Individual growth layer groups (GLG) in the dentine portion of beluga and walrus teeth were counted as 1 year each (Garlich-Miller et al. 1993; Stewart et al. 2006).

Chemistry Techniques

Approximately 0.2 and 0.1 g of muscle and liver tissue, respectively, were used to analyze THg content. The samples were submerged in acids overnight and then placed on a heating block for 2 h the following morning. After samples were diluted with deionised water, they were analyzed for THg using Cold Vapour

Table 2 Annual mean ages, lengths and THg (in muscle, µg/g wet weight) ± standard errors in Hudson Bay beluga by location and sex

Location, sex	Year	N	Age	Length (cm)	THg in muscle
Arviat, female	1984	11	21.6 ± 3.8	326 ± 22	1.1 ± 0.1
	1986	10	21.8 ± 5.7	306 ± 17	1.2 ± 0.3
	1997	5	33.2 ± 4.0	367 ± 19	1.1 ± 0.2
	1999	10	19.8 ± 1.9	329 ± 10	0.9 ± 0.08
	2003	13	19.7 ± 2.4	324 ± 7	1.1 ± 0.1
	2005	2	17.0 ± 13.0		0.84 ± 0.47
	2007	3	31.0 ± 11.6	230 ± 14	0.86 ± 0.2
	2008	6	23.7 ± 10.1	363 ± 16	0.75 ± 0.08
Arviat, male	1984	3	24.0 ± 9.9	299 ± 40	1.1 ± 0.3
	1986	4	30.5 ± 2.7	372 ± 5	1.3 ± 0.069
	1997	4	27.8 ± 5.1	371 ± 16	0.88 ± 0.13
	1999	7	24.4 ± 3.6	362 ± 16	0.84 ± 0.047
	2003	16	18.3 ± 3.1	334 ± 13	0.68 ± 0.049
	2005	9	21.3 ± 3.9		1.1 ± 0.32
	2007	9	7.9 ± 3.3	329 ± 20	0.78 ± 0.15
	2008	6	17.3 ± 6.5	363 ± 15	0.98 ± 0.20
Sanikiluaq, female	1994	12	26.4 ± 3.4	307 ± 12	0.71 ± 0.037
	1995	7	24 ± 5.5	383 ± 23	1.0 ± 0.15
	1998	9	28.7 ± 3.9		1.2 ± 0.17
	2003	4	17.8 ± 8.0	273 ± 16	0.58 ± 0.12
	2004	8	40.9 ± 7.2	375 ± 15	1.2 ± 0.17
	2005	4	32 ± 11.7		1.2 ± 0.38
	2007	3	16.3 ± 5.1	304 ± 20	0.49 ± 0.057
	2008	5	18.5 ± 3.7	328 ± 2.6	0.63 ± 0.022
Sanikiluaq, male	1994	15	29.9 ± 2.8	376 ± 14	1.3 ± 0.17
	1995	6	38.7 ± 7.2	403 ± 55	1.8 ± 0.53
	1998	10	23.1 ± 3.3		1.5 ± 0.40
	2002	6	22.5 ± 3.9	372 ± 32	0.89 ± 0.28
	2003	7	17.9 ± 3.3	353 ± 33	0.70 ± 0.14
	2005	6	22.2 ± 1.6		0.92 ± 0.13
	2007	6	18.3 ± 5.2	355 ± 16	0.95 ± 0.16
	2008	7	16.0 ± 2.7	362 ± 10	0.73 ± 0.11

Atomic Absorption Spectroscopy (CVAAS; Armstrong and Uthe 1971). LUTS-1, TORT-2 and CRM 2976 were used as standard reference material (80–90% recovered). The limit of detection was 0.005 µg/g wet weight. All data are reported in wet weight.

Liver samples were analyzed for stable isotope (SI) ratios ($\delta^{15}N$ and $\delta^{13}C$). We chose to analyze $\delta^{15}N$ and $\delta^{13}C$ from liver tissue because SI ratios appear to have a shorter turnover time there, reflecting relatively recent prey consumption and incorporation into tissues (Loseto et al. 2008b). No liver samples from 1994 Sanikiluaq were available for this analysis. Prior to SI analysis, lipids were removed from the samples to reduce the bias in the $\delta^{13}C$ values (Kurle and Worthy 2002). A 2:1 chloroform/methanol solution was added to 0.2 g freeze-dried liver in test tubes followed by mixing, centrifuging, and removing and replacing the solvent (three times; based on Folch et al. 1957). Dried samples were packaged and sent to the University of Winnipeg Stable Isotope Laboratory for SI analysis by Continuous Flow Ion Ratio Mass Spectroscopy (CFIR-MS). PeeDee Belemnite and IAEA-N-1 were used as standards for ^{13}C and ^{15}N analysis, respectively. The calculation for SI ratios is given in Loseto et al. (2008a, b). Here units of the stable isotope ratios are given in per mil (‰) with δ notation. Average error reported in the SI analysis was 0.17‰ for ^{13}C and 0.30‰ for ^{15}N.

Climate Analysis

The length of each ice-free season in Hudson Bay and Foxe Basin over the last several decades was observed to significantly increase over the study period (data adapted from Rodrigues 2009). Because ‘year’ and ‘length of the ice-free season’ were significantly correlated, we used ‘year’ only in the statistical analysis and focused on temporal trends. The annual wintertime NAO index (December–March, Gough et al. 2004) in the winter prior to the summer harvest was calculated from monthly data available on the National Oceanic and Atmospheric Administration website (NOAA, http://www.cpc.noaa.gov/products/precip/CWlink/pna/nao.shtml). We note that the NAO index was not significantly correlated to year.

Statistical Analysis

After log-transforming all data, we tested for the effects of age, length, day of year samples were harvested, location, sex and the interaction of these variables separately for each marine mammal species. Raw $\delta^{15}N$ and $\delta^{13}C$ values were normally distributed so we did not log-transform these for the statistical analyses. A student’s t-test examined differences in THg between the sexes for each year. For the time-series analysis we first used a general linear model (GLM) to assess which variables were significantly associated with THg, $\delta^{15}N$ and $\delta^{13}C$ (including the NAO index, year, $\delta^{15}N$, $\delta^{13}C$, and a categorical variable defining individuals by sex and location). The temporal variation of raw THg concentrations was illustrated with boxplots. Finally we tested for differences between mean annual (log-transformed) THg within species using an analysis of covariance (ANCOVA) and Bonferroni post-hoc tests. All statistical analyses were carried out with SYSTAT 11.

Results

Mercury Concentrations in Three Marine Mammals

Beluga

Due to observations in our time-series analysis, we report mean data of the sexes separately in Tables 1 and 2. The interval of mean annual liver THg concentrations in Arviat beluga (5–21 μg/g) was half that of Sanikiluaq beluga (5–43 μg/g). The large range of liver THg values is likely a result of the wide range of ages. Mean THg values presented here agree with values obtained from other Canadian Arctic beluga in the 1990s and early 2000s (Muir 2005, 6–44 μg/g ww). Mean muscle THg concentrations in beluga fell within 0.68–1.3 μg/g in Arviat and 0.49–1.8 μg/g in Sanikiluaq and are similar to values reported in Beaufort Sea beluga (Loseto et al. 2008a, 0.67–1.70 μg/g ww[1]).

Sex and location were not significant factors influencing beluga liver THg concentrations. In muscle, however, mean THg concentrations in female Arviat animals were higher than those of Sanikiluaq females ($F = 2.88$, $p = 0.036$, age as covariate). This may be attributed to the longer length of the food web (which results in higher contaminant exposure to top predators) in western Hudson Bay in comparison to eastern Hudson Bay (Pazerniuk 2007). When 'year' was accounted for, the significance dropped ($p = 0.078$).

Age was significantly related to liver and muscle THg in beluga from both Arviat ($r = 0.60$, $p < 0.001$; $r = 0.58$, $p < 0.001$) and Sanikiluaq ($r = 0.60$, $p < 0.001$; $r = 0.69$, $p < 0.001$). In addition, length was related to THg in both tissues for both locations ($p < 0.05$). Positive relationships between age, length and THg in liver and muscle have been previously documented (Hansen et al. 1990; Wagemann et al. 1995; Siebert et al. 1999; Woshner et al. 2001). In beluga, age tends to be strongly associated with THg in liver whereas length is more closely associated with muscle THg (Loseto et al. 2008b). In this study there was no length data for 2005 Arviat or 1998 and 2005 Sanikiluaq beluga. However, length and age were significantly related (Arviat $r = 0.56$, $p < 0.001$; Sanikiluaq $r = 0.51$, $p < 0.001$). Taken together, these findings lead us to use age as a covariate to control variation in our temporal analysis of both liver and muscle THg.

Walrus

No significant effects were observed with sex, day of year, or location, so we pooled sexes and samples from both communities at Igloolik and Hall Beach. Liver

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[1] Conversion of THg concentration in dry weight to wet weight was calculated assuming 74% moisture content in beluga muscle (DFO archives).

THg concentrations fell within a small range of 1–5 µg/g (Table 1) which is similar to concentrations reported for walrus at Inukjuaq (2.64 µg/g ww in liver, Muir et al. 2000). Age was significantly related to THg in liver ($r = 0.56$, $p < 0.001$), as was length to THg ($r = 0.46$, $p < 0.001$). However, without age data for either community in 2007 and 2008, and with walrus age and length significantly related ($r = 0.87$, $p < 0.001$), we chose to use length as a covariate in controlling variation of data in the temporal analysis.

Narwhal

Narwhal liver samples were collected at Repulse Bay during 1993, 1999 and 2001. We pooled the sexes since there were fewer samples. Mean annual liver THg values fell between 8 and 12 µg/g (Table 1) and coincided with concentrations measured in narwhal from western Greenland (2–17 µg/g ww, Dietz et al. 2004) and in Pond Inlet (mean THg in liver: 12.9 µg/g ww, Wagemann et al. 1983). Narwhal length was positively related to concentrations of liver THg ($r = 0.37$, $p = 0.048$), so it was selected as a covariate for the time series analysis.

Time Series Analysis

Beluga

Female beluga from Arviat had significantly higher liver THg content than males in 2003 ($t = -2.173$, $p = 0.038$), and 1994 Sanikiluaq male beluga had significantly higher liver THg than females ($t = 2.377$, $p = 0.025$). Due to these observations we plotted male and female concentrations of THg in liver separately (Fig. 1a–d). Regression coefficients of age-adjusted liver THg by year are presented in Table 3. Only the female beluga from Arviat had liver THg concentrations that significantly changed (increased) over the years of collection. There were no significant differences in annual mean liver THg between sex or location. Apart from age, the GLM detected no other significant factors associated with liver THg concentrations.

Males from Sanikiluaq had significantly higher THg concentrations in muscle compared to females in 2007 ($t = 2.877$, $p = 0.024$) (Fig. 2c, d). In contrast to liver, age-adjusted muscle THg concentrations declined over time in all beluga and the trend was significant in Arviat female animals (Table 3). THg concentrations in Sanikiluaq female beluga were significantly higher in 1998 compared to 1994 (Bonferroni $p = 0.018$). The GLM for muscle THg revealed that age was the only significant factor (Table 4).

The importance of $\delta^{15}N$ and $\delta^{13}C$ driving beluga THg concentrations was assessed with GLMs. $\delta^{13}C$ was the only variable to significantly influence $\delta^{15}N$ values (Table 4). The GLM for $\delta^{13}C$ detected four significant variables: $\delta^{15}N$, year,

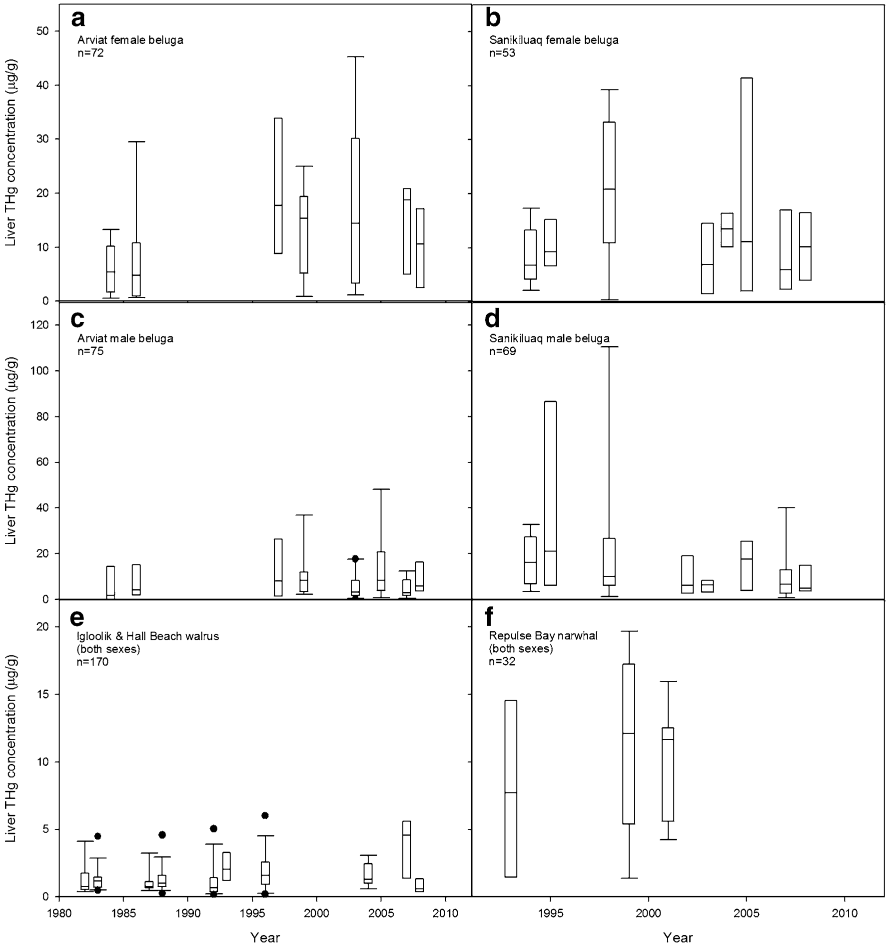

Fig. 1 Box and whisker plots of yearly THg concentrations (µg/g ww) in beluga liver sampled in Arviat (females (**a**), males (**c**)) and Sanikiluaq (females (**b**), males (**d**)) and walrus (**e**) and narwhal (**f**) sampled in Foxe Basin. *Lines in boxes* represent mean concentrations and *whiskers* represent 5th and 95th percentiles. Outliers are indicated by black dots

NAO index and sex/location. An ANCOVA (year as covariate) revealed no significant difference in $\delta^{13}C$ between sexes for Arviat and Sanikiluaq beluga. However, Arviat beluga were significantly enriched in $\delta^{13}C$ compared to Sanikiluaq beluga ($F = 32.52$, $p < 0.001$). Mean annual $\delta^{13}C$ was regressed as a function of both year and NAO by location (sex pooled) to illustrate these specific relationships (Fig. 3). $\delta^{13}C$ decreased over time in beluga sampled from Arviat ($r = -0.70$, $p = 0.05$). Only in Sanikiluaq animals did $\delta^{13}C$ appear to respond to the NAO index in a positive relationship ($r = 0.77$, $p = 0.03$). After removal of the

Table 3 Time-series analysis of THg in liver and muscle tissues of Hudson Bay marine mammals via linear regression. Prior to analysis THg concentrations in beluga were adjusted to age; THg concentrations in walrus and narwhal were adjusted to length

	Regression coefficient	
Species, location	Liver	Muscle
Beluga, Arviat (females)	0.87*	−0.80*
Beluga, Arviat (males)	0.46	−0.23
Beluga, Sanikiluaq (females)	−0.18	−0.34
Beluga, Sanikiluaq (males)	−0.42	−0.31
Walrus, Hall Beach and Igloolik (both sexes)	0.51	–
Narwhal, Repulse Bay (both sexes)	0.59	–

*p < 0.05

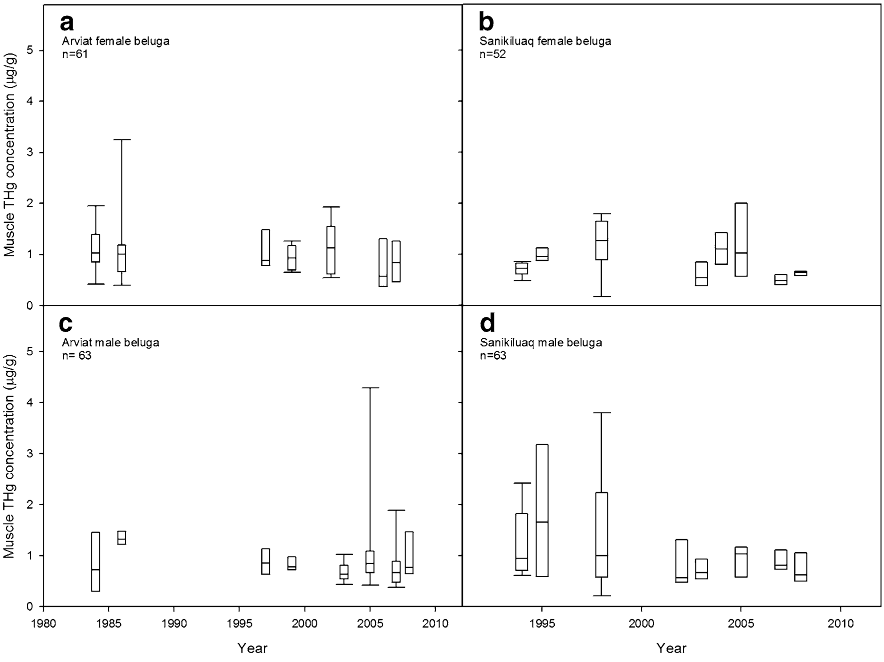

Fig. 2 Box and whisker plots of annual THg concentrations in beluga muscle (µg/g ww) sampled at Arviat (females (**a**), males (**c**)) and Sanikiluaq (females (**b**), males (**d**)). *Lines in boxes* represent mean concentrations and *whiskers* represent fifth and 95th percentiles

highest mean $\delta^{13}C$ in Sanikiluaq beluga in 1995 (NAO 1.34), the significance of both year and the NAO index was lost from the GLM. We also tried removing 1995 Sanikiluaq samples from the GLM, but the significance of 'year' in the model was retained.

Table 4 Summary of three generalized linear models of THg in muscle, $\delta^{15}N$ and $\delta^{13}C$ for Hudson Bay beluga. Bold variables explain observed variance in each model at a significant level of $\alpha \leq 0.05$

Response variable	Explanatory variable	Sum of squares	Degrees of freedom	Mean-square	F-ratio	p
THg in muscle	Sex_Location	0.04	3	0.01	0.41	0.74
	Year	0.41	1	0.41	13.70	<0.001
	Age	3.57	1	3.57	118.20	<0.001
	$\delta^{15}N$	0.03	1	0.03	1.07	0.30
	$\delta^{13}C$	0.002	1	0.002	0.06	0.81
	NAO	0.05	1	0.05	1.65	0.20
	Error	5.38	178	0.03		
$\delta^{15}N$	Sex_Location	13.38	3	4.46	1.91	0.13
	Year	3.12	1	3.12	1.33	0.25
	Age	1.90	1	1.90	0.81	0.37
	$\delta^{13}C$	17.71	1	17.71	7.57	0.007
	NAO	8.35	1	8.35	3.57	0.06
	Error	451.61	193	2.34		
$\delta^{13}C$	**Sex_Location**	26.53	3	8.84	7.222	<0.001
	Year	6.74	1	6.74	5.50	0.02
	Age	1.27	1	1.27	1.04	0.31
	$\delta^{15}N$	9.27	1	9.27	7.57	0.007
	NAO	7.62	1	7.62	6.22	0.01
	Error	236.32	193	1.22		

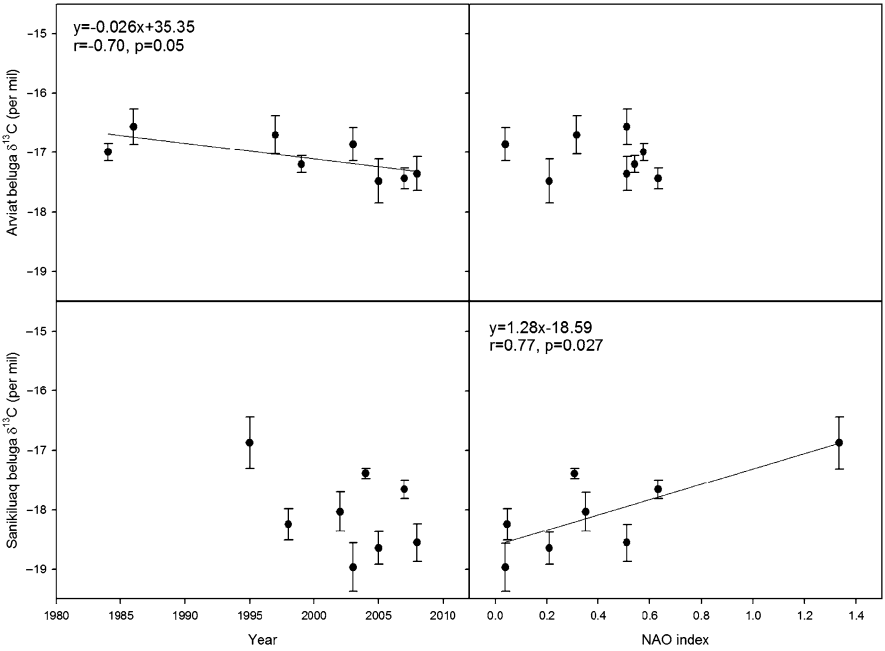

Fig. 3 Annual means and standard error bars of $\delta^{13}C$ in beluga from both Arviat (*top panels*) and Sanikiluaq (*lower panels*) regressed as a function of year and the NAO index

Walrus

Length-adjusted THg concentrations in Igloolik and Hall Beach walrus liver did not change significantly over the 26-year period (Fig. 1e, Table 3). Liver THg concentrations in samples from 2007, however, were significantly higher than those from 1982, 1983 and 1992 (length as covariate; Bonferroni p = 0.02, 0.024, 0.011, respectively). The NAO index was not significantly associated with liver THg concentrations as assessed with the GLM.

Narwhal

Length-adjusted liver THg concentrations in Repulse Bay narwhal also did not show any significant temporal trend (Fig. 1f, Table 3). There were no significant differences between THg concentrations analyzed from the 3 years samples were collected. Again the NAO index was not significantly associated with liver THg concentrations in narwhal.

Discussion

Trends in Liver Mercury Concentrations

An increasing trend in liver THg was observed only in female beluga from Arviat. The lack of a trend in male belugas may suggest habitat segregation and differences between prey selection between the sexes (Loseto et al. 2006, 2008b). Liver THg increased in Mackenzie Delta beluga from 1984 to 1996, although sexes were pooled in the analysis because there was no effect of gender (Lockhart et al. 2005). However, similar to this study, the remaining Canadian Arctic beluga populations examined showed no strong trends in liver THg from the early 1980s to 2000s. Walrus THg did not change significantly, corresponding to previous work in Northwest Greenland (Riget et al. 2007), nor did THg in narwhal. Because the half-life of Hg in liver is relatively long (10 years in humans; Friberg et al. 1979) and MeHg is ultimately converted to HgSe in marine mammal liver (Koeman et al. 1973; Ikemoto et al. 2004), liver THg may be better suited to monitoring longer term trends of Hg in the environment compared to muscle.

Beluga: Trends in Muscle Mercury and Stable Isotopes

Mercury concentrations in muscle may be more useful for dietary related trend analysis since these concentrations reflect relatively recent MeHg exposure (Loseto et al. 2008b; Gaden et al. 2009). The THg concentrations in muscle of female beluga from Arviat declined from 1984 to 2008, suggesting a decrease in dietary exposure. Although little is known about the Hudson Bay beluga diet, capelin (*Mallotus villosus*) was found to be an important prey species in the 1980s (Kelley et al. this volume), however Arctic cod (*Boreogadus saida*) is thought to dominate the diet of other circumpolar beluga populations (e.g. Loseto et al. 2009; Dahl et al. 2000). There are no temporal fish data sets in Hudson Bay to support trends in dietary exposure. A long term temporal data set for High Arctic land locked char (*Salvelinus alpinus*) showed a decrease in muscle THg in 2006 relative to 1992 that was related to diet (measured via δ^{15}N levels) (Gantner et al. 2009). Opposite trends in muscle THg were recorded in female adult hooded seals (*Cystophora cristata*) from the Greenland Sea where 1999 concentrations were significantly higher than 1985 values (Brunborg et al. 2006). Few data sets have found significant increasing or decreasing trends largely due to a few data points in which to extract significant time trends (Arctic Monitoring and Assessment Programme 2007). The inclusion of SI ratios can significantly alter results in THg trend analysis (Riget et al. 2007). The relatively unchanged δ^{15}N values in this study imply that beluga in Hudson Bay are not necessarily consuming different species or at least not switching to higher or lower trophic level prey.

δ^{13}C, on the other hand, significantly decreased in Arviat animals, but δ^{13}C was not significantly related to THg in muscle. In beluga, concentrations of muscle THg are associated with SI ratios in liver, although δ^{13}C may have a longer turnover rate compared to THg (Loseto et al. 2008b), which may explain why THg in muscle in this study was not significantly related to δ^{13}C in liver even though both variables declined over time. The downward, increasingly negative trend in δ^{13}C signatures in Arviat beluga samples suggest that belugas that summer in Arviat may be spending more time foraging in further offshore regions or that they are more dependent on pelagic sources of prey where δ^{13}C are typically more depleted (Hobson and Welch 1992; Cherel and Hobson 2007).

Dietary changes have been observed elsewhere in Hudson Bay. Thick-billed murres (*Uria lomvia*) in northern Hudson Bay gradually began favouring capelin and sandlance in place of Arctic cod and sculpin from the early 1980s to 2002 (Gaston et al. 2003; Braune 2009; Mallory et al. this volume). Braune (2009) reported lower δ^{15}N in the eggs of thick-billed murres on Coats Island (northern Hudson Bay) from 1998 to 2007 in comparison to 1993, but there was no clear temporal trend in THg. The lower δ^{15}N in the seabirds in the more recent years was likely a result of the lower trophic position of sandlance and capelin in comparison to Arctic cod (Braune 2009). The shift in prey species may be the response of increasingly longer ice-free seasons (Gaston and Hipfner 1998; Gaston et al. 2009; Mallory et al. this volume). McKinney et al. (2009) also observed a climate-related dietary shift in the western Hudson Bay subpopulation of polar bear: with longer open water periods, the proportion of open-water seals (harbour and harp seals; *Phoca vitulina* and *Phoca groenlandica*, respectively) increased and the fraction of seals living in pack ice (bearded seals; *Erignathus barbatus*) decreased in the diet.

We believe the decreasing trends in THg and δ^{13}C in female beluga from Arviat are driven primarily by ecological shifts influenced by warming in Hudson Bay

(Gagnon and Gough 2005; Rodrigues 2009) – not by changing ambient THg concentrations. To illustrate this point, we refer to a study by Kirk and St. Louis (2009): river flows and Hg export to Hudson Bay were particularly high from the Nelson and Churchill Rivers in 2005 (Kirk and St. Louis 2009), yet we observed no parallel increases in THg in Arviat or Sanikiluaq beluga the same or following years.

Other ecological factors which may have contributed to the declining THg concentrations in female Arviat beluga are (1) elevated primary production levels associated with warming temperatures and (2) shifts in the migrations of the beluga. The first example involves high volumes of pelagic organic matter produced as a result of the lengthening ice-free season. The large quantity of organic matter may have adsorbed a higher quantity of MeHg from the water column, sunk to the benthos, and overall lowered the contaminant concentration for bioaccumulation (Outridge et al. 2005, 2007, 2008; Kuzyk et al. in press). Additionally, the warming climate may have affected the migrations of beluga, either in the timing or routes, due to altered prey distribution or travel accessibility (Tynan and DeMaster 1997; Learmonth et al. 2006; Stern and Macdonald 2005). Accounting for migration patterns in relation to diet shifts (and associated Hg exposure) in Hudson Bay beluga presents a challenge. The seasonal distribution and migration patterns appear variable even at the individual level (Richard and Orr 2003) paralleling genetic studies showing beluga stocks can mix amongst each other (De Marche and Postma 2003). Whereas we cannot submit evidence to suggest Hudson Bay beluga may have changed their migration patterns over the study period, we also cannot overlook the potential this possibility holds for explaining our observations.

The NAO index did not appear to influence THg in any marine mammals, nor $\delta^{15}N$ or $\delta^{13}C$ in beluga; its impacts to dietary exposure of MeHg and prey selection may be indirect and more complex. However, we did observe relatively high $\delta^{13}C$ signatures in Sanikiluaq beluga in 1995 which had the maximal NAO index (1.34) for the sampling years here (Fig. 3). High positive phases of the NAO tend to be associated with low temperatures and more ice in Hudson Bay (Qian et al. 2008). The Canadian Ice Service (http://ice-glaces.ec.gc.ca/) also reports above-average ice cover in 1994 and 1995 for eastern Hudson Bay. Thus the high $\delta^{13}C$ signatures in 1995 Sanikiluaq beluga may indicate that animals were foraging upon more inshore or benthic prey that year due to heavy ice conditions (Hobson and Welch 1992; Cherel and Hobson 2007).

Conclusion

Decreasing trends in muscle THg and $\delta^{13}C$ in Arviat beluga indicate that with increasing temperatures and longer duration of the ice-free period over time in Hudson Bay (Gagnon and Gough 2005), foraging activities of beluga summering in western Hudson Bay may be shifting spatially in favour of offshore, less MeHg-contaminated prey. Additionally, dietary MeHg exposure may have declined over time in response to a transfer of pelagic MeHg to the benthic environment (Outridge et al. 2005, 2007, 2008; Kuzyk et al. in press). Progressive warming in the Arctic and sub-arctic (Walsh 2008) may continue to ultimately lower THg concentrations in beluga muscle tissue. It is imperative that temporal and spatial studies on beluga and the food webs that support them continue to be able to identify changes within the marine ecosystem that are responding to climate change.

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