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Mercury bioaccumulation and biomagnification in a small Arctic polynya ecosystem

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HIGHLIGHTS

- Polynyas are recurring sites of open water in polar marine areas
- Mercury (Hg) biomagnification was studied in a small polynya near Nasaruvaalik Island, NU, Canada
- Hg biomagnification estimates for invertebrates to fish were low compared to other Arctic systems
- Factors underlying this result are unknown but may relate to primary productivity in small polynyas

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ABSTRACT

Recurring polynyas are important areas of biological productivity and feeding grounds for seabirds and mammals in the Arctic marine environment. In this study, we examined food web structure (using carbon and nitrogen isotopes, $\delta^{13}C$ and $\delta^{15}N$) and mercury (Hg) bioaccumulation and biomagnification in a small recurring polynya ecosystem near Nasaruvaalik Island (Nunavut, Canada). Methyl Hg (MeHg) concentrations increased by more than 50-fold from copepods (*Calanus hyperboreus*) to Arctic terns (*Sterna paradisaea*), the abundant predators at this site. The biomagnification of MeHg through members of the food web – using the slope of log MeHg versus $\delta^{15}N$ – was 0.157 from copepods (*C. hyperboreus*) to fish. This slope was higher (0.267) when seabird chicks were included in the analyses. Collectively, our results indicate that MeHg biomagnification is occurring in this small polynya and that its trophic transfer is at the lower end of the range of estimates from other Arctic marine ecosystems. In addition, we measured Hg concentrations in some poorly studied members of Arctic marine food webs [e.g. Arctic alligatorfish (*Ulcina olrikii*) and jellyfish, Medusozoa], and found that MeHg concentrations in jellyfish were lower than expected given their trophic position. Overall, these findings provide fundamental information about food web structure and mercury contamination in a small Arctic polynya, which will inform future research in such ecosystems and provide a baseline against which to assess changes over time resulting from environmental disturbance.

1. Introduction

Polynyas are ice-free areas of the polar oceans, which typically occur at the same location and time each year (Stirling, 1980). They are created by physical processes in the ocean and overlying atmosphere (e.g., currents, tidal action, wind), and are key zones of biological productivity in the Arctic marine environment (Stirling, 1997). As such, they are critical feeding grounds for seabirds and large mammals (Gilchrist and Robertson, 2000; Mallory and Gilchrist, 2005; Karnovsky et al., 2007). Because polynyas are highly productive, they are also important zones of contaminant transfer from the physical environment (e.g., atmosphere, water column, and seabed) into marine food webs. Indeed, bioaccumulation and biomagnification of mercury (Hg) and other pollutants have been documented in the large Northwater Polynya (NOW) of Baffin Bay (Fisk et al., 2001; Hobson et al., 2002; Campbell et al., 2005). However, little is known about contaminant uptake and trophic transfer through food webs in the multitude of small polynyas that occur throughout the high Arctic. This is a significant knowledge gap given the productivity of these areas, and uncertainty about how the occurrence and distribution of polynyas will be affected by changing sea ice conditions and sea level rise resulting from climate change.

Contaminants such as Hg can biomagnify to potentially harmful levels in Arctic marine food webs. Concentrations of methyl Hg

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(MeHg, the organic, biomagnifying form) may be exceptionally high in some wildlife (Braune et al., 2006), and often exceed toxicity thresholds in human inhabitants of the Arctic, whose traditional diets include top predators of these food webs (Kirk et al., 2012). Therefore, understanding the processes that affect the accumulation and biomagnification of Hg in Arctic marine ecosystems is essential. Biomagnification of this contaminant is studied in a variety of ways, including trophic magnification slopes (TMSs), that is, slopes of regressions of $\log_{10}$-Hg concentrations versus raw or baseline-corrected nitrogen isotope ratios, expressed as δ^{15}N (Kidd et al., 2012a; Lavoie et al., 2013). TMS values for MeHg in Arctic marine systems range from 0.13 in the eastern Canadian Arctic (van der Velden et al., 2013) to 0.34 in waters of the lower west coast of Greenland (Rigét et al., 2007). Extending from the TMS, trophic magnification factors (TMFs) are another means of assessing food web transfer of Hg, and are calculated as the antilog of the slope of $\log_{10}$-Hg concentrations regressed against discrete trophic levels (calculated from δ^{15}N assuming an average trophic enrichment factor, Δ^{15}N, through the food web; Borgå et al., 2012; Kidd et al., 2012a). Both TMS and TMF estimates in the literature vary considerably in terms of the organisms on which calculations are based. For instance, some studies include only fish and seabirds in their estimates (e.g., Jæger et al., 2009), whereas others looked at particulate organic matter through to apex predators such as polar bears (*Ursus maritimus*; Atwell et al., 1998). In addition, the inclusion of large-bodied animals (e.g. birds, mammals) typically requires that Hg analyses be conducted on muscle (or other individual tissues such as liver, as in Jæger et al., 2009) rather than on whole-body homogenates, which are more easily analyzed for smaller fishes and invertebrates. As such, estimates of Hg biomagnification often rely on a mix of muscle and whole-body Hg concentrations for organisms of different trophic positions.

In this study, we collected components of the seabird food web at a small polynya in the Canadian high Arctic to determine Hg concentrations and food web structure using stable carbon (δ^{13}C) and nitrogen isotope (δ^{15}N) analyses. Our goal was to determine whether Hg bioaccumulation and biomagnification (as indicated by TMSs and TMFs) in this polynya differed from larger polynyas or open water regions at high latitudes. As productive marine sites at high latitudes (Hoppema and Anderson, 2007; Lavoie et al., 2013), we predicted that the food

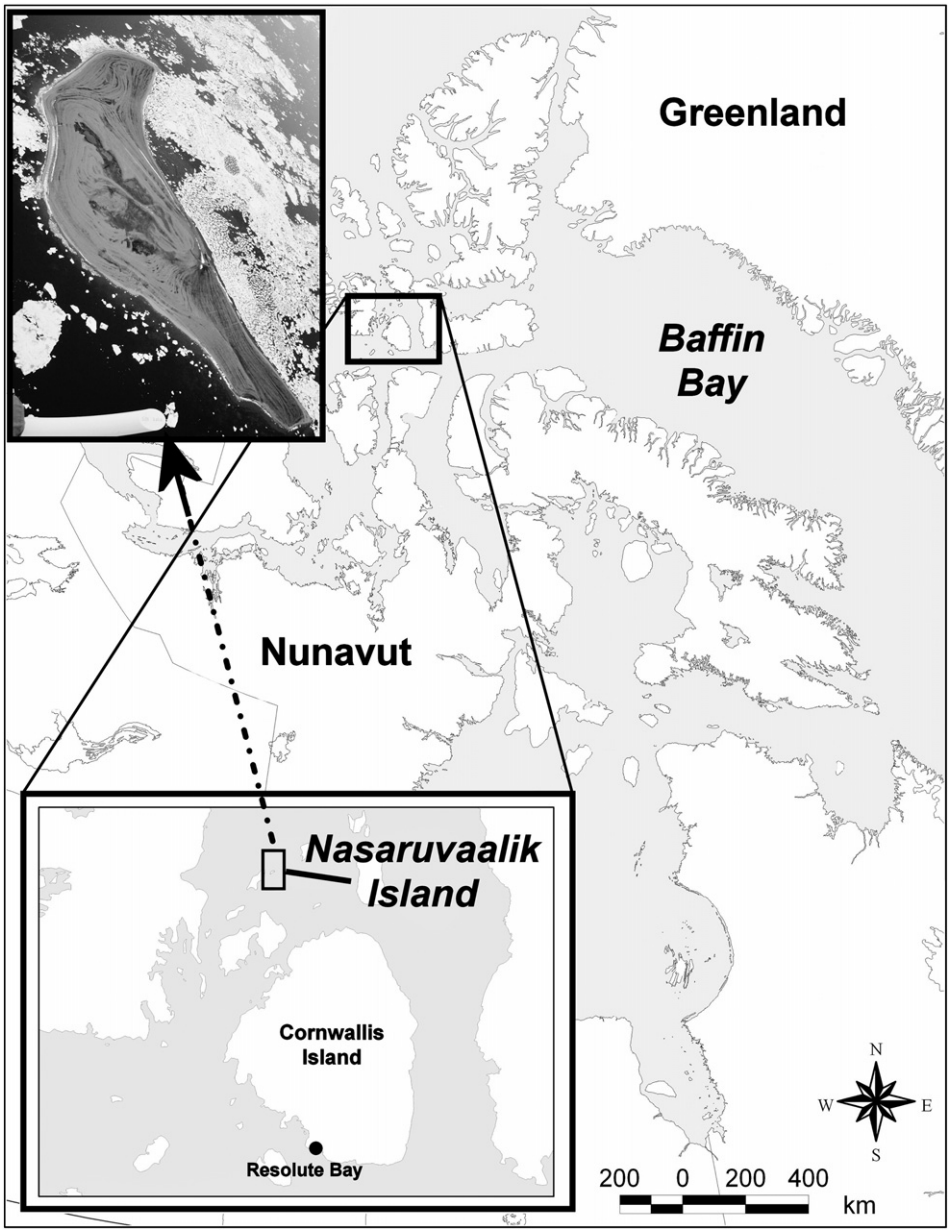

Fig. 1. Location of Nasaruvaalik Island (Nunavut, Canada) and recurring polynya (visible dark open water area immediately west of the gray island in the inset photograph).

Table 1
Mean (±SD) methyl and total mercury (MeHg and THg), proportion of MeHg (% of THg as MeHg), MeHg bioaccumulation factor (BAF) relative to seawater,[a] stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope values, trophic level (TL), and carbon and nitrogen contents (%C, %N) and carbon to nitrogen content ratio (C:N) for organisms of a small polynya near Nasaruvaalik Island (Nunavut, Canada), sampled June–August, 2011.

Organism	Tissue analyzed	*n*	MeHg (ng ng/g dw)	THg (ng ng/g dw)	%MeHg	MeHg BAF ($\times 10^6$)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	TL	%C	%N	C:N
Fishes												
Arctic alligatorfish (*Ulcina olrikii*)	Half body	1	119	366	32.6	1.98						
	Muscle	1	205	383	53.5	3.42	−19.55	13.81	3.39	46.5	13.3	3.5
Arctic cod (*Boreogadus saida*)	Half body	5	74 ± 41	270 ± 66	26.0 ± 9.4	1.22						
	Muscle	5	122 ± 73	363 ± 94	31.9 ± 12.3	2.03	−21.55 ± 0.46	13.31 ± 0.86	3.26 ± 0.23	49.6 ± 2.3	11.9 ± 0.8	4.2 ± 0.5
Seabirds[b]												
Arctic tern (*Sterna paradisaea*)	Egg	17		2109 ± 531				15.67 ± 1.03				
	Heart	2	879/946	1190/1240	73.9/73.6	15.2						
	Kidney	2	76/437	353/644	21.4/67.8	4.27						
	Liver	11	1170 ± 898	1443 ± 904	73.7 ± 16.3	19.5						
	Muscle	12	696 ± 734	907 ± 758	68.5 ± 16.8	11.6	−19.64 ± 1.28	13.85 ± 0.55	3.77 ± 0.14	50.3 ± 3.5	11.5 ± 1.2	4.5 ± 0.9
Common eider (*Somateria mollissima*)	Egg	14		876 ± 211				13.94 ± 0.43				
	Liver	2	496/882	769/1105	64.5/79.8	11.5						
	Muscle	2	675/1134	911/1410	74.0/80.4	15.1	−19.25/−19.07	13.57/14.44	3.70/3.93	50.7/52.7	10.9/11.3	4.6/4.7
Long-tailed duck (*Clangula hyemalis*)	Egg	4	850 ± 503					13.89 ± 0.66				
	Heart	2	220/815	509/1124	43.3/72.5	8.62						
	Kidney	1	335	527	63.5	5.58						
	Liver	3	477 ± 115	718 ± 130	65.9 ± 4.0	7.93						
	Muscle	3	364 ± 72	619 ± 55	58.5 ± 8.0	6.05	−18.00 ± 0.53	12.19 ± 0.52	3.34 ± 0.14	49.0 ± 1.8	12.7 ± 0.7	3.9 ± 0.1
Invertebrates												
Jellyfish (Scyphozoa)[c]	Part	3	12 ± 5			0.20	−21.87	13.24	3.24	36.9	7.9	4.7
Calanus hyperboreus	Whole body	2	11/15	182/258	6.0/6.2	0.22	−24.81/−24.46	8.28/8.74	1.94/2.06	52.1/59.1	6.0/6.1	8.5/9.9
Bulk crustaceans	Whole body	4	137 ± 26	384 ± 57	35.7 ± 3.6	2.28	−18.69 ± 0.91	13.11 ± 0.53	3.21 ± 0.14	37.9 ± 2.3	7.1 ± 0.5	5.4 ± 0.2
Gammaracanthus spp. (Amphipoda)	Whole body	3	44 ± 0.009	282 ± 14	15.9 ± 3.9	0.73	−19.88 ± 0.52	12.46 ± 0.30	3.04 ± 0.08	40.7 ± 4.2	6.9 ± 0.63	6.0 ± 0.9
Gammarus setosus (Amphipoda)	Whole body	12	27 ± 9	232 ± 58	11.8 ± 2.7	0.45	−18.74 ± 0.51	10.08 ± 0.39	2.41 ± 0.10	38.6 ± 1.9	7.6 ± 0.7	5.1 ± 0.5
Bulk krill (Amphipoda and Euphausiacea)	Whole body	1	43	352	12.3	0.72	−19.30	9.22	2.19	36.4	5.8	6.3
Other												
Saccharina latissima (kelp)	Part	5	3 ± 1	93 ± 19	3.8 ± 1.0	0.05	−18.85 ± 2.76	5.90 ± 1.49		32.2 ± 2.3	2.2 ± 0.5	15.1 ± 3.7

[a] BAF calculated as the ratio of MeHg in an organism to MeHg in seawater (0.06 ng/L).
[b] The muscle, heart, liver and kidney analyses were conducted on seabird chicks.
[c] N = 1 for jellyfish isotope, TL, %C, %N and C:N data; three separate jellyfish samples were analyzed for MeHg, but pooled to one homogenized sample for isotope analysis.

web at our small polynya would exhibit similar Hg biomagnification as the larger, well-studied sites like the North Water Polynya.

2. Methods

2.1. Field sampling

Sampling was conducted in a small, tidally-driven polynya on the west side of Nasaruvaalik Island in the Canadian Arctic Archipelago (75.8°N, 96.3°W; Nunavut, Canada; Fig. 1) in June–August of 2011. In this region, water flows south from ice-covered areas to the North, and is exposed to direct sunlight near the small polynya, and then flows under ice again (Mallory and Fontaine, 2004). Fish (Arctic cod, *Boreogadus saida*, and Arctic alligatorfish, *Ulcina olrikii*) were collected near nests when these prey items were dropped by marine birds feeding their chicks, and bird eggs or chicks (Arctic tern, *Sterna paradisaea*; common eider, *Somateria mollissima*; long-tailed duck, *Clangula hyemalis*; Sabine's gull, *Xema sabini*) were collected opportunistically from abandoned nests (most of which had been recently abandoned during sampling of adults for other studies). Field observations since 2007 indicate that these birds generally arrive at the polynya in mid-June and leave by mid-September, and that their foraging is concentrated immediately offshore or in other shallow areas of the polynya. Kelp (*Saccharina latissima*) samples were collected opportunistically along the shore. Marine aquatic macroinvertebrates (unidentified Medusozoa jellyfish, bulk krill consisting primarily of Amphipoda and Euphausiacea orders, bulk crustaceans, *Gammaracanthus* spp. and *Gammarus setosus* amphipods, *Calanus hyperboreus* copepods) were collected by taking sweeps with an aquatic dipnet along the island shoreline and around the edges of landfast sea-ice, consistent with the areas in which the birds' foraging was focused. Open-water zooplankton tows yielded few specimens. With the exception of eggs and chicks, all samples were gathered by personnel wearing nitrile gloves, and samples were placed in clean plastic storage bags and frozen at ~−20 °C within 2 h of collection, and remained frozen through transport to the Center for Analytical Research on the Environment (CARE) laboratory at Acadia University (Wolfville, NS, Canada). Water and melted snow samples (1 L) were collected from freshwater ponds or the ocean as shoreline grabs, using clean, sterile amber glass bottles with Teflon-lined caps, filled to maximum capacity and acidified to 1% HCl. Bottles were placed in the dark in a cooler and transported to CARE for analysis.

2.2. Mercury and stable isotope analyses

Total Hg and MeHg analyses were conducted on individual bird (muscle, liver, kidney, heart, and egg tissues), fish (muscle and whole-body homogenates) and jellyfish (whole-body homogenates) samples. Total Hg was measured in bird eggs as a proxy for MeHg, since generally >90% of THg in eggs is MeHg (Ackerman et al., 2013). Whole pooled bodies of all invertebrate samples were analyzed. Given the opportunistic nature of the sampling, invertebrate samples varied in their level of taxonomic grouping (e.g. samples of bulk krill or crustaceans compared to samples of *G. setosus* amphipods and *C. hyperboreus* amphipods; see Table 1 for more information about sample sizes and groupings). All biotic samples were freeze-dried and homogenized prior to Hg and stable isotope analyses. Methyl Hg and THg analyses were conducted at CARE. Approximately 10 mg (±0.01 mg) of freeze-dried tissue was digested following the established procedures (Cai et al., 1997; Edmonds et al., 2012). An aliquot of 20–40 μL of each digested sample was injected into a glass reaction bubbler for isolation and quantification of Hg species by aqueous phase ethylation, purge and trap, and gas chromatography followed by detection with atomic fluorescence spectrometry using a Brooks Rand Model III (US EPA, 2001, 2002). Method detection limits for MeHg and THg were 1 ng/g and 37 ng/g, respectively. Mean % recovery of MeHg and THg in certified reference materials was 83 ± 9% and 89 ± 12% ($n = 13$), respectively, in DOLT-4 (National Research Council of Canada); mean coefficients of variation between method replicates for MeHg and THg analyses were 6.9 ± 3.0% and 13 ± 5% ($n = 6$). All biotic Hg data are expressed in ng/g of dry weight (dw). THg and MeHg analyses of water and snow samples were also conducted at Acadia University (US EPA, 2001, 2002). Method detection limits for these analyses were 2.85 ng/L and 0.05 ng/L for THg and MeHg, respectively. The relative standard deviation among replicates was 16% for MeHg ($n = 3$ samples analyzed in triplicate) and 22% for THg ($n = 9$ samples analyzed in triplicate).

Stable isotope analysis of bird eggs is described in Akearok et al. (2010). Isotope analyses for all other taxa and tissues were done at the Stable Isotopes in Nature Laboratory (SINLAB, University of New Brunswick, Fredericton, NB, Canada). Approximately 0.20–0.35 mg of each dried sample was weighed into tin capsules and analyzed on a NC2500 Elemental Analyzer and Finnigan Delta Plus XP mass spectrometer as described in Clayden et al. (2013). Approximately 10% of samples were analyzed in triplicate; the coefficients of variation were within 3.4% for $\delta^{13}C$ and 1.7% for $\delta^{15}N$ ($n = 5$ samples analyzed in triplicate). Analysis of international standards for $\delta^{13}C$ and $\delta^{15}N$ (polyethylene IAEA-CH-7 and ammonium sulfate IAEA-N-2, respectively; International Atomic Energy Agency) resulted in relative differences from certified values of 1.1% for $\delta^{13}C$ and 0.2% for $\delta^{15}N$.

2.3. Data analysis

Total Hg and MeHg concentrations and %MeHg (ratio of MeHg to THg in each sample × 100%) of all organisms were $\log_{10}$-transformed to approximate normality (Kolmogorov–Smirnov tests, $\alpha \geq 0.15$), whereas the distributions of other variables ($\delta^{15}N$, $\delta^{13}C$, %N and %C) were not improved through transformation and left untransformed for statistical analysis.

The trophic position (TP) of each organism was calculated relative to the phytoplanktivorous copepod *C. hyperboreus* (with a TP of 2 and mean $\delta^{15}N$ of 8.5‰; Table 1) and assuming an average trophic fractionation factor ($\Delta^{15}N$) of 3.8‰ (Hobson and Welch, 1992) as follows:

$$TP_{consumer} = 2 + \left(\delta^{15}N_{consumer} - \delta^{15}N_{C.\ hyperboreus}\right)/3.8.$$

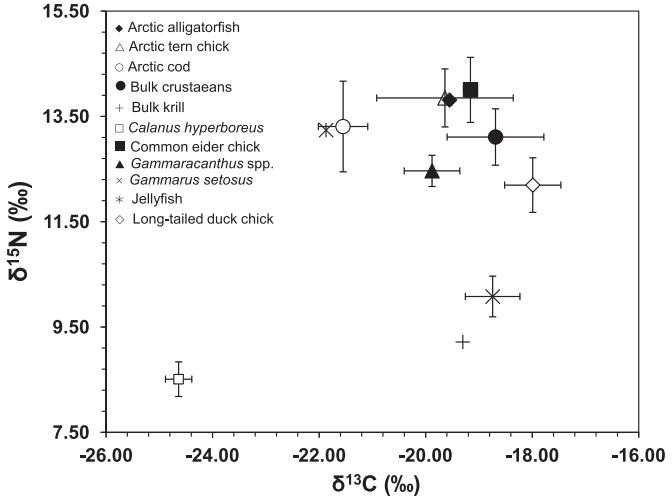

Fig. 2. Mean ± SD $\delta^{13}C$ and $\delta^{15}N$ (‰) values of invertebrates, fishes and seabirds from a small polynya food web near Nasaruvaalik Island (Nunavut, Canada).

For seabirds, however, $\Delta^{15}N$ is lower (approximately 2.4‰; Hobson and Clark, 1992), so TP for these organisms was determined as:

$$TP_{bird} = 3 + \left(\delta^{15}N_{bird} - 10.9\right)/3.8.$$

For further information about these calculations, see Fisk et al. (2001) and Hobson et al. (2002). MeHg bioaccumulation factors for each organism were calculated as the ratio of MeHg concentrations in a given taxon to the concentration of MeHg in seawater (see Table 1; Kidd et al., 2012a). Total Hg and MeHg biomagnification through members of the Nasaruvaalik polynya food web was calculated using TMSs and TMFs. MeHg and THg TMSs were determined as the regression slopes of log-MeHg or log-THg concentrations (ng/g of dry weight) versus $\delta^{15}N$ (‰) values of organisms. TMSs and TMFs were calculated for invertebrates through fishes (using whole body MeHg or THg concentrations for all organisms) and for invertebrates through seabirds (using muscle MeHg or THg concentrations of fishes and birds and whole-body concentrations of invertebrates). THg and MeHg TMFs were calculated using the antilog of the slope (b) of the regression of log THg (or MeHg) versus TP (Borgå et al., 2012), as follows:

$$\log_{10}Hg = a + b * TP, \quad \text{and} \quad TMF = 10^{b}.$$

To determine whether $\delta^{13}C$, %C or %N content explained additional variability in log-MeHg or log-THg concentrations of the organisms in the food web, multiple linear regression analyses were conducted with these additional variables. Models that included raw and lipid-corrected $\delta^{13}C$ values were evaluated. Lipid-correction followed the recommendations of Post et al. (2006) using the formula $\delta^{13}C = \delta^{13}C_{raw} - 3.32 + 0.99 \times C{:}N$. Models that included all possible subsets of variables were evaluated, and an Akaike Information Criterion (AIC) adjusted for small sample size (AIC_c) was used to assess each model; the best model was identified as the one with the lowest (most negative) AIC_c score (Burnham and Anderson, 2002). Collinearity among the independent variables was ruled out based on condition indices <15 (IBM SPSS 20 Statistics).

3. Results & discussion

3.1. Food web structure

Arctic tern chick muscle and eggs and Arctic alligatorfish muscle had among the highest $\delta^{15}N$ values of the organisms sampled in the Nasaruvaalik Island polynya food web (13.85‰ and 13.81‰, respectively; Table 1). Although the muscle $\delta^{15}N$ of this seabird species was similar to that of the alligatorfish in our sampling, the $\delta^{15}N$ of tern muscle varies annually and is often much higher at this site (Boadway, 2012) reflecting the dietary variability in this predator, and likely their higher trophic position as adults than that of fish. Jellyfish and bulk crustaceans were at a higher trophic position than other invertebrates (13.24‰ and 13.11‰, respectively, compared to 12.46‰ for amphipods, *Gammaracanthus* spp.).

In the Nasaruvaalik polynya ecosystem, values of $\delta^{13}C$ in biota ranged from −24.81‰ in *C. hyperboreus* to −18.00‰ in long-tailed duck chicks (Table 1, Fig. 2). Thus, *C. hyperboreus* were lower in $\delta^{13}C$ when compared to those of the NOW polynya located approximately 600 km to the east (~−21‰; Hobson et al., 2002; Campbell et al., 2005). These differences may be a function of different basal $\delta^{13}C$ values among these ecosystems. Amphipods from the nearby Barrow Strait, 200 km to the south, had $\delta^{13}C$ values within the range of those from the Nasaruvaalik polynya (−19.1‰ in Hobson and Welch, 1992, compared to −18.74‰ and −19.88‰ in *G. setosus* and *Gammaracanthus* spp., Table 1, Fig. 2). In seabirds from the current study, mean $\delta^{13}C$ values were similar for terns and eiders (−19.64 to −19.07, respectively) and slightly higher for long-tailed duck, but variation in $\delta^{13}C$ among samples was relatively high for terns and long-tailed ducks (Table 1; Fig. 2). Both of these species may have diverse diets reflecting their ability to exploit available prey resources (Robertson and Savard, 2002; Boadway, 2012), perhaps more so than benthivorous eiders. Nonetheless, these findings are also close to the mean values and range observed for seabirds in the Barrow Strait (−19.2‰ to −17.3‰; Hobson and Welch, 1992) and the NOW polynya (−19.9‰ to −18.7‰; Hobson et al., 2002). Likely reflecting the lower $\delta^{13}C$ at the base of the food web, Arctic cod collected from the polynya near Nasaruvaalik Island had slightly lower $\delta^{13}C$ values (−21.55‰) than others in the region (−18.9 ± 1.0 to −19.3 ± 0.1‰; Campbell et al., 2005; Hobson and Welch, 1992).

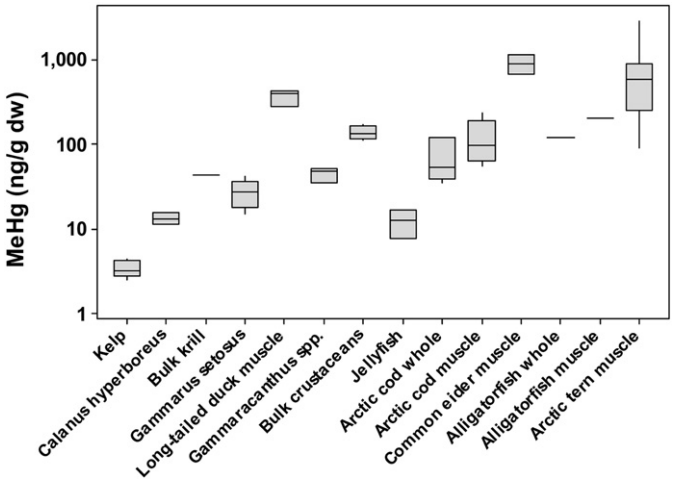

Fig. 3. Boxplot of MeHg (ng/g dry weight, dw) concentrations in organisms from a small polynya near Nasaruvaalik Island (Nunavut, Canada), in order of increasing mean $\delta^{15}N$ (see Table 1). Horizontal lines in boxes represent the median MeHg concentrations, whereas vertical bars represent the upper and lower 25% of the distribution of MeHg concentrations in each organism.

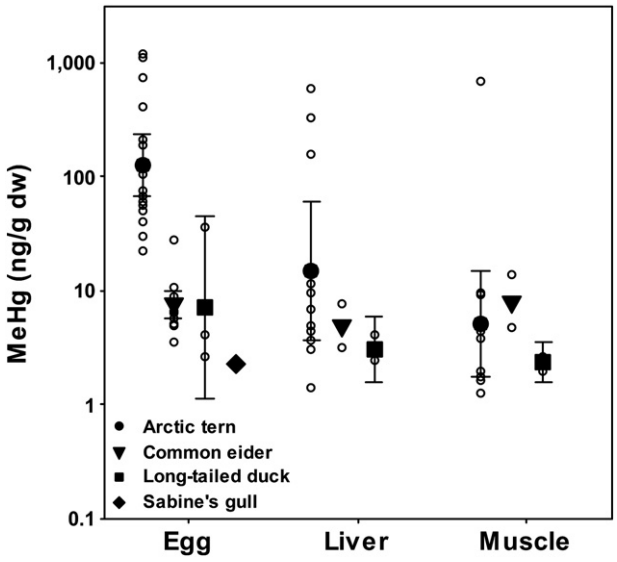

Fig. 4. Mean MeHg concentrations (ng/g of dry weight, dw) with 95% confidence intervals in the egg, liver and muscle of four seabird species collected around the Nasaruvaalik polynya. Open circles represent the individual values and confidence intervals are not shown where $n \leq 2$.

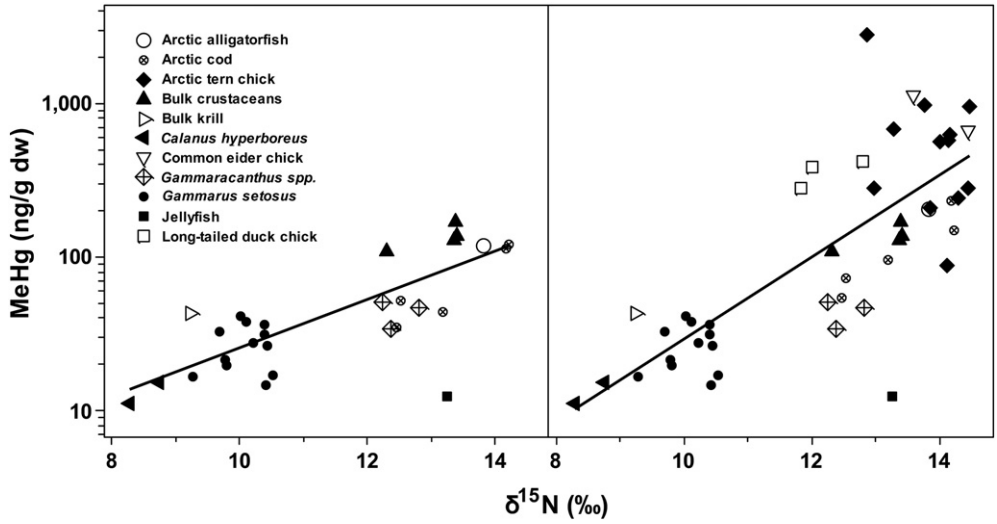

Fig. 5. Linear regressions of MeHg concentrations (ng/g of dry weight, dw) versus $\delta^{15}N$ (‰) in Nasaruvaalik organisms (left panel includes whole body MeHg concentrations of invertebrates and fishes, log-MeHg = $0.157 \times \delta^{15}N - 3.161$, $R^2 = 0.712$, $p < 0.001$, $n = 28$; right panel includes whole body concentrations of invertebrates and muscle concentrations of fishes and seabirds, log-MeHg = $0.267 \times \delta^{15}N - 4.199$, $R^2 = 0.642$, $p < 0.001$, $n = 45$). Jellyfish were not included in these regressions.

3.2. Hg bioaccumulation in Nasaruvaalik Island polynya organisms

Overall, kelp had the lowest MeHg concentrations of the organisms sampled in this study (3 ± 1 ng/g; Table 1, Fig. 3), and concentrations in these primary producers were approximately 50,000 times higher than the surrounding seawater (0.06 ng/L filtered; Table 1). This is lower than the bioconcentration seen in some freshwater primary producers (>100,000 times; Pickhardt and Fisher, 2007), perhaps because MeHg is more bioavailable in freshwater (van der Velden et al., 2013, and references therein). MeHg concentrations in freshwater ponds adjacent to the polynya were higher than those in seawater (0.16–0.59 ng/L filtered). MeHg bioaccumulation factors ranged up to 19,500,000 in Arctic tern livers relative to seawater (Table 1). Among invertebrates, mean MeHg concentrations ranged more than 10-fold (i.e., 11 ng/g in *C. hyperboreus* copepods to 137 ± 26 ng/g in bulk crustacean samples; Table 1). This range is approximately twice as large as that observed in zooplankton taxa of the much larger NOW polynya in Baffin Bay (approximately 5 to 50 ng/g assuming 80% moisture; Fig. 3a in Campbell et al., 2005), but similar to the range of 11 ng/g for *C. hyperboreus* to 142 ng/g in *Themisto libellula* from the same system (calculated from Table 7 in Campbell et al., 2005).

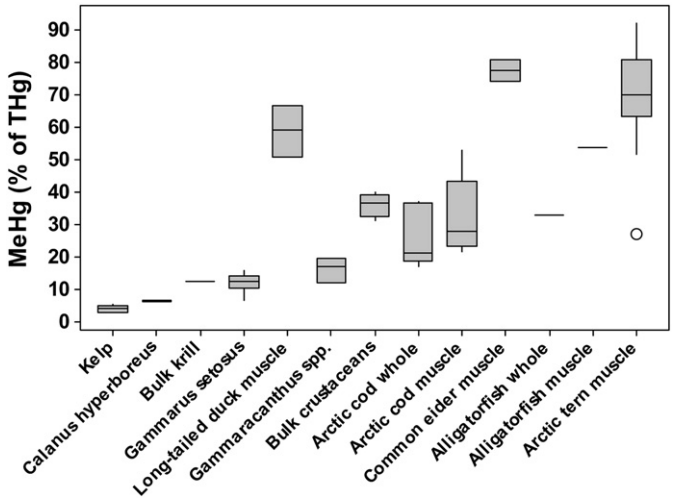

Fig. 6. Boxplot of %MeHg concentrations (MeHg as a percentage of THg) in organisms from a small polynya near Nasaruvaalik Island (Nunavut, Canada), shown in order of increasing $\delta^{15}N$ (see Table 1). Horizontal lines in boxes represent the median %MeHg, whereas vertical bars represent the upper and lower 25% of the distribution of %MeHg in each organism; open circles represent outliers (values >1.5 times the central 50% of the data distribution).

Data in our study were limited for fish, but Hg concentrations ranged less than two-fold in both muscle and body samples across species (Table 1). The muscle MeHg concentrations were generally lower than those seen in other marine fish species from western Greenland (190–250 ng/g; Rigét et al., 2007). However, Arctic cod in our study had almost twice as much THg in the muscle (363 ng/g) than this species in Lancaster Sound (190 ± 30 ng/g, Atwell et al., 1998).

Among seabird species collected near the small polynya, MeHg concentrations in the muscle differed by approximately two-fold (364 ± 72 ng/g in the muscle of long-tailed ducklings to 696 ± 794 ng/g in Arctic tern chicks; Table 1); this was consistent with known differences in THg between adults of these species (e.g., Mallory and Braune, 2012). Common eider ducklings in the current study had three times more mean MeHg in the muscle than did adult common eiders from Western Greenland (675–1134 ng/g, Table 1, Fig. 3, and 170 ± 170 ng/g in Rigét et al., 2007). The highest MeHg concentrations that we found in biota around Nasaruvaalik Island were in seabird livers (477 ± 0.115 ng/g to 1170 ± 898 ng/g in long-tailed ducklings and Arctic tern chicks, respectively) and eggs (Fig. 4). The range of hepatic THg in seabirds from the island (716–1443 ng/g) was generally lower than the reported values for seabirds elsewhere in this region (2640–9440 ng/g; Braune and Scheuhammer, 2008), but in this other study, liver samples were from adult birds, which would be expected to have higher Hg concentrations than chicks. These concentrations are below those considered a risk for wildlife health (Thompson, 1996).

Across seabird species, Arctic terns typically had the highest MeHg concentrations, which likely reflects their tendency to feed at higher trophic levels than eiders or ducks (e.g., Table 1; Akearok et al., 2010). However, between-species comparisons were not done because of small sample sizes available for some species and tissues. Overall, the relatively high concentrations of MeHg in seabirds in this study are consistent with studies showing that THg tends to be higher in seabirds in Arctic Canada than in the European Arctic (e.g., Braune et al., 2006; Miljeteig et al., 2009).

3.3. Biomagnification of mercury in the Nasaruvaalik polynya

Methyl Hg concentrations biomagnified between the lower and upper trophic levels of this polynya ecosystem (Table 1, Figs. 3, 5

Table 2
Trophic magnification factors (TMFs) and trophic magnification slopes (TMS) of methyl and total mercury (MeHg and THg) from different Arctic marine food webs.

Source	Location	Organisms	Contaminant	TMF	TMS	Method[a]
This study[b]	Nasaruvaalik Island (Nunavut)	*C. hyperboreus* — fish	MeHg	3.96	0.157	dw
		C. hyperboreus — fish	THg	1.37	0.036	dw
		C. hyperboreus — seabird	MeHg	7.65	0.267	dw
		C. hyperboreus — seabird	THg	2.13	0.095	dw
Atwell et al. (1998)	Lancaster Sound	POM — polar bear	THg		0.20	dw
Campbell et al. (2005)	NOW polynya (Baffin Bay)	Zooplankton — seal	MeHg		0.223	ww
			THg		0.197	ww
Foster et al. (2012)	Hudson Bay, Hudson Strait, Foxe Basin	Zooplankton	THg	1.1–1.9		dw
			MeHg	0.8–9.3		dw
Jæger et al. (2009)	Svalbard	Seabirds	THg	3.02	0.14	ww
		Fish — seabird	THg	4.87		
Loseto et al. (2008)	Beaufort Sea	Zooplankton — beluga	MeHg		0.25–0.31	dw
McMeans et al. (2010)	Iceland	Shrimp — shark	THg		0.26	dw
van der Velden	Eastern Canadian Arctic	Invertebrates — fish	THg	1.59–2.82 (2.25)	0.06–0.13 (0.10)	dw
			MeHg	2.72–5.70 (3.98)	0.13–0.22 (0.17)	dw
Rigét et al. (2007)	Western Greenland	Mussel — seal	MeHg		0.339	dw

[a] Concentrations of MeHg and/or THg determined on a dry or wet weight basis (dw and ww, respectively).
[b] *C. hyperboreus* — fish TMS and TMF were calculated using whole-body fish Hg concentrations, whereas *C. hyperboreus* — seabird TMS and TMF used muscle Hg of fish and seabirds.

and 6). However, although jellyfish were approximately one trophic position above copepods and are known to feed omnivorously on larval fish, other jellyfish and zooplankton (Martinussen and Bamstedt, 1999), their MeHg and THg concentrations were among the lowest of the organisms sampled (Table 1). Thus, MeHg concentrations in these organisms were inconsistent with the trend of increasing MeHg with trophic position (Figs. 3 and 5). Although it is not clear why jellyfish had lower MeHg than would be predicted by their trophic position, we speculated that it may be due to low protein content (Hsieh et al., 2001) relative to other secondary consumers, since MeHg tends to accumulate in proteinaceous tissues (Lemes and Wang, 2009). However, % nitrogen content (which is a proxy of total protein content; FAO, 2003) of jellyfish was comparable to or higher than that of other invertebrates (Table 1). Thus, other aspects of jellyfish elemental composition, respiration or metabolism may also affect their bioaccumulation of Hg. For instance, they have been shown to have faster growth rates and slower respiration rates than other marine invertebrates (Pitt et al., 2013). We are not aware of any other published MeHg data in jellyfish, and only one published estimate of THg (70 ng/g, *Mertensia ovum*; Atwell et al., 1998), but our findings indicate that these organisms are anomalous members of marine food webs with respect to their bioaccumulation and biomagnification of MeHg. This may have important implications for Hg dynamics and storage in food webs, since jellyfish may be proliferating or invading (and therefore becoming a more dominant source of biomass) in some marine food webs in association with climate- and/or pollution-driven ecological changes in the world's oceans (Mills, 2001).

The TMS of MeHg through the Nasaruvaalik polynya (from *C. hyperboreus* to fish) was 0.157 ($R^2 = 0.71$, $p < 0.001$, $n = 28$; Fig. 5; Table 2), which indicates that MeHg was biomagnified from invertebrates to fish. However, this value is lower than recent estimates of the average TMS for marine polar ecosystems (0.21; Lavoie et al., 2013), and lower than the MeHg TMS of the NOW polynya (0.22; Campbell et al., 2005). Not surprisingly, when seabirds were included in these regressions for the small polynya, the TMS for MeHg was higher (0.267, $R^2 = 0.642$, $n = 45$), but this may have been confounded by the fact that only muscle rather than whole-body Hg concentrations were available for birds, although this is consistent with TMS calculations for the NOW polynya (Campbell et al., 2005).

The TMS indicates that the biomagnification of MeHg in this polynya was at the lower end of the range observed for other Arctic marine food webs (0.14–0.34; Table 2). However, many of these other studies included muscle or other individual tissues of higher predators (e.g., polar bear, shark) in their calculations, which may account for the higher slopes they found compared to the current study. It is difficult to say what accounts for the variability that is seen across systems (Table 2) given that studies have different experimental designs and have included many different marine organisms in their calculations. A global review of food web studies found higher TMSs for aquatic systems at higher latitudes, although much of the variability in these data remained unexplained (Lavoie et al., 2013). Interestingly, the range of TMSs in Arctic marine systems spans the range of those from tropical to Arctic food webs (Table 2), which suggests that local or regional factors, such as water chemistry or physical characteristics of the sites (Kidd et al., 2012b; Clayden et al., 2013), may also affect Hg biomagnification across ecosystems. For instance, TMSs may be lower in systems with higher aqueous nutrients (Clayden et al., 2013). Thus, it is possible that the relatively high primary productivity of polynya ecosystems may help to explain why the TMS for the Nasaruvaalik polynya was somewhat low in invertebrates to fish compared to other Arctic marine environments.

When the TMS was determined using the regression of log THg versus $\delta^{15}N$, it was less than half of that for MeHg (slope = 0.095 with birds included, slope = 0.036 without birds). These slopes are also markedly lower than in other studies that have calculated Hg biomagnification through marine food webs using THg concentrations (see Table 2 and Lavoie et al., 2013). However, again these other studies included higher predators (e.g., polar bear, shark) and/or more mature organisms in their calculations (e.g., bird chicks rather than adults were included herein), which may account for the higher slopes they found compared to the current study. Also, studies with large-bodied animals have typically used muscle or other individual tissue Hg concentrations in their calculations (e.g., Rigét et al., 2007), which are generally higher in Hg than whole bodies, and TMS or TMF values may therefore be higher than they would be if whole bodies were used throughout the food web. Furthermore, the relatively high concentrations of both THg and MeHg in some invertebrates from our study (see Section 3.2) compared to low trophic level organisms such as bulk zooplankton from the NOW polynya (Tables 2 and 7 in Campbell et al., 2005) may have driven up the intercepts of the log-Hg versus $\delta^{15}N$ regressions, and thereby decreased the slopes (for discussion of the interdependence of slopes and intercepts, see Clayden et al., 2013; Lavoie et al., 2013). However, the sample sizes of key invertebrates such as copepods were low. Future studies in polynyas would benefit from larger sample sizes of all organisms, but particularly those at low trophic levels such as copepods, which represent an important proportion of the total biomass in these aquatic ecosystems. Moreover, recent

Table 3
Results of multiple regression analyses of log-MeHg and log-THg concentrations in Nasaruvaalik food web organisms (invertebrates and fishes) from a small polynya near Nasaruvaalik Island (Nunavut, Canada) with carbon[a] and nitrogen isotopes ($\delta^{13}C$ and $\delta^{15}N$), % carbon (%C), and % nitrogen (%N) of their tissues. The strongest model (with most negative AIC_c)[b] is shown in bold.[c]

Regression equation	R^2_{adj}	Standard error	AIC_c
MeHg			
$0.733 \times \delta^{15}N + 0.234 \times \delta^{13}C_{raw}$	0.559	0.229	−81.802
$0.874 \times \delta^{15}N + 0.313 \times \delta^{13}C_{lipid}$	**0.583**	**0.222**	**−83.458**
$0.774 \times \delta^{15}N + 0.513 \times \delta^{13}C_{lipid} + 0.321 \times \%N$	0.605	0.216	−83.451
$0.928 \times \delta^{15}N + 0.455 \times \delta^{13}C_{lipid} + 0.179 \times \%C$	0.584	0.222	−81.957
$0.827 \times \delta^{15}N + 0.658 \times \delta^{13}C_{lipid} + 0.322 \times \%N + 0.180 \times \%C$	0.607	0.216	−81.856
THg			
$0.482 \times \delta^{15}N$	**0.203**	**0.118**	**−117.042**
$0.659 \times \delta^{15}N - 0.287 \times \%N$	0.226	0.117	−116.457
$0.502 \times \delta^{15}N - 0.201 \times \%C$	0.214	0.118	−116.015
$0.478 \times \delta^{15}N + 0.119 \times \delta^{13}C_{raw}$	0.186	0.120	−115.047
$0.588 \times \delta^{15}N + 0.251 \times \delta^{13}C_{lipid}$	0.227	0.117	−116.467

[a] Models using raw or lipid corrected $\delta^{13}C$ values were evaluated, and results are shown for those models that met the selection criteria.[b]
[b] An Akaike Information Criterion adjusted for small sample size (AICc) was used to identify the best models (see Methods and Burnham and Anderson, 2002); models within ±2 of the model with the lowest AIC_c are also shown.
[c] Standardized regression coefficients are shown for each variable; $n = 34$ for MeHg models and $n = 33$ for THg models (no THg data available for jellyfish), $p < 0.001$ for all models.

findings suggest that *C. hyperboreus* is an important source of MeHg in Arctic marine food webs (Pućko et al., 2014), so further research on MeHg bioaccumulation and biomagnification in polynyas would do well to include extensive sampling of these zooplankton.

TMFs for MeHg were higher than those for THg in both *C. hyperboreus*-seabird and *C. hyperboreus*-fish food webs, and were 3.40 and 7.24 for MeHg and 1.36 and 2.10 for THg when excluding and including seabirds, respectively (Table 2). When comparing similar food webs, the TMFs for the Nasaruvaalik polynya fell within the range calculated for other systems; for example, the TMF for invertebrates to fishes for MeHg was 3.40 in the current study and between 2.7 and 5.7 in the eastern Canadian Arctic (van der Velden et al., 2013). However, because the data for Arctic marine systems are limited and varied in the biota that were analyzed, it was not possible to compare TMFs of Hg for invertebrates to birds across studies.

In general, %MeHg increased with increasing $\delta^{15}N$ (log-%MeHg = $0.114 \times \delta^{15}N - 0.098$; $R^2 = 0.845$, $p < 0.001$, $n = 33$; Table 1; Fig. 6), similar to MeHg concentrations (Fig. 3), again demonstrating the biomagnification of this form of Hg. Although most of the Hg in fishes is typically present as MeHg, %MeHg in Arctic cod from the Nasaruvaalik polynya was at least three times lower than that reported for the NOW polynya (Campbell et al., 2005). Percent MeHg in Arctic alligatorfish was also low compared to other Arctic fishes (Table 1; van der Velden et al., 2013). Muscle %MeHg in seabirds of the NOW polynya ranged from 75% in glaucous gulls (*Larus hyperboreus*) to 86% in ivory gulls (*Pagophila eburnea*; Campbell et al., 2005), which is higher than the range of 59% in long-tailed ducks to 77% in common eiders we observed in this study. These differences may arise from the opportunistic feeding habits of glaucous and ivory gulls, whose diets include more high-mercury items such as fish, carrion or waterfowl eggs and chicks (Mallory et al., 2008), compared to the eiders and long-tailed ducks in this study, for whom aquatic invertebrates are more important diet items (Goudie et al., 2000; Robertson and Savard, 2002).

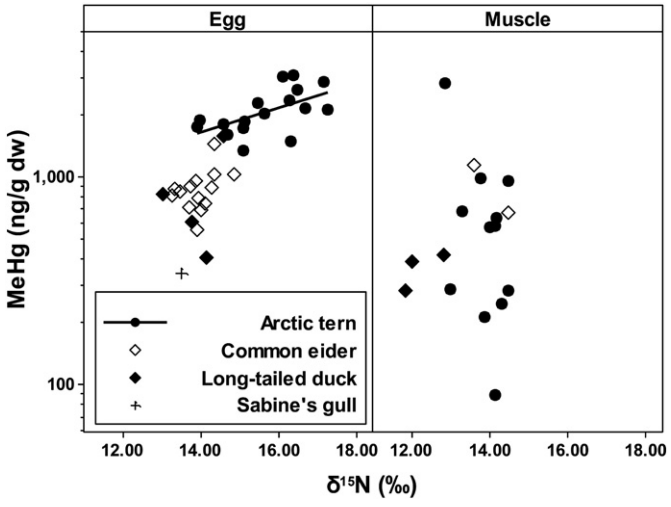

Fig. 7. Concentrations of MeHg (ng/g of dry weight, dw) versus nitrogen isotopes ($\delta^{15}N$, ‰) in egg and muscle tissue of four seabird species sampled around a small polynya near Nasaruvaalik Island (Nunavut, Canada). Linear regression analysis was only significant for Arctic tern eggs (log MeHg = $-0.618 + 0.0593 \times \delta^{15}N$, $R^2 = 0.33$, $p = 0.016$).

Among invertebrates and fish, MeHg concentrations were generally better explained by models that included lipid-corrected $\delta^{13}C$ in addition to $\delta^{15}N$, compared to those that included raw $\delta^{13}C$ (Table 3). Lipid-corrected $\delta^{13}C$ rather than raw values also explained slightly more variability in THg concentrations in these organisms (Table 3). These findings may be influenced by the high C:N ratios of some of the invertebrates, particularly copepods (Table 1). Such high ratios in these organisms may be indicative of relatively high lipid content and/or the dominance of exoskeleton in their overall body mass, but may also have been affected by the time of year of sampling and the limitations of mathematically correcting for lipid content (Syväranta and Rautio, 2010). Thus, future research could be helped by chemically extracting lipids from samples to better assess how $\delta^{13}C$ affects Hg bioaccumulation in polynya organisms. In addition, %C and %N of the organisms slightly increased the explanatory power of models of MeHg in fish and invertebrates (Table 3). As with $\delta^{13}C$, these relationships could partly be influenced by the organisms' lipid content. Moreover, MeHg concentrations in some freshwater food webs have also been shown to vary with the protein content of organisms, as inferred from %N (Clayden et al., 2013). Within individual taxa, $\delta^{13}C$ was a significant predictor of MeHg concentrations in Arctic terns (log-MeHg = $0.305 \times \delta^{13}C + 5.34$, $R^2 = 0.492$, $p = 0.011$), in addition to $\delta^{15}N$ (Fig. 7). Campbell et al. (2005) also found that $\delta^{13}C$ was a significant positive predictor of MeHg and THg concentrations across seabird species from the NOW polynya. These relationships may indicate that individuals feeding more in shoreline areas accumulate more Hg than those feeding in deeper waters, since littoral carbon sources (e.g., ice algae) typically have less negative $\delta^{13}C$ values than pelagic sources (e.g., particulate organic matter; Hobson et al., 2002). However, our findings from this small polynya and those of Campbell et al. (2005) from the larger NOW polynya differ somewhat from those of Choy et al. (2009), who showed a positive relationship between THg in fish

of the North Pacific Ocean and their depth of occurrence, suggesting that more pelagic fish had higher concentrations of this contaminant. It is not clear why littoral carbon sources in these Arctic polynyas might have higher MeHg. As such, more research on polynyas is needed to better understand how unique characteristics such as their morphometry affect the dynamics of Hg in both abiotic and biotic components of these ecosystems.

4. Conclusions

The analyses of biota from a small polynya in the Canadian high Arctic revealed that both MeHg and THg are biomagnified through this system. Hg transfer through this food web was higher when seabirds were included but, with or without seabirds, at the lower end of what has been observed in other marine food webs in the Arctic. The mechanisms underlying the lower trophic transfer of Hg in this small polynya remain to be understood, but may be linked to the higher productivity that is typical of polynyas. Future studies of contaminants in small polynya food webs would benefit from the collection of other primary producers in addition to kelp (e.g., ice algae, POM) to improve our understanding of how carbon sources affect Hg accumulation in consumers. Given recent findings that Hg trophic transfer estimates vary among systems in concert with their ecological characteristics, further research on other small polynyas would also facilitate among-system comparisons. This would, in turn, help to understand whether biological, physical or chemical characteristics affect Hg bioaccumulation and trophic transfer in these ecosystems. Finally, it is very likely that the distribution and occurrence of polynyas are being affected by climate change, so better characterization of the food webs and contaminant dynamics in these ecosystems will provide more information to assess the overall effects that climate change is having in Arctic environments.

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