Environmental Science & Technology

ARTICLE

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Differences in Mercury Bioaccumulation between Polar Bears (*Ursus maritimus*) from the Canadian high- and sub-Arctic

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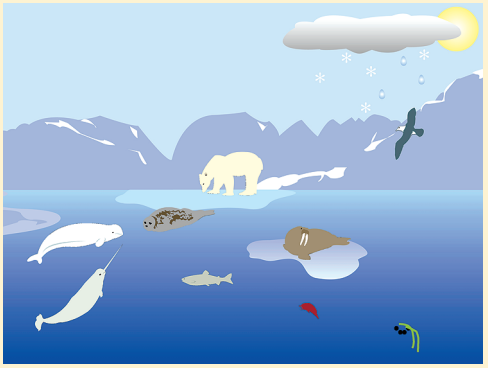
ABSTRACT: Polar bears (*Ursus maritimus*) are being impacted by climate change and increased exposure to pollutants throughout their northern circumpolar range. In this study, we quantified concentrations of total mercury (THg) in the hair of polar bears from Canadian high- (southern Beaufort Sea, SBS) and sub- (western Hudson Bay, WHB) Arctic populations. Concentrations of THg in polar bears from the SBS population ($14.8 \pm 6.6\ \mu g\ g^{-1}$) were significantly higher than in polar bears from WHB ($4.1 \pm 1.0\ \mu g\ g^{-1}$). On the basis of $\delta^{15}N$ signatures in hair, in conjunction with published $\delta^{15}N$ signatures in particulate organic matter and sediments, we estimated that the pelagic and benthic food webs in the SBS are ~4.7 and ~4.0 trophic levels long, whereas in WHB they are only ~3.6 and ~3.3 trophic levels long. Furthermore, the more depleted $\delta^{13}C$ ratios in hair from SBS polar bears relative to those from WHB suggests that SBS polar bears feed on food webs that are relatively more pelagic (and longer), whereas polar bears from WHB feed on those that are relatively more benthic (and shorter). Food web length and structure accounted for ~67% of the variation we found in THg concentrations among all polar bears across both populations. The regional difference in polar bear hair THg concentrations was also likely due to regional differences in water-column concentrations of methyl Hg (the toxic form of Hg that biomagnifies through food webs) available for bioaccumulation at the base of the food webs. For example, concentrations of methylated Hg at mid-depths in the marine water column of the northern Canadian Arctic Archipelago were $79.8 \pm 37.3\ pg\ L^{-1}$, whereas, in HB, they averaged only $38.3 \pm 16.6\ pg\ L^{-1}$. We conclude that a longer food web and higher pelagic concentrations of methylated Hg available to initiate bioaccumulation in the BS resulted in higher concentrations of THg in polar bears from the SBS region compared to those inhabiting the western coast of HB.



INTRODUCTION

Polar bears (*Ursus maritimus*) are currently listed as threatened under the United States Endangered Species Act.[1] This listing is in part because temporal and spatial reductions in sea ice extent have led to deteriorations in polar bear body condition, and declines in growth, birth, and survival rates.[2] Pollutants are also influencing polar bear condition and survival by, for example, compromising immune system functioning.[3,4] Alterations in distribution or abundance of this apex predator could have widespread cascading ecological impacts throughout the northern circumpolar region.

Although some pollutants, such as mercury (Hg), are ubiquitous in the environment, their concentrations have increased in organisms inhabiting remote polar regions through transport from more industrialized areas via the atmosphere, rivers, ocean currents, and even biological vectors (e.g., ref 5). Hg deposition phenomena called Atmospheric Mercury Depletion Events (AMDEs) also occur in high latitude regions.[6] AMDEs are heterogeneous atmospheric reactions that generally occur in the springtime atmosphere when reactive bromine species (released in bromine explosions over areas of first-year ice and open water leads in the sea ice) oxidize gaseous elemental Hg(0) to inorganic Hg(II) species operationally defined as reactive gaseous Hg and particulate bound Hg (e.g., ref 7). Both of these oxidized Hg species readily fall out of the atmosphere, primarily onto seascapes and coastal landscapes. However, once deposited, most of the oxidized Hg(II) in snow packs is rapidly photoreduced to Hg(0), which is then emitted back to the atmosphere between AMDEs, resulting in little net deposition of Hg(II).[8,9] Regardless of the source of inorganic Hg(II) to polar regions, it is methyl Hg (MeHg), not Hg(II), that is the potent vertebrate neurotoxin that bioaccumulates in organisms and biomagnifies through food webs. MeHg can be produced through the microbial methylation of Hg(II) in, for example, saturated wetland regions of freshwater watersheds that drain into polar marine waters (e.g., via the Mackenzie and Churchill rivers; refs 11,12), or in the water column and sediments of oceans themselves (e.g., ref 13).

Received: January 7, 2011
Accepted: June 1, 2011
Revised: May 29, 2011
Published: June 16, 2011

dx.doi.org/10.1021/es2000672 | *Environ. Sci. Technol.* 2011, 45, 5922–5928

Polar bears forage at the top of marine food webs throughout their high- and sub-Arctic range, making them extremely susceptible to toxic, biomagnifying pollutants such as MeHg. The proportion of different prey species in their diet may vary geographically, however. For example, the proportion of ringed seal (*Pusa hispida*), bearded seal (*Erignathus barbatus*), and beluga whale (*Delphinapterus leucas*) in the diet of bears from the southern Beaufort Sea (SBS) region of Canada, determined by fatty acid signature analyses, was 64 ± 5%, 23 ± 5%, and 13 ± 2%, respectively, whereas the proportion of these prey items in the diet of western Hudson Bay (WHB) bears ~2500 km away was 67 ± 7%, 13 ± 6%, and 0.1 ± 0.5%, respectively, as well as 15 ± 6% harbor seal (*Pagophilus vitulina*) and 5 ± 1% harp seal (*Pagophilus groenlandicus*).[14] This type of geographical variation in diet has the potential to result in regional differences in MeHg burdens in polar bears.

In this study, we quantified concentrations of total Hg (THg, all forms of Hg in a sample) in guard hairs of polar bears collected from the SBS and WHB populations. Polar bear coats are water repellent consisting of a dense and insulating layer of under hair covered by stiff and hollow guard hairs up to 15 cm long that are molted and renewed annually in May/June and into July for bears residing in the very far north.[15] The molt can last several weeks, during which time elements circulating in blood can be deposited into growing hair, which in turn can provide a snapshot of diet[16] or contaminant load during the period of hair regrowth.[17] We also analyzed hair for stable isotopes of nitrogen (δ^{15}N) and carbon (δ^{13}C) to elucidate the relationship between THg concentrations in polar bears and food web length and structure in the study regions (e.g., ref 18).

METHODS

Polar Bear Hair Collection. Guard hair samples were collected from 151 adult (>5 years old) polar bears populating two Canadian regions: 1) SBS off the northern coast of the Northwest Territories, and 2) WHB near Churchill, Manitoba (Figure 1). Seventy-seven bears (38 females [ages 6−28] and 39 males [ages 6−23]) were sampled from the SBS population in April−May 2004, whereas 74 bears (21 females [ages 7−27] and 53 males [ages 6−26]) were sampled from the WHB population in August−October 2004. As a result, hairs collected from the SBS population would have grown following the 2003 molt, whereas hairs from the WHB population would have grown following the 2004 molt. Bears were sampled nonselectively by immobilization with Telazol using standard techniques [19] as part of long-term population monitoring studies. Hair was collected from the rump of each bear by shaving an area ~2.5 cm in diameter with a scalpel. The hair was subsequently stored in WhirlPak polyethylene bags until processing. Sex of bears was determined in the field, whereas individuals were aged using a vestigial premolar as described in Calvert and Ramsay.[20]

Analytical Methods. Hair samples were washed before analyses to remove repellant oils and extraneous dirt. Hair was agitated, using stainless steel forceps, in warm Milli-Q water containing a small amount of mild Ivory dishwashing detergent, then rinsed three times with Milli-Q water and air-dried overnight in the University of Alberta Biogeochemical Laboratory (UABL; Edmonton, AB) class-100 clean room.

Total Hg Analyses. Hair samples were analyzed for THg in the UABL using standard protocols. Whole hair samples (20.9 ± 5.2 mg, mean ± S.D.) were digested in 60 mL sealed Teflon digestion vessels using 7 mL of 7:3 (v/v) HNO_3/H_2SO_4. Digestion vessels were heated in a vented oven for two hours at 125 °C. Once cooled, 19 mL of Milli-Q water and 1 mL of BrCl were added to each digestion vessel, and vessels were reheated overnight at 60 °C. A 0.5 mL subsample of the digest was then diluted with Milli-Q water to a final volume of 50 mL, and 0.04% (v/v) hydroxylamine hydrochloride was added to neutralize excess BrCl. Sample reduction, delivery, and cold-vapor atomic fluorescence detection were accomplished using an automated Tekran 2600 THg analyzer. Spike recoveries in samples were 98.9 ± 11.6%, and duplicate analyses were within 10% of one another. Average concentrations of THg measured in Certified Reference Materials (National Research Council Canada) DORM-2 and DORM-3 (4.42 ± 0.17 μg g^{-1} and 0.40 ± 0.01 μg g^{-1}, respectively) were within their certified ranges (4.64 ± 0.26 μg g^{-1} and 0.382 ± 0.060 μg g^{-1}, respectively). The analytical limit of detection was 0.5 ng g^{-1}.

δ^{15}N and δ^{13}C Analyses. Hair samples were also analyzed for δ^{15}N and δ^{13}C in the UABL using standard protocols. A whole hair (0.70−0.90 mg) was transferred into a tin capsule and weighed using a microbalance. Samples were analyzed for δ^{15}N and δ^{13}C ratios using a EuroVector EuroEA3028-HT elemental analyzer coupled to a GV Instruments IsoPrime continuous-flow isotope ratio mass spectrometer. δ^{15}N and δ^{13}C ratios (‰) were determined using the following equation:

$$\delta R‰ = ((R_{sample}/R_{standard}) - 1) \times 1000$$

where R_{sample} was the ratio of ^{15}N/^{14}N or ^{13}C/^{12}C in the sample, and $R_{standard}$ for ^{13}C or ^{15}N was referenced to that in Pee Dee Belemnite and air, respectively. National Institute of Standards and Technology 8415 whole egg powder Standard Reference Material was used as an in-house δ^{15}N and δ^{13}C QA/QC check throughout analyses; the standard deviations for these δ^{15}N and δ^{13}C values were below 0.1‰.

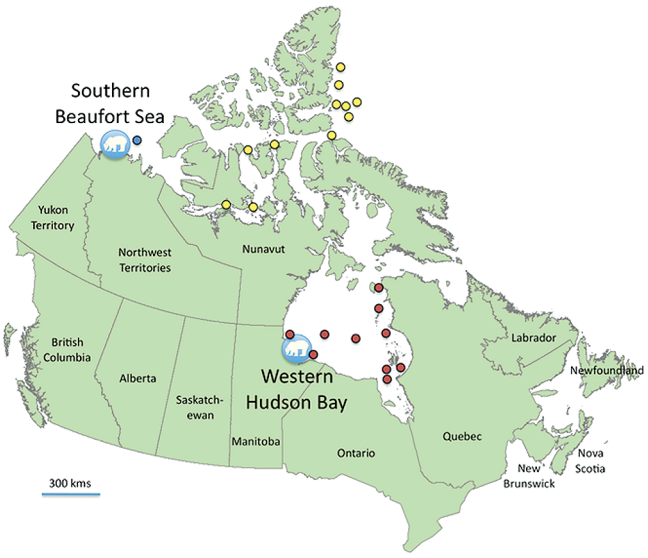


Figure 1. General locations where polar bears were sampled in the southern Beaufort Sea and western Hudson Bay regions of Canada. Dots denote sites that were previously sampled[50] for concentrations of total Hg (THg) and methylated Hg species in the water column of Hudson Bay (red), the Canadian Arctic Archipelago (yellow), and the Beaufort Sea (blue).

Table 1. Average (± S.D.) Total Hg (THg) Concentrations and Stable Isotope Ratios (δ^{15}N and δ^{13}C) in Hair from Polar Bears in the Southern Beaufort Sea (SBS) and Western Hudson Bay (WHB) Regions of Canada[a]

population	# samples	age (yrs)	THg (μg g^{-1})	δ^{15}N (‰)	δ^{13}C (‰)
SBS					
all bears	77	12.8 ± 5.1	14.8 ± 6.6	20.55 ± 0.49	−17.21 ± 0.35
males	39	12.1 ± 4.0	13.1 ± 5.0	20.49 ± 0.53	−17.25 ± 0.34
females	38	13.4 ± 6.0	16.6 ± 7.7	20.63 ± 0.43	−17.17 ± 0.35
WHB					
all bears	74	14.5 ± 6.0	4.1 ± 1.0	19.36 ± 0.56	−16.67 ± 0.24
males	53	13.9 ± 5.5	3.9 ± 0.8	19.60 ± 0.46	−16.60 ± 0.22
females	21	16.0 ± 6.9	4.5 ± 1.3	18.77 ± 0.30	−16.84 ± 0.22

[a] Average age of the bears sampled is also provided.

RESULTS AND DISCUSSION

Concentrations of THg in hair of polar bears from the SBS population (14.8 ± 6.6 μg g^{-1}) were significantly higher than in hair of polar bears from WHB (4.1 ± 1.0 μg g^{-1}) (F = 351, p < 0.001) (Table 1, part A of Figure 2) consistent with the results of previous studies (e.g., refs 21−24). Overall, THg concentrations were significantly higher in female than in male bears (Table 1, F = 4.53, p = 0.035), and the magnitude of differences was consistent between the two populations (interaction of site and sex; F = 0.43, p = 0.511) but inconsequential compared to differences between populations. There was no significant difference in THg concentrations in the hair of female polar bears with or without cubs (F = 0.1, p = 0.756), suggesting that nursing did not alter Hg metabolism in females. Alternatively, differences in THg concentrations between the sexes are likely a function of sexual size dimorphism in polar bears, with females: 1) hunting smaller prey species that may proportionally concentrate more THg in their smaller masses when feeding at similar trophic levels as larger prey species, and 2) scavenging on protein- (and Hg-) rich carcasses in which the energy-rich fatty layer has already been consumed by larger males.[14,25] During certain times of year, females also have different movement (and hence foraging) patterns than males.[26] There were no significant relationships between THg concentrations and age of polar bears sampled in the SBS region (t = 0.39, p = 0.536) or along WHB (t = 0.37, p = 0.710) (part A of Figure 2), similar to findings of Carbona−Marek et al.[25] This is likely because THg concentrations in hair is only representative of blood MeHg concentrations during the spring/early summer molt period when follicles are producing hair, as opposed to muscle tissue, for example, which integrates Hg bioaccumulated over much longer periods of time (e.g., refs 17,27).

We propose two overarching reasons why concentrations of THg in the hair of polar bears from the SBS population are higher than in polar bears from WHB: 1) the marine food web length/structure is different in the two regions, and 2) there is a greater source of MeHg to the base of the food web in the SBS than in WHB.

1) Food Web Length/Structure. δ^{15}N ratios in organisms are used to estimate trophic position because δ^{15}N ratios of predators are typically enriched by 3−4‰ relative to their prey,[28] with an average 3.8‰ enrichment per trophic level previously documented for the Barrow Strait−Lancaster Sound (eastern Canadian Arctic) marine food web.[29] Polar bears from the SBS population had significantly higher baseline-uncorrected δ^{15}N ratios (20.55 ± 0.49‰) than polar bears from WHB (19.36 ± 0.56‰) (F = 217, p < 0.001) (Table 1, part B of Figure 2). However, δ^{15}N signatures among different polar bear populations are not directly comparable unless they have been corrected for δ^{15}N signatures at the base of the food web (e.g., ref 28). For example, higher δ^{15}N signatures in SBS polar bears could just reflect higher δ^{15}N signatures at the base of the SBS food web relative to that at the base of the WHB food web. However, on the basis of data published elsewhere, quite the opposite appears to be the case. For reasons not yet understood, δ^{15}N ratios in both particulate organic matter (POM, surrogate for the base of the pelagic food web) and sediments (surrogate for the base of the benthic food web) were higher in HB than they were in the BS (Table 2, refs 31,32). By subtracting average δ^{15}N ratios of POM (Table 2) from average δ^{15}N ratios of polar bears (apex predator, Table 1), then dividing the difference by an average enrichment of 3.8‰ per trophic level previously observed in an eastern Canadian Arctic marine food web,[29] we calculate that the pelagic food web in the SBS is 4.5−4.8 trophic levels long, whereas in WHB it is only 3.3−3.9 trophic levels long. By doing similar calculations with sediments (Table 2) instead of POM, we estimate that the benthic food web in the SBS is 4.0 trophic levels long but only 3.3 trophic levels long in WHB. These calculations suggest that pelagic and benthic foodwebs are on average 0.6−1.5 trophic levels longer in SBS than in WHB. However, this first approximation of regional differences in food web length requires more rigorous field testing, including better incorporation of seasonal and spatial variation in δ^{15}N ratios at the base of the food webs (e.g., Table 2), as well as analyses of organisms throughout the food webs as was done, for example, by Hobson and Welch[29] for an eastern Canadian Arctic marine food web.

Polar bears from the SBS population were also significantly more depleted in δ^{13}C (−17.21 ± 0.35‰) than polar bears from the WHB population (−16.67 ± 0.24‰) (F = 130, p < 0.001) (Table 1, part B of Figure 2). Unlike δ^{15}N ratios, δ^{13}C ratios change little as carbon moves through food webs but can be used to differentiate sources of carbon for organisms. In aquatic systems, more depleted δ^{13}C ratios are indicative of more pelagic- (as opposed to benthic-) based food webs,[28] further suggesting that SBS polar bears feed on food webs that are more pelagic (and longer), whereas their WHB counterparts feed on food webs that are more benthic (and shorter). Feeding on a longer, more pelagic food web appears to have led to higher THg concentrations in polar bears from the SBS region. Multiple regression analyses revealed that 36% of the variation in THg concentrations among all polar bears was explained by trophic feeding position as determined by baseline-uncorrected δ^{15}N ratios in hair (t = 9.44, p < 0.001, part C of Figure 2). Another

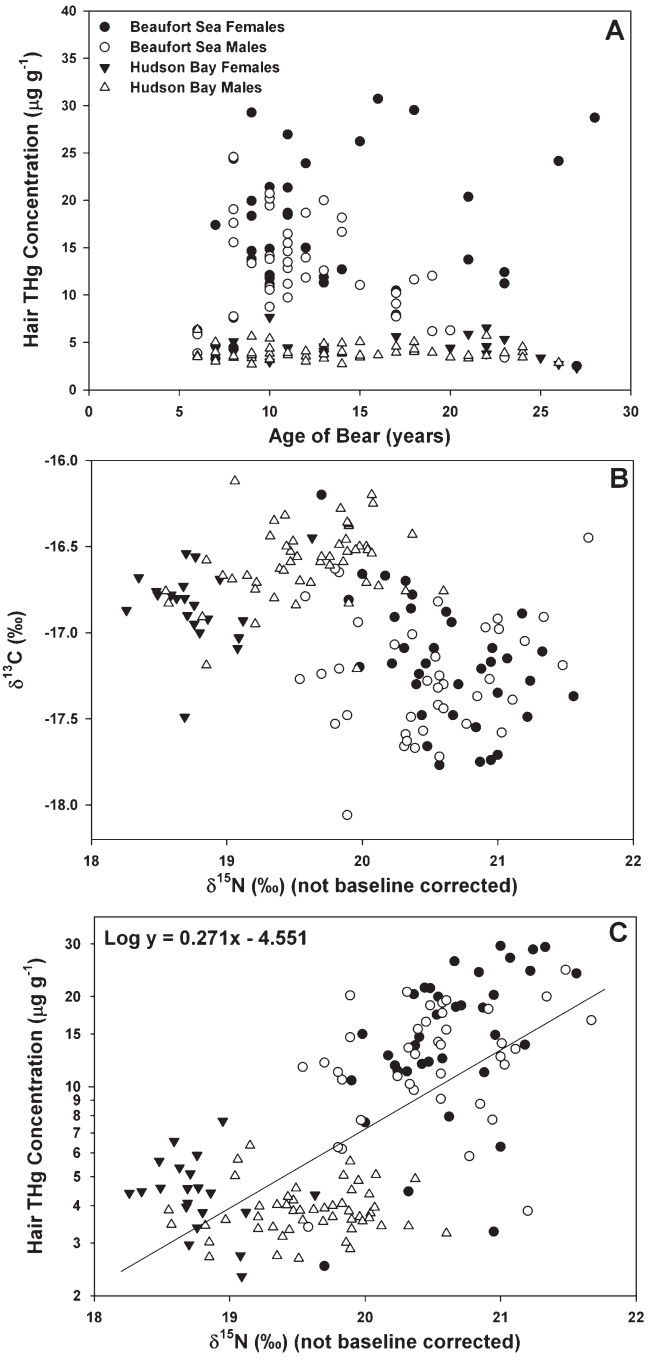


Figure 2. Total Hg (THg) concentrations (A) and δ^{15}N and δ^{13}C ratios (B) in hair of individual polar bears sampled from the southern Beaufort Sea and western Hudson Bay populations. C: The relationship between δ^{15}N and log THg concentration in polar bear hair. Note that δ^{15}N ratios have not been baseline corrected (Table 2).

31% of the variation in THg concentrations among polar bears was explained by δ^{13}C ratios ($t = -8.43, p < 0.001$). These results are consistent with the fact that pelagic food webs are often longer than benthic ones and that MeHg biomagnifies up the food web.

Where are the extra steps in the SBS polar bear food web occurring? We first examined the proportion of prey items in polar bear diets in combination with δ^{15}N ratios in those prey to determine if the lengthening of the SBS food web is occurring in the upper trophic level between polar bears and their immediate prey. The diet of polar bears in the SBS consists of approximately 64% ringed seal, 23% bearded seal, and 13% beluga whale.[14] Published baseline-uncorrected δ^{15}N ratios in the muscle of these prey items from the SBS region are 17.2 ± 0.7‰, 16.7 ± 0.9‰, and 16.9 ± 0.2‰, respectively.[33,34] Polar bears in WHB consume on average 67% ringed seal, 13% bearded seal, and 15% harbor seal.[14] Published baseline-uncorrected δ^{15}N ratios in the muscle of those prey items in WHB are 14.6 ± 0.3‰, 14.5 ± 0.3‰, and 17.2 ± 0.1‰, respectively.[35] Having bearded seal in the diet instead of beluga whale should result in a lengthening of the food web in WHB relative to that in the SBS and not vice versa. Furthermore, recent studies have shown that concentrations of THg in muscle tissues of ringed seals from the SBS (0.41−0.62 μg g^{-1} w.w., 36) were higher than those in muscle tissues of ringed seals from WHB (0.22 ± 0.07 μg g^{-1} w.w., 35), a pattern similar to that found in the hair of polar bears from these two regions. Thus, we propose that, in the SBS, the relatively more pelagic food web experienced by polar bears is longer either because of the presence of microbial activity in productive regions of the water column (i.e., a microbial loop) and/or a predatory zooplankton species, both of which are absent in the more benthic food web experienced by polar bears in WHB. This hypothesis warrants further scientific investigation.

Alternatively, there is some evidence that the length of the HB food web has recently been truncated. The relative abundance of fish species in northern HB, as determined from trends in fish fed to nestling thick-billed murres (*Uria lomvia*), appears to have changed over the past three decades due to concomitant climate warming and reductions in annual duration of sea-ice cover. Between 1984−1987 and 1998−2002, the mean proportion of Arctic cod (*Boreogadus saida*) in nestling diets fell from 43% to 15%, whereas capelin (*Mallotus villosus*) increased from 15% to 51%, and sand lance (*Ammodytes* spp.) from 3% to 15%.[37] Capelin (δ^{15}N, 11.7−13.0‰) and sand lance (δ^{15}N, 12.0‰) feed approximately half a trophic level lower than Arctic cod (δ^{15}N, 13.7‰) (ref 38 and data within). If ringed seal diet changed in a similar pattern as murres' diet in the HB region then, in recent decades, ringed seals, and hence polar bears, would be feeding at a lower trophic position than in the past. Because THg concentrations in polar bears are primarily explained by their trophic feeding position (e.g., part C of Figure 2), if the WHB food web has recently shortened, WHB bears should have had higher concentrations of Hg three decades ago than they do now (4.1 ± 1.0 μg g^{-1}), possibly resembling those of current SBS

Table 2. Published δ^{13}C and δ^{15}N Ratios in the Base of the Marine Pelagic and Benthic Food Webs in the Southern Beaufort Sea (SBS) and Western Hudson Bay (WHB)

	δ^{15}N (‰)	δ^{13}C (‰)	ref
SBS			
POM[a] (chlorophyll max, autumn 2003)	2.2 ± 1.8	−27.3 ± 0.7	31
POM (chlorophyll max, summer 2004)	3.5 ± 2.2	−23.4 ± 2.0	31
sediments (surface)	6.7 ± 0.2	−22.6 ± 0.7	31
WHB			
POM (surface waters, autumn 2005)	4.6 ± 0.8	−24.7 ± 1.3	32
POM (chlorophyll max, autumn 2005)	6.7 ± 1.4	−25.0 ± 1.7	32
sediments (surface)	7.8 ± 1.1	−21.8 ± 1.0	32

[a] Particulate organic matter.

bears ($14.8 \pm 6.6\ \mu g\ g^{-1}$). However, THg concentrations in hair collected from the WHB population in 1980 and 1988 (2.54 and 3.04 $\mu g\ g^{-1}$, respectively) were similarly lower than concentrations in hair collected from the BS population in those same years (18.54 and 9.6 $\mu g\ g^{-1}$).[21,22] These results suggest that any potential decline in food web length in HB in the past few decades has had little or no impact on THg concentrations in polar bears there.

Although differences in food web length likely explain a large percentage of the elevated $\delta^{15}N$ ratios and THg concentrations in SBS bears relative to WHB bears, both $\delta^{15}N$ ratios and THg concentrations can also be affected by body condition, age, and growth rates (e.g., ref 39). For example, organisms with faster, more efficient growth often have lower THg concentrations due to growth dilution (e.g., ref 40), whereas the relationship between $\delta^{15}N$ and body condition can be negative because of increased recycling of nitrogen within an organism's body during times of nutritional stress.[41] If organisms in the SBS food web grow slower and less efficiently than those in the WHB food web because it is annually colder and less productive there, this could result in higher THg concentrations and $\delta^{15}N$ ratios in apex predators like polar bears. Similarly, if productivity is higher at the base of the HB food web, biomass dilution should reduce the rate of MeHg biomagnification through the food web[42] resulting in lower THg concentrations in bears from WHB relative to those from the SBS. There is some evidence to suggest that this is the case. Recent measurements showed that, in the summer, pelagic primary productivity in HB (240−490 mg C m^{-2} d^{-1}) was higher than primary productivity in the BS (40−200 mg C m^{-2} d^{-1}) (ref 43 and data within). Higher primary productivity could conceivably lead to greater secondary productivity throughout the HB food web relative to that in the BS. In fact, significant differences in asymptotic size and growth rate have been found between the SBS and WHB populations of polar bears. For example, female polar bears in the WHB population reached 97% of their asymptotic body length in 4.1 years, whereas females in the BS region took 4.7 years to reach the same proportion.[44] However, it should be noted that earlier summer break-up of sea ice in WHB over the last 30 years [45,46] is resulting in declines in polar bear body condition, natality, survival, and population size there.[47−49] Therefore, further research is required to determine the exact relationship between the productivity of marine food webs, polar bear condition, and the bioaccumulation of MeHg.

Overall, stepwise regression showed that food web length/structure accounted for ~67% of the variation in hair THg concentrations among all polar bears in the two populations we studied. To our knowledge, the difference in trophic feeding positions between the two populations has not been previously recognized. However, the difference in concentrations of THg in polar bears between regions is likely also affected by availability of MeHg for bioaccumulation at the base of the food web.

2) Sources of Methylated Hg to the Base of Food Webs in the BS and HB. There are numerous sources of MeHg to polar oceans, including atmospheric deposition, export from major river systems, and in situ production in marine sediments and water columns.[13] Unfortunately, though, there is a paucity of published MeHg data for polar oceans. In the most comprehensive study published to date, Kirk et al.[50] measured concentrations of THg and methylated Hg species (the sum of both MeHg and dimethyl Hg) in marine waters of the Canadian Arctic Archipelago (CAA) and throughout HB in August to October 2005 (Figure 1). THg concentrations were low throughout the

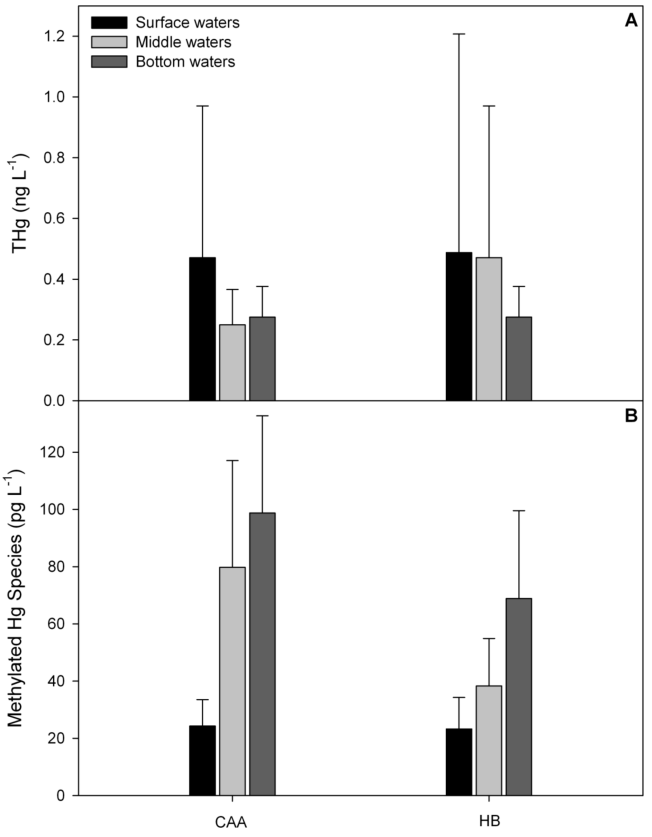


Figure 3. Average (± S.D.) concentrations of (A) total Hg (THg) and (B) methylated Hg species at the surface, bottom, and midway through the marine water column among sites in the Canadian Arctic Archipelago (CAA) and Hudson Bay (HB) (adapted from ref 50).

water column in both the CAA and HB (0.39 ± 0.40 ng L^{-1} and 0.42 ± 0.53 ng L^{-1}, respectively) (part A of Figure 3). At most sites in the CAA and HB regions, concentrations of methylated Hg species were low in surface waters, but higher at mid and bottom depths where water-column methylation of Hg(II) has been shown to occur.[13] Concentrations of methylated Hg species were similar in the surface waters of the CAA and HB regions (t = 1.234, p = 0.237), averaging only 24.3 ± 9.2 pg L^{-1} and 23.3 ± 11.0 pg L^{-1}, respectively (part B of Figure 3). However, at mid depths of the water column in the CAA, concentrations of methylated Hg species were 79.8 ± 37.3 pg L^{-1}, and increased to 98.8 ± 33.9 pg L^{-1} at the bottom (part B of Figure 3). Concentrations of methylated Hg species were also measured at one site in the SBS in 2007 (Figure 1) using sampling/analytical protocols described in Kirk et al.[50] and suggest that concentrations there are at least as high as those measured in the CAA (91 pg L^{-1} at the depth of chlorophyll maximum, and 140 pg L^{-1} at the oxycline; I. Lehnherr; unpublished data). In HB, concentrations of methylated Hg species were significantly lower than those in the BS (F = 20.06, p < 0.001), averaging 38.3 ± 16.6 pg L^{-1} midway through the water column, and 68.8 ± 30.7 pg L^{-1} at the bottom (part B of Figure 3). These data suggest that, although inorganic Hg(II) concentrations are similar among sites, overall there are higher concentrations of methylated Hg species available at the base of the pelagic marine food web for bioaccumulation in the SBS and CAA regions than in WHB. Unfortunately, neither MeHg concentrations, or the potential for sediments to methylate Hg(II), have been quantified in sediments

of the BS and HB, so we cannot determine if MeHg sources to the benthic food web also differ between these two regions. However, concentrations of THg in surface sediments are low in both the BS (averaging 10−100 ng g^{-1} d.w. depending on % clay in sediments, ref 51) and WHB (15−58 ng g^{-1} d.w., ref 52), and only a small fraction of this THg is likely comprised of MeHg suggesting that sediments would be minimal sources of MeHg to the base of benthic food webs at both sites. For example, in surface sediments from Chesapeake Bay and the mid-Atlantic continental margin, MeHg concentrations averaged only 1% of THg concentrations.[53]

Overall, we conclude that a longer, more pelagic food web and higher water column concentrations of methylated Hg in the BS resulted in higher concentrations of Hg in polar bears from the SBS population relative to those living along the western coast of HB. These findings have important implications for understanding the flow of not only energy and Hg but also other bioaccumulating and biomagnifying contaminants of concern, through marine food webs and into polar bears. Because polar bears are being heavily impacted by climate change throughout their circumpolar range (e.g., ref 49), understanding ways in which to minimize further risk from contaminants is of utmost importance.

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ACKNOWLEDGMENT

We thank G. Theimann, N. Morata, and Z.Z. Kuzyk for calculating specific averages from their published data for use here. Funding for this project was provided through the Natural Sciences and Engineering Research Council of Canada (NSERC), ArcticNet (a Network of Centres of Excellence of Canada), Canadian Wildlife Federation, Manitoba Conservation Sustainable Development Innovations Fund, Polar Bears International, Polar Continental Shelf Project, and World Wildlife Fund (Canada).

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