Nitrogen and sulfur isotopes predict variation in mercury levels in Arctic seabird prey

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A B S T R A C T

Mercury (Hg) biotransformation and biomagnification are processes that affect Hg burdens in wildlife. To interpret variation in Hg in seabird eggs, used as Hg bioindicators in the Arctic, it is important to understand how Hg biomagnifies through the food web. We evaluated the use of $\delta^{34}S$, along with other commonly used stable isotope signatures ($\delta^{15}N$ and $\delta^{13}C$), for the determination of possible sources of Hg in an Arctic food web (56 individuals of 15 species of fish and invertebrates). Hg correlated with $\delta^{34}S$ ($R^2 = 0.72$). When the combined effects of $\delta^{34}S$ and $\delta^{15}N$ were considered in mixed-effects models, both $\delta^{34}S$ and $\delta^{15}N$ together described Hg patterns in Arctic food webs better than either isotope alone. Our results demonstrate the usefulness of $\delta^{34}S$ to account for variation in Hg among marine animals and to study the possible underlying effects that MeHg production may have on Hg pathways in Arctic ecosystems.

1. Introduction

Human activity directly and indirectly produces multiple sources of pollutants that are affecting and modifying the environment (Persson et al., 2013; Rockström et al., 2009). For example, mercury (Hg) deposition has increased three-fold since preindustrial times (Driscoll et al., 2013; Selin, 2009). Currently, 1900–4000 t are released into the atmosphere every year from human primary sources (Driscoll et al., 2013; Selin, 2009). Hg poses a special threat to polar regions (Fort et al., 2014; Kirk et al., 2012). Hg speciation between its volatile and deposited forms can lead to processes such as global distillation and mercury depletion events during which Hg is transferred from equatorial or temperate regions into polar environments (Ariya et al., 2004; Braune et al., 2015; O'Driscoll et al., 2005; Rigét et al., 2011; Skov et al., 2004). Between 208 and 305 tons of Hg are deposited each year in the Arctic due to these types of processes despite local emissions in the region being low (Ariya et al., 2004; Skov et al., 2004). Mercury is most toxic in its organic form, methylmercury (MeHg), which is also easily assimilated and bioaccumulated by organisms (Dietz et al., 2013; Liu et al., 2008).

Animals, particularly seabirds, can be used to monitor contaminants in ecosystems because they can integrate signals over large foraging areas and return to a central site (colony) where they can be sampled relatively easily (Elliott and Elliott, 2013, 2016; Furness and Camphuysen, 1997). Dietary analyses can be used to determine whether increasing Hg concentrations in animal tissue are due to an increase of available MeHg in the ocean or due to changes in the animal's feeding habits (Elliott and Elliott, 2016; Kidd et al., 1995; McKinney et al., 2010, 2015). Stable isotope ratios have been widely used as indicators of trophic level and feeding location when quantifying Hg in tissues (Atwell et al., 1998; Nisbet et al., 2002; Overman and Parrish, 2001; Vo et al., 2011).

The $\delta^{15}N$ ratio (^{15}N to ^{14}N, expressed in relation to an international isotopic reference) is an index of the relative trophic position of an organism as ^{15}N content increases with trophic level (Carr et al., 2017; Hobson et al., 1994). As differences in dietary trophic level can explain significant proportions of Hg variation, especially when temporal trends are being studied, correcting for the $\delta^{15}N$ ratio can help to control for this confounding effect (Fig. 1: 'trophic position hypothesis'; Bentzen et al., 2016; Kidd et al., 1995; McKinney et al., 2012; Vo et al., 2011). Another commonly used isotope signature is the ratio of ^{13}C to ^{14}C, $\delta^{13}C$ (as expressed relative to an international isotopic reference), which can describe changes in food sources associated with habitat to a greater degree than trophic level. For example, benthic feeding organisms are enriched in ^{13}C compared to pelagic feeders (Carr et al., 2017; Hobson et al., 1994; Nisbet et al., 2002) and terrigenous organic carbon is associated with lower $\delta^{13}C$ values compared to marine carbon (Foster et al., 2012; Schell et al., 1998). Differential feeding from

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Trophic Position Hypothesis

Sulfate Availability Hypothesis

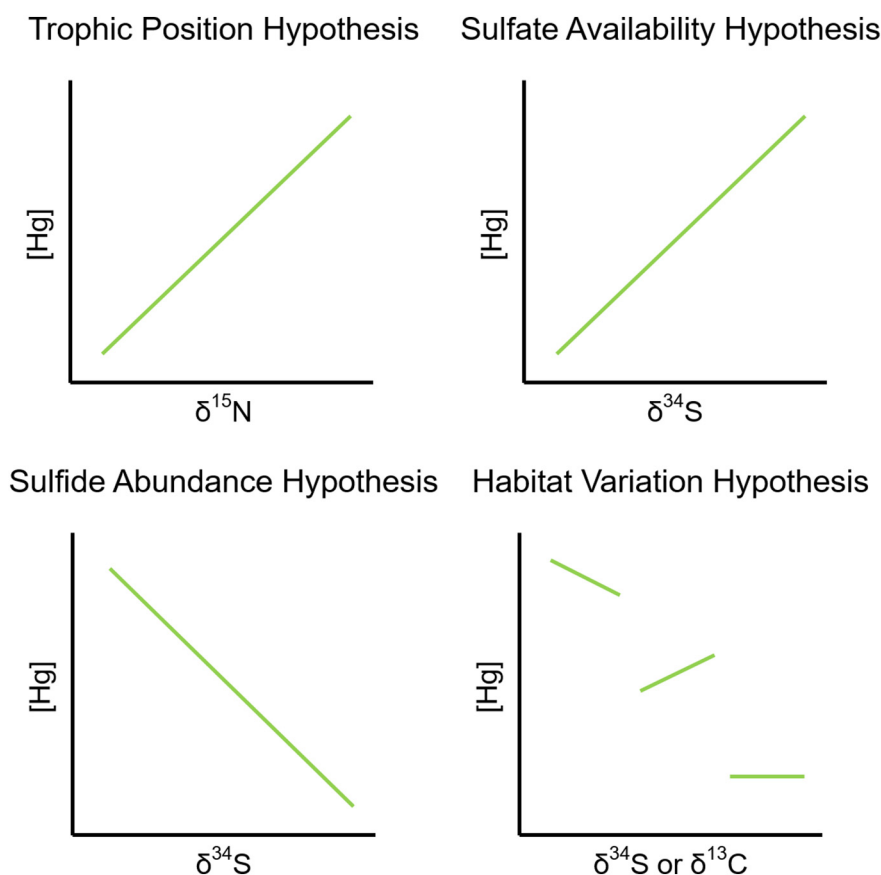## Sulfide Abundance Hypothesis

Habitat Variation Hypothesis

Fig. 1. Hypotheses for relationship between Hg and isotopes tested within this paper: trophic position hypothesis, sulfate availability hypothesis, sulfide abundance hypothesis, and habitat variation hypothesis.

diverse habitats may cause variation in the Hg concentrations uptaken (Fig. 1: 'habitat variation hypothesis'). Specifically, if Hg levels are higher in one habitat (i.e. benthos) than another habitat (i.e. pelagic waters), due to long range transport or other mechanisms, then an association between Hg and isotopic values might be expected. For example, fish that were originally river residents had higher Hg than other fish caught in the same rivers and that migrated from a lake; $\delta^{13}C$ and $\delta^{34}S$ (see below) could distinguish between the river residents and the lake migrants (Carr et al., 2017). These commonly used ratios can help to elucidate observed differences in Hg concentrations that are the result of the animal's feeding habits but fail to account for an important aspect of the mercury cycle that is independent of top predator's life history, the production of MeHg.

Sulfate-reducing bacteria (and iron-reducing or methanogenic bacteria, to a lesser extent) are the main drivers of the conversion of inorganic Hg into MeHg in many environments (Driscoll et al., 2013; Morel et al., 1998; Selin, 2009). These bacteria, which inhabit anoxic aquatic environments, use sulfate as the final electron acceptor for respiration and can methylate Hg during the process, presumably by the involvement of the acetyl-coenzyme A pathway (Parks et al., 2013; Pollman and Axelrad, 2014; Selin, 2009). The central role played by the sulfate-reducing bacteria suggests that the production of MeHg is not limited by the initial concentration of inorganic Hg but rather by sulfate concentrations and, thus, sulfate reduction rates. The addition of an inhibitor of sulfate reduction to anoxic sediments led to an almost complete reduction of MeHg production (Compeau and Bartha, 1985). Similarly, no methylation occurred in cultures of sulfate-reducing bacteria where no sulfate was added (King et al., 2000). Increased concentration of available sulfate in sediments and lakes led to an increase in MeHg production (Gilmour et al., 1992). As these bacteria respire, sulfur in the water column is converted from sulfate to sulfide

causing the remaining sulfate to become enriched in the heavier sulfur isotope, ^{34}S (Krouse and Mayer, 2000; Peterson and Fry, 1987). This makes the ^{34}S to ^{32}S ratio as expressed relative to an international isotopic reference, $\delta^{34}S$, useful for the detection of sulfate reduction and, thus, mercury methylation via sulfate-reducing bacteria, so that variation in environmental MeHg levels can be accounted for (Fig. 1: 'sulfate availability hypothesis'; Elliott and Elliott, 2016). Conversely, if sulfate-reducing bacteria are limited by factors other than sulfate, we may expect a negative relationship between $\delta^{34}S$ and Hg (Carr et al., 2017; Fry and Chumchal, 2012). High levels of sulfide associated with low $\delta^{34}S$ values could be indicative of the active presence of sulfate-reducing bacteria (which, if limited by factors other than sulfate abundance, would convert most sulfate into sulfide) leading to higher methylation rates and thus, higher Hg bioaccumulation potential (Fig. 1: 'sulfide abundance hypothesis').

Thick-billed murres (*Uria lomvia*) are a key monitoring species for mercury in the Canadian Arctic (Braune, 2007; Braune et al., 2016). A recent study showed that variation in Hg among both thick-billed murres and their prey was associated with variation in $\delta^{15}N$ (Braune et al., 2014a, 2014b). A dietary shift towards prey lower in $\delta^{15}N$ masked an increase in Hg across time, and accounting for variation in $\delta^{15}N$ improved assessment of Hg trends over time (Braune et al., 2014b). Our study aims to compare the usefulness of $\delta^{34}S$, along with $\delta^{15}N$ and $\delta^{13}C$, for the determination of Hg bioaccumulation sources in Arctic food webs. To this end, we sampled individuals from 15 species of fish and invertebrates that are common prey of thick-billed murres breeding at Coats Island in northern Hudson Bay. We predicted that Hg levels would be associated with both sulfate availability ($\delta^{34}S$) and trophic position ($\delta^{15}N$).

2. Materials and methods

2.1. Sample collection and preparation

Representative samples of small fish and invertebrates were collected opportunistically from the breeding ledges of thick-billed murres at Coats Island (62°98′N, 82°00′W) in northern Hudson Bay, Nunavut, Canada (see Braune et al., 2014a for details). Fifty-six individuals from 15 species were collected between 2007 and 2009. The samples were a subsample of those analyzed as part of Braune et al. (2014a), and Hg and $\delta^{15}N$ data used in this study were presented in that paper. Species sampled included Arctic cod (*Boreogadus saida*), capelin (*Mallotus villosus*), sand lance (*Ammodytes* spp.), Atlantic poacher (*Leptagonus decagonus*), Arctic shanny (*Stichaeus punctatus*), daubed shanny (*Leptoclinus maculatus*), banded gunnel (*Pholis fasciata*), fish doctor (*Gymnelus viridis*), fourline snake blenny (*Eumesogrammus praecisus*), sculpin (*Cottidae*), snailfish (*Liparis* sp.), sea butterfly (*Clione limacina*), squid (*Gonatus fabricii*), gammarid (*Gammaridae*), euphausiids (*Euphausiacea*), and jellyfish (*Medusozoa*). Fresh fish and invertebrates collected from the ledges were identified and measured as described by Elliott and Gaston (2008). Samples were washed, individually wrapped in foil, placed in plastic bags and frozen at −20 °C in the field before being shipped to the National Wildlife Research Centre (NWRC), Ottawa, Ontario, where they were stored at −40 °C prior to chemical analysis. Fish were analyzed either individually or as composite samples (pools) comprised of 2–8 fish (see Braune et al., 2014a). Pooled samples were created by taking equal aliquots from each fish. In some cases, only sagittal sections were available after the other half of the fish was used for fatty acid analyses, but it was assumed that the sagittal sections were representative of the whole fish.

2.2. Mercury and stable isotope analysis

For Hg quantification, samples were homogenized, freeze-dried, homogenized again, and weighed into nickel combustion boats. Total Hg was analyzed using an Advanced Mercury Analyzer (AMA-254) equipped with an ASS-254 autosampler for solid samples as described elsewhere (EPA Method 7473; see Braune et al., 2014a for details). Stable isotope analyses for $\delta^{34}S$, $\delta^{15}N$, and $\delta^{13}C$ were performed on the homogenate at the G. G. Hatch Stable Isotope Laboratory (Ottawa, ON; $\delta^{15}N$, $\delta^{13}C$, and all $\delta^{34}S$) or University of Winnipeg Stable Isotope Laboratory (Winnipeg, MB; only $\delta^{15}N$ and $\delta^{13}C$), with no significant inter-lab variability as reported by Braune et al. (2014b). Homogenates were freeze-dried and powdered. Lipids were removed using a 2:1 chloroform: methanol soak and rinse. Stable nitrogen and carbon isotope assays were performed on 1 mg subsamples of homogenized material loaded into tin cups. Samples were analyzed using an isotope cube elemental analyzer (Elementar, Germany) interfaced with a Delta Advantage continuous-flow isotope ratio mass spectrometer (Thermo, Germany) coupled with a ConFlo III (Thermo, Germany). A glutamic acid laboratory standard was included for every 10 unknown samples. Quality control was maintained by running sample duplicates. All measurements are reported in standard δ-notation in parts per thousand (‰) relative to the AIR international standard. Replicate measurements of internal laboratory standards [C-55 (glutamic acid)] indicated measurement error of ± 0.2‰, respectively. Stable sulfur isotope analyses were performed on 10 mg subsamples of the homogenized fish loaded into tin capsules (lipids were not extracted from homogenates as lipid extraction is known to alter $\delta^{34}S$ and lipids should not contain sulfur; Elliott et al., 2014). Samples were analyzed with an isotope cube elemental analyzer (Elementar, Germany) interfaced with a Finnigan DeltaPlus XP isotope ratio mass spectrometer (Thermo Germany) coupled with a ConFlo IV (Thermo Germany). All measurements are reported in parts per thousand (‰) relative to the VDCT international standard. Calibrated internal standards were used to normalize the data with a precision of ± 0.4‰.

2.3. Statistical analyses

Statistical analyses were conducted in R 3.3.2 using linear models and linear mixed-effect models with Hg concentrations (log-transformed) as the dependent variable and isotope ratio values as the independent variables. Linear and mixed-effect models with single isotopes (only $\delta^{15}N$, $\delta^{34}S$, and $\delta^{13}C$) and groupings of two and all three isotopes ($\delta^{15}N + \delta^{34}S$; $\delta^{15}N + \delta^{13}C$; $\delta^{34}S + \delta^{13}C$; and $\delta^{15}N + \delta^{34}S + \delta^{13}C$) were included and ranked using Akaike's Information Criterion, adjusted for small sample sizes (AIC$_c$; Burnham and Anderson, 2002). With the linear models, for species which more than one individual was collected, we used a single average data point, which was averaged across all individuals of a given species; data for species with a single collected individual were also included in these models. With the mixed-effect models, data were not pooled and species was included as a random effect. We report *P*-values for all top models. The top 6 AIC$_c$ of the mixed-effect models with centralized predictors were used to develop a conditional model average (Grueber et al., 2011). Model averaging can rank and weigh these models to obtain quantitative measures of relative support of each of the used model to a general, averaged model (Grueber et al., 2011).

3. Results

All three isotopes correlated weakly with each other (Fig. 2). $\delta^{13}C$ and $\delta^{15}N$ were positively correlated each other ($t_{54} = 4.77$, $P < 0.00002$). $\delta^{34}S$ was negatively correlated with $\delta^{13}C$ ($t_{54} = -3.21$, $P < 0.003$) and with $\delta^{15}N$ ($t_{54} = -3.46$, $P < 0.002$). Using an average species value of the raw $\delta^{15}N$ data (raw data available at: Góngora et al., 2018), we observed that banded gunnels, poachers, fourline snake blennies, and Arctic shannies occupied the highest trophic position in the studied food web, and sea butterflies, gammarids, and jellyfish occupied the lowest. $\delta^{13}C$ and $\delta^{34}S$ (average value per species) imply that the species more associated with pelagic feeding habits were sea butterflies and jellyfish, and that the species with more benthic feeding habits included euphausiids, fourline snake blennies, daubed shannies, and poachers. However, the habitat assignation obtained from the values for those two isotopes were not always in accordance with each other; $\delta^{13}C$ characterized sand lance as pelagic feeders while $\delta^{34}S$ characterized them as benthopelagic feeders, for example. All data are archived at Góngora et al. (2018) and included in the Supplementary material.

For the data pooled by species (linear models), Hg variation was best explained by $\delta^{34}S$ ($t_{15} = -6.20$, $P < 0.00002$) in accordance with the AIC$_c$ model selection (Table 1), as Hg decreased with increasing $\delta^{34}S$ (Fig. 3). AIC$_c$ values and weights for the non-pooled data (mixed-effect models) did not show strong statistically significant evidence for the selection of a model (Table 1) but models including $\delta^{13}C$ were discarded as this regression coefficient was not statistically significant. The averaged mixed-effect model showed that the $\delta^{15}N$ and $\delta^{34}S$ ratios were both important in explaining Hg variation. Hg concentrations increased as $\delta^{15}N$ values increased (averaged mixed-effect model, $z = 2.96$, $P_{|z|} < 0.004$) and decreased with increasing $\delta^{34}S$ (averaged mixed-effect model, $z = 2.40$, $P_{|z|} < 0.02$).

4. Discussion

The $\delta^{34}S$ ratio consistently explained variation in Hg levels, both across and within species. We observed a negative relationship between the $\delta^{34}S$ and Hg concentrations which is consistent with the sulfide abundance hypothesis (Fig. 1). $\delta^{15}N$ also explained variation in Hg levels when all individual samples were included (mixed-effect models), in support of the trophic position hypothesis. Thus, across an Arctic food web from zooplankton to predatory fish, trophic position and sulfide abundance (an index of the abundance of sulfate-reducing bacteria and their activity) together predicted levels of Hg. We urge the
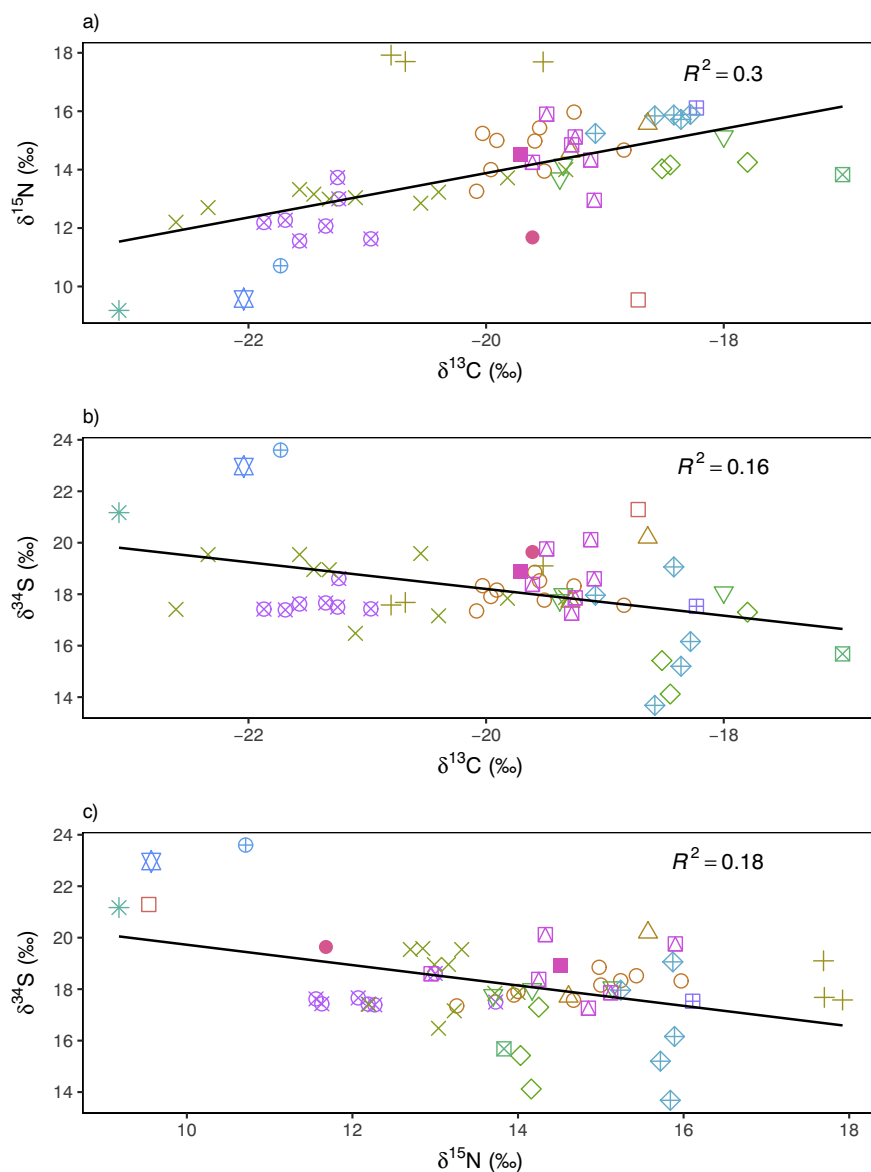
Fig. 2. Relationships between a) δ^{13}C and δ^{15}N, b) δ^{13}C and δ^{34}S, and c) δ^{15}N and δ^{34}S for Arctic fish and invertebrates from Coats Island. Data from all (N = 56) individual samples was included and was not pooled by species. See Fig. 3 for species legend.

Table 1
Statistical output for models of Hg variation either pooled or not pooled by species.

	df	ΔAIC_c	AIC_c weight
Linear model (pooled by species)			
Null	2	18.604	9.09×10^{-05}
δ^{13}C	3	12.952	0.00153395
δ^{15}N	3	12.304	0.00212078
δ^{34}S	3	0	0.99625437
Mixed-effect model (not pooled by species)			
Null	3	7.4295	0.01774927
δ^{15}N	4	2.4844	0.21036928
δ^{34}S	4	5.6455	0.04330757
δ^{15}N + δ^{34}S	5	0	0.72857389

inclusion of δ^{34}S, alongside the commonly used δ^{15}N, to predict levels of Hg in Arctic ecosystems.

Significant relationships between Hg and δ^{34}S or δ^{15}N have also been observed for birds, fish, and invertebrates from marine, estuarine, and freshwater environments (Table 2). An explanation for the

consistent relationships with δ^{34}S (8 out of 8 studies showing significant relationships, Table 2) compared with δ^{15}N (4 out of 8 studies showing consistent significant relationships for all sites/samples/species included; studies with variable results of positive and negative were excluded) is that previous studies of marine ecosystems were generally focused on a species or multiple species that are at relatively the same trophic level (Bentzen et al., 2016; Elliott and Elliott, 2016; Ramos et al., 2013). Thus, differential Hg accumulation is more likely caused by other factors (e.g. sulfate availability and sulfide abundance) when trophic levels are similar. Elliott and Elliott (2016) present an example of how δ^{34}S can explain patterns in Hg accumulation when δ^{15}N cannot; seabirds feeding further offshore, with the highest levels of δ^{34}S, had the highest Hg concentrations compared to other seabirds feeding nearshore. Thus, species such as Leach's storm-petrel (*Oceanodroma leucorhoa*) had high levels of Hg despite having low trophic position (Table 2). In that study, both Hg and δ^{34}S declined over time in cormorants despite no variation in δ^{15}N. If one only accounted for δ^{15}N and not δ^{34}S, researchers would have concluded that Hg was decreasing when, in actuality, it was only diet that had changed. Another example of the future potential δ^{34}S as a complement of δ^{15}N in the study of Hg
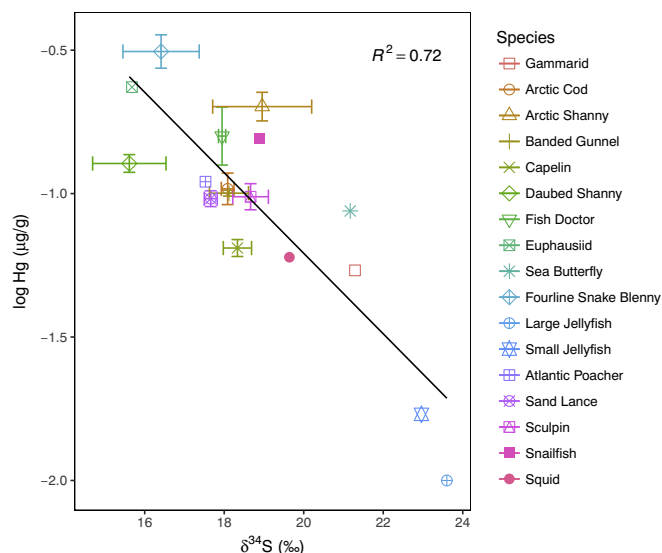
Fig. 3. Log Hg decreased with $\delta^{34}S$ for samples averaged by species. Error bars represent SE for a given species; for species with no error bars, only one individual was collected for that species.

bioaccumulation is the unexpected result observed for halibut in the western Aleutian region in Bentzen et al. (2016); this region had the lowest $\delta^{15}N$ while having the highest Hg levels in the study. It is possible that a methylation hotspot could have been detected in this region if they would have also measured $\delta^{34}S$ for the same individuals (see footnote 2 in Table 2). Finally, in our own study, daubed shanny had higher Hg levels than banded gunnel (as correctly predicted by $\delta^{34}S$) despite being 1.5‰ lower in $\delta^{15}N$.

The negative correlation between $\delta^{34}S$ and Hg observed in our study is consistent with the sulfide abundance hypothesis (high sulfide abundance evidencing high methylation and high Hg accumulation). Where bacteria are limited by sulfate availability, one might expect a positive relationship between bacteria and sulfate as areas with high sulfate abundance can sustain high bacteria populations (Fig. 1). However, where bacteria are limited by other factors (such as oxygen concentrations, competition with other biota, sulfate recycling, temperature, presence of compounds complexing with Hg, or MeHg photodegradation; Marschall et al., 1993, Muyzer and Stams, 2008, Point et al., 2011, Schaefer et al., 2011, O'Driscoll et al., 2011, Graham et al., 2012, Antler et al., 2013, St. Pierre et al., 2014, Schartup et al., 2015),

one could expect that the abundance of sulfide might be an index of sulfate-reducing bacteria (more bacteria, more sulfate reduction, more sulfide), so that there is a negative relationship between bacteria, mercury, and $\delta^{34}S$. Both possibilities could explain the results presented in Table 2. As future studies continue to use $\delta^{34}S$, along with other commonly used stable isotope ratios, such as $\delta^{15}N$, recurring patterns will permit better understanding of $\delta^{34}S$ and its association with Hg. Increasing sampling efforts in the Arctic will help us to explain the role sulfur stable isotopes play in Hg accumulation in the Arctic marine ecosystems.

Previous studies used $\delta^{34}S$ primarily as a spatial indicator of the origin of food sources in parallel to how $\delta^{13}C$ is used and, thus, those studies used $\delta^{34}S$ to demonstrate differential Hg levels for samples originating from different habitats (i.e. lake vs. river or marine vs. freshwater vs. estuarine; Hobson, 1999, Fry and Chumchal, 2012, Lavoie et al., 2015, Carr et al., 2017). Indeed, the negative correlation could also be caused by spatial differences in the animals' feeding strategies (Fig. 1: 'habitat variation hypothesis'). Fry and Chumchal (2012) suggest the presence of Hg hotspots that may arise due to the interaction of sulfate-reducing bacteria with aquatic macrophytes and epiphytes. As most of the fish and invertebrates in our study are carnivorous, such an interaction was not considered. However, zooplankton feeding on particulate organic material and algae can play a central role in the introduction of MeHg into the food web (Chételat et al., 2014; Pućko et al., 2014), and similar processes may be involved at our study site. Interactions at different levels of the ecosystem could cause the observed trends in our study. Foster et al. (2012) suggest that studying animals in low trophic position could provide information on what is occurring at higher trophic positions in terms of Hg trends. Foster et al. (2012) also emphasized that using $\delta^{15}N$ as a predictor of trophic level and trophic magnification factors is only meaningful if the specific interactions within a given food web can be identified. This proposal comes in opposition to using an entire data set without clearly identifying the species included and whether they co-occurred across food webs. Given that our sampling strategy depended on what the thick-billed murres brought back to their nests, it was not possible to characterize the entire food web in such detail but future studies should consider having greater sampling efforts that would allow for better descriptions of entire Arctic food webs. Some of the trends reported by Foster et al. (2012) might be clarified if $\delta^{34}S$ were also measured and, indeed, the lack of a correlation with $\delta^{15}N$ may be due to variation in sulfate-reducing bacteria at the base of their food web rather than variation in food web interactions.

Other possible causes for the inverse correlation between Hg and

Table 2
Summary of published studies that use sulfur and nitrogen isotopes as explanatory variables for Hg concentrations.

Studied organisms	Location	Marine/Freshwater	Hg - $\delta^{15}N$ correlation	Hg - $\delta^{34}S$ correlation	Reference
Fish and invertebrates	Nunavut, Canada	Marine	Positive	Negative	This study
Seabirds	British Columbia, Canada	Marine	N/S	Positive	Elliott and Elliott (2016)
Fish	Northwest Territories, Canada	Freshwater	Positive	Negative	Carr et al. (2017)
Halibut	Alaska, USA	Marine	Negative[1]	Not studied[2]	Bentzen et al. (2016)
Cormorants and terns	Ontario, Canada	Both	Negative to N/S[3]	Negative[3]	Lavoie et al. (2015)
Yellow-legged gull	Eastern Iberian coast, Spain	Marine	N/S	Positive	Ramos et al. (2013)
Fish	Ontario, Canada	Freshwater	N/S	Positive	Ethier et al. (2008)
Fish and invertebrates	Louisiana, USA	Estuary	Positive	Negative	Fry and Chumchal (2012)
Fish and invertebrates	Missouri, USA	Freshwater	Negative to Positive	Negative to Positive	Schmitt et al. (2011)
Fish and invertebrates	Nova Scotia, Canada	Freshwater	Positive	Negative to Positive[4]	Clayden et al. (2013, 2017)

[1] $\delta^{15}N$ for the western Aleutian region was the lowest from the study while having the highest total Hg concentrations; for the rest of the sites the correlation was positive.

[2] Although this study doesn't analyze the $\delta^{34}S$ ratio, it does consider that a possible source of high Hg levels is marine Hg methylation, which could be evidenced if the $\delta^{34}S$ signature for the studied sites was measured.

[3] At the ^{13}C-rich site, low $\delta^{15}N$ was associated with high total Hg values for cormorants while low $\delta^{34}S$ was associated with high total Hg for both cormorants and terns.

[4] The relationship between $\delta^{34}S$ and Hg (total Hg and MeHg) was negative when data for all fish and invertebrates within a given lake were combined. Relationships for species considered individually were not significant, except for yellow perch, which was negative among all but one of the studied lakes.

δ^{34}S are high inputs of terrestrial carbon and organic matter originating from rivers and thaw ponds flowing into the Canadian Arctic Ocean, especially Hudson Bay, and the consequent external sources of Hg entering the food web (Braune et al., 2005, 2015; Foster et al., 2012; MacMillan et al., 2015). Dissolved organic matter (DOM) can increase Hg bioavailability by forming Hg nanoparticles that are taken up more easily by sulfate-reducing bacteria (Graham et al., 2012). Additionally, Hg methylation has been detected in methanogenic archaea that were isolated from northern peat lands, which suggests that MeHg could be imported from terrestrial environments through runoffs during thawing events in the summer months (Gilmour et al., 2013). Although δ^{13}C, widely-used to distinguish between terrestrial and marine carbon sources, was not a significant predictor of Hg concentration patterns in our dataset, sulfur isotopes may be used to distinguish Hg sources (Fig. 1: 'habitat variation hypothesis'). The negative association between δ^{34}S and Hg could have been caused by species with low δ^{34}S (more terrestrial input; Lott et al., 2003) feeding on prey more closely associated with high-Hg terrestrial runoffs. A marine-terrestrial cut-off, initially used to distinguish between high δ^{34}S marine raptors from other coastal birds with lower δ^{34}S, was established at a value of 10‰ (Lott et al., 2003). Despite the large extent of the Hudson Bay watershed, we argue that terrestrial input was likely low in our system, given that our lowest δ^{34}S value was 13.7‰. Regardless, Hg bioaccumulation in freshwater and saltwater Arctic ecosystems, and the interaction between the two, is complex and warrants further study (Chételat et al., 2015; Chételat et al., 2008).

Even though thick-billed murres are a highly-studied species in the Arctic, not much is known about the ecology of the prey they consume. Our stable isotope data can help elucidate some of the characteristics of these animals. As expected, zooplankton species such as gammarids and sea butterflies occupy a low trophic position. However, euphausiids (though we only studied one species) had a higher δ^{15}N than the rest of the studied zooplankton, even though euphausiids mostly feed on phytoplankton. Unlike fish, euphausiids have a relatively low MeHg:Hg ratio, and it is possible that the high level of Hg in euphausiids is not indicative of a high level of methylmercury. Fish occupied higher trophic positions, suggesting they feed on invertebrates (intermediate δ^{15}N values; i.e. capelin) or on other fish (high δ^{15}N values; i.e. sculpin). Nonetheless, if species foraged in different food webs, with different baselines or discrimination factors, then δ^{15}N may be an imperfect measure of trophic position (Foster et al., 2012). In general, δ^{13}C and δ^{34}S allowed us to describe the feeding habits of various fish and invertebrates. Capelin, sea butterflies, and jellyfish are associated with pelagic feeding, while daubed shannies and fourline snake blennies are benthic feeders. However, for some other species, the resolution of the isotope ratios included in our study was not sufficient to clearly determine a feeding habit (i.e. sculpin, a benthic species, has intermediate values suggesting it is a benthopelagic feeder) or the isotopes evidenced different results, as occurred for sand lance. Whereas the δ^{13}C signal implied that sand lance fed primarily in pelagic waters, as they come to the surface at night to feed; δ^{34}S, in contrast, implied that sand lance were benthic feeders.

5. Conclusions

Stable isotopes are a valuable tool for understanding multiple aspects of the ecology and toxicology of marine fauna which, otherwise, could not be studied. Using isotope ratios other than the more traditional δ^{15}N and δ^{13}C can expand our knowledge of multiple biological systems that cannot be completely explained. Given the ubiquity of Hg in polar ecosystems, we encourage the incorporation of δ^{34}S into Hg monitoring plans in the Arctic, along with more detailed studies of the links between δ^{34}S, Hg, MeHg, sulfate, and sulfate-reducing bacteria.

Conflicts of interest

None.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2018.07.075.

References

Antler, G., Turchyn, A.V., Rennie, V., Herut, B., Sivan, O., 2013. Coupled sulfur and oxygen isotope insight into bacterial sulfate reduction in the natural environment. Geochim. Cosmochim. Acta 118, 98–117. https://doi.org/10.1016/j.gca.2013.05.005.

Ariya, P.A., Dastoor, A.P., Amyot, M., Schroeder, W.H., Barrie, L., Anlauf, K., Raofie, F., Ryzhkov, A., Davignon, D., Lalonde, J., Steffen, A., 2004. The Arctic: a sink for mercury. Tellus Ser. B Chem. Phys. Meteorol. 56, 397–403. https://doi.org/10.1111/j.1600-0889.2004.00118.x.

Atwell, L., Hobson, K. a, Welch, H.E., 1998. Biomagnification and bioaccumulation of mercury in an arctic marine food web: insights from stable nitrogen isotope analysis. Can. J. Fish. Aquat. Sci. 55, 1114–1121. https://doi.org/10.1139/cjfas-55-5-1114.

Bentzen, R., Castellini, J.M., Gerlach, R., Dykstra, C., O'Hara, T., 2016. Mercury concentrations in Alaska Pacific halibut muscle relative to stable isotopes of C and N and other biological variables. Mar. Pollut. Bull. 113, 110–116. https://doi.org/10.1016/j.marpolbul.2016.08.068.

Braune, B.M., 2007. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975-2003. Environ. Pollut. 148, 599–613. https://doi.org/10.1016/j.envpol.2006.11.024.

Braune, B.M., Outridge, P.M., Fisk, A.T., Muir, D.C.G., Helm, P.A., Hobbs, K., Hoekstra, P.F., Kuzyk, Z.A., Kwan, M., Letcher, R.J., Lockhart, W.L., Norstrom, R.J., Stern, G.A., Stirling, I., 2005. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: an overview of spatial and temporal trends. Sci. Total Environ. 351–352, 4–56. https://doi.org/10.1016/j.scitotenv.2004.10.034.

Braune, B.M., Gaston, A.J., Elliott, K.H., Provencher, J.F., Woo, K.J., Chambellant, M., Ferguson, S.H., Letcher, R.J., 2014a. Organohalogen contaminants and total mercury in forage fish preyed upon by thick-billed murres in northern Hudson Bay. Mar. Pollut. Bull. 78, 258–266. https://doi.org/10.1016/j.marpolbul.2013.11.003.

Braune, B.M., Gaston, A.J., Hobson, K.A., Gilchrist, H.G., Mallory, M.L., 2014b. Changes in food web structure alter trends of mercury uptake at two seabird colonies in the Canadian Arctic. Environ. Sci. Technol. 48, 13246–13252. https://doi.org/10.1021/es5036249.

Braune, B., Chételat, J., Amyot, M., Brown, T., Clayden, M., Evans, M., Fisk, A., Gaden, A., Girard, C., Hare, A., Kirk, J., Lehnherr, I., Letcher, R., Loseto, L., Macdonald, R., Mann, E., McMeans, B., Muir, D., O'Driscoll, N., Poulain, A., Reimer, K., Stern, G., 2015. Mercury in the marine environment of the Canadian Arctic: review of recent findings. Sci. Total Environ. 509–510, 67–90. https://doi.org/10.1016/j.scitotenv.2014.05.133.

Braune, B.M., Gaston, A.J., Mallory, M.L., 2016. Temporal trends of mercury in eggs of five sympatrically breeding seabird species in the Canadian Arctic. Environ. Pollut. 214, 124–131. https://doi.org/10.1016/j.envpol.2016.04.006.

Burnham, K.P., Anderson, D.R., 2002. Model Selection and Multimodel Inference: A Practical Information-Theoretic Approach, Second edition. Springer-Verlag, New York, NY. https://doi.org/10.1007/b97636.

Carr, M.K., Jardine, T.D., Doig, L.E., Jones, P.D., Bharadwaj, L., Tendler, B., Chételat, J., Cott, P., Lindenschmidt, K.-E., 2017. Stable sulfur isotopes identify habitat-specific foraging and mercury exposure in a highly mobile fish community. Sci. Total Environ. 586, 338–346. https://doi.org/10.1016/j.scitotenv.2017.02.013.

Chételat, J., Amyot, M., Cloutier, L., Poulain, A., 2008. Metamorphosis in chironomids,

more than mercury supply, controls methylmercury transfer to fish in high Arctic lakes. Environ. Sci. Technol. 42, 9110–9115. https://doi.org/10.1021/es801619h.

Chételat, J., Poulain, A.J., Amyot, M., Cloutier, L., Hintelmann, H., 2014. Ecological determinants of methylmercury bioaccumulation in benthic invertebrates of polar desert lakes. Polar Biol. 37, 1785–1796. https://doi.org/10.1007/s00300-014-1561-3.

Chételat, J., Amyot, M., Arp, P., Blais, J.M., Depew, D., Emmerton, C.A., Evans, M., Gamberg, M., Gantner, N., Girard, C., Graydon, J., Kirk, J., Lean, D., Lehnherr, I., Muir, D., Nasr, M., Poulain, A., J., Power, M., Roach, P., Stern, G., Swanson, H., van der Velden, S., 2015. Mercury in freshwater ecosystems of the Canadian Arctic: recent advances on its cycling and fate. Sci. Total Environ. 509–510, 41–66. https://doi.org/10.1016/j.scitotenv.2014.05.151.

Clayden, M.G., Kidd, K.A., Wyn, B., Kirk, J.L., Muir, D.C.G., O'Driscoll, N.J., 2013. Mercury biomagnification through food webs is affected by physical and chemical characteristics of lakes. Environ. Sci. Technol. 47, 12047–12053. https://doi.org/10.1021/es4022975.

Clayden, M.G., Lescord, G.L., Kidd, K.A., Wang, X., Muir, D.C.G., O'Driscoll, N.J., 2017. Using sulfur stable isotopes to assess mercury bioaccumulation and biomagnification in temperate lake food webs. Environ. Toxicol. Chem. 36, 661–670. https://doi.org/10.1002/etc.3615.

Compeau, G.C., Bartha, R., 1985. Sulfate-reducing bacteria: principal methylators of mercury in anoxic estuarine sediment. Appl. Environ. Microbiol. 50, 498–502.

Dietz, R., Sonne, C., Basu, N., Braune, B., O'Hara, T., Letcher, R.J., Scheuhammer, T., Andersen, M., Andreasen, C., Andriashek, D., Asmund, G., Aubail, A., Baagøe, H., Born, E.W., Chan, H.M., Derocher, A.E., Grandjean, P., Knott, K., Kirkegaard, M., Krey, A., Lunn, N., Messier, F., Obbard, M., Olsen, M.T., Ostertag, S., Peacock, E., Renzoni, A., Rigét, F.F., Skaare, J.U., Stern, G., Stirling, I., Taylor, M., Wiig, Ø., Wilson, S., Aars, J., 2013. What are the toxicological effects of mercury in Arctic biota? Sci. Total Environ. 443, 775–790. https://doi.org/10.1016/j.scitotenv.2012.11.046.

Driscoll, C.T., Mason, R.P., Chan, H.M., Jacob, D.J., Pirrone, N., 2013. Mercury as a global pollutant: sources, pathways, and effects. Environ. Sci. Technol. 47, 4967–4983. https://doi.org/10.1021/es305071v.

Elliott, J.E., Elliott, K.H., 2013. Tracking marine pollution. Science (80-.) 340, 556–558. https://doi.org/10.1126/science.1235197.

Elliott, K.H., Elliott, J.E., 2016. Origin of sulfur in diet drives spatial and temporal mercury trends in seabird eggs from Pacific Canada 1968–2015. Environ. Sci. Technol. 50, 13380–13386. https://doi.org/10.1021/acs.est.6b05458.

Elliott, K.H., Gaston, A.J., 2008. Mass-length relationships and energy content of fishes and invertebrates delivered to nestling Thick-billed Murres Uria lomvia in the Canadian Arctic, 1981-2007. Mar. Ornithol. 36, 25–34.

Elliott, K.H., Davis, M., Elliott, J., 2014. Equations for lipid normalization of carbon stable isotope ratios in aquatic bird eggs. PLoS One 9, 33–36. https://doi.org/10.1371/journal.pone.0083597.

Ethier, A.L.M., Scheuhammer, A.M., Bond, D.E., 2008. Correlates of mercury in fish from lakes near Clyde Forks, Ontario, Canada. Environ. Pollut. 154, 89–97. https://doi.org/10.1016/j.envpol.2008.01.009.

Fort, J., Robertson, G.J., Grémillet, D., Traisnel, G., Bustamante, P., 2014. Spatial ecotoxicology: migratory arctic seabirds are exposed to mercury contamination while overwintering in the Northwest Atlantic. Environ. Sci. Technol. 48, 11560–11567. https://doi.org/10.1021/es504045g.

Foster, K.L., Stern, G.A., Pazerniuk, M.A., Hickie, B., Walkusz, W., Wang, F., Macdonald, R.W., 2012. Mercury biomagnification in marine zooplankton food webs in Hudson Bay. Environ. Sci. Technol. 46, 12952–12959. https://doi.org/10.1021/es303434p.

Fry, B., Chumchal, M.M., 2012. Mercury bioaccumulation in estuarine food webs. Ecol. Appl. 22, 606–623. https://doi.org/10.1890/11-0921.1.

Furness, R.W., Camphuysen, C.J., 1997. Seabirds as monitors of the marine environment. ICES J. Mar. Sci. 54, 726–737. https://doi.org/10.1006/jmsc.1997.0243.

Gilmour, C.C., Henry, E.A., Mitchell, R., 1992. Sulfate stimulation of mercury methylation in freshwater sediments. Environ. Sci. Technol. 26, 2281–2287. https://doi.org/10.1021/es00035a029.

Gilmour, C.C., Podar, M., Bullock, A.L., Graham, A.M., Brown, S.D., Somenahally, A.C., Johs, A., Hurt, R.A., Bailey, K.L., Elias, D.A., 2013. Mercury methylation by novel microorganisms from new environments. Environ. Sci. Technol. 47, 11810–11820. https://doi.org/10.1021/es403075t.

Góngora, E., Braune, B., Elliott, K., 2018. Dataset from Nitrogen and sulfur isotopes predict variation in mercury levels in Arctic seabird prey. In: Mendeley Data v3, https://doi.org/10.17632/4rngbjg4r8.3.

Graham, A.M., Aiken, G.R., Gilmour, C.C., 2012. Dissolved organic matter enhances microbial mercury methylation under sulfidic conditions. Environ. Sci. Technol. 46, 2715–2723. https://doi.org/10.1021/es203658f.

Grueber, C.E., Nakagawa, S., Laws, R.J., Jamieson, I.G., 2011. Multimodel inference in ecology and evolution: challenges and solutions. J. Evol. Biol. 24, 699–711. https://doi.org/10.1111/j.1420-9101.2010.02210.x.

Hobson, K.A., 1999. Tracing origins and migration of wildlife using stable isotopes: a review. Oecologia 120, 314–326. https://doi.org/10.1007/s004420050865.

Hobson, K.A., Piatt, J.F., Pitocchelli, J., 1994. Using stable isotopes to determine seabird trophic relationships. J. Anim. Ecol. 63, 786. https://doi.org/10.2307/5256.

Kidd, K.A., Hesslein, R.H., Fudge, R.J.P., Hallard, K.A., 1995. The influence of trophic level as measured by δ15N on mercury concentrations in freshwater organisms. Water Air Soil Pollut. 80, 1011–1015. https://doi.org/10.1007/BF01189756.

King, J.K., Kostka, J.E., Frischer, M.E., Saunders, F.M., 2000. Sulfate-reducing bacteria methylate mercury at variable rates in pure culture and in marine sediments. Appl. Environ. Microbiol. 66, 2430–2437. https://doi.org/10.1128/AEM.66.6.2430-2437.2000.Updated.

Kirk, J.L., Lehnherr, I., Andersson, M., Braune, B.M., Chan, L., Dastoor, A.P., Durnford, D.,

Gleason, A.L., Loseto, L.L., Steffen, A., St. Louis, V.L., 2012. Mercury in Arctic marine ecosystems: sources, pathways and exposure. Environ. Res. 119, 64–87. https://doi.org/10.1016/j.envres.2012.08.012.

Krouse, H.R., Mayer, B., 2000. Sulphur and oxygen isotopes in sulphate. In: Environmental Tracers in Subsurface Hydrology. Springer US, Boston, MA, pp. 195–231. https://doi.org/10.1007/978-1-4615-4557-6_7.

Lavoie, R.A., Kyser, T.K., Friesen, V.L., Campbell, L.M., 2015. Tracking overwintering areas of fish-eating birds to identify mercury exposure. Environ. Sci. Technol. 49, 863–872. https://doi.org/10.1021/es502813t.

Liu, G., Cai, Y., Philippi, T., Kalla, P., Scheidt, D., Richards, J., Scinto, L., Appleby, C., 2008. Distribution of total and methylmercury in different ecosystem compartments in the Everglades: implications for mercury bioaccumulation. Environ. Pollut. 153, 257–265. https://doi.org/10.1016/j.envpol.2007.08.030.

Lott, C.A., Meehan, T.D., Heath, J.A., 2003. Estimating the latitudinal origins of migratory birds using hydrogen and sulfur stable isotopes in feathers: influence of marine prey base. Oecologia 134, 505–510. https://doi.org/10.1007/s00442-002-1153-8.

MacMillan, G.A., Girard, C., Chételat, J., Laurion, I., Amyot, M., 2015. High methylmercury in arctic and subarctic ponds is related to nutrient levels in the warming Eastern Canadian Arctic. Environ. Sci. Technol. 49, 7743–7753. https://doi.org/10.1021/acs.est.5b00763.

Marschall, C., Frenzel, P., Cypionka, H., 1993. Influence of oxygen on sulfate reduction and growth of sulfate-reducing bacteria. Arch. Microbiol. 159, 168–173. https://doi.org/10.1007/BF00250278.

McKinney, M.A., Stirling, I., Lunn, N.J., Peacock, E., Letcher, R.J., 2010. The role of diet on long-term concentration and pattern trends of brominated and chlorinated contaminants in western Hudson Bay polar bears, 1991–2007. Sci. Total Environ. 408, 6210–6222. https://doi.org/10.1016/j.scitotenv.2010.08.033.

McKinney, M.A., McMeans, B.C., Tomy, G.T., Rosenberg, B., Ferguson, S.H., Morris, A., Muir, D.C.G., Fisk, A.T., 2012. Trophic transfer of contaminants in a changing Arctic marine food web: Cumberland Sound, Nunavut, Canada. Environ. Sci. Technol. 46https://doi.org/10.1021/es302761p. (120907113320002).

McKinney, M.A., Pedro, S., Dietz, R., Sonne, C., Fisk, A.T., Roy, D., Jenssen, B.M., Letcher, R.J., 2015. A review of ecological impacts of global climate change on persistent organic pollutant and mercury pathways and exposures in arctic marine ecosystems. Curr. Zool. 61, 617–628. https://doi.org/10.1093/czoolo/61.4.617.

Morel, F.M.M., Kraepiel, A.M.L., Amyot, M., 1998. The Chemical Cycle and Bioaccumulation of Mercury. Annu. Rev. Ecol. Syst. 29, 543–566. https://doi.org/10.1146/annurev.ecolsys.29.1.543.

Muyzer, G., Stams, A.J.M., 2008. The ecology and biotechnology of sulphate-reducing bacteria. Nat. Rev. Microbiol. 45, 187–192. https://doi.org/10.1038/nrmicro1892.

Nisbet, I., Montoya, J., Burger, J., Hatch, J., 2002. Use of stable isotopes to investigate individual differences in diets and mercury exposures among common terns Sterna hirundo in breeding and wintering grounds. Mar. Ecol. Prog. Ser. 242, 267–274. https://doi.org/10.3354/meps242267.

O'Driscoll, N.J., Rencz, A., Lean, D.R.S., Beaney, P., 2005. The biogeochemistry and fate of mercury in the environment. In: Sigel, A., Sigel, H., Sigel, R.K.O. (Eds.), Metal Ions in Biological Systems. Biogeochemical Cycles of Elements, Metal Ions in Biological Systems Vol. 43. CRC Press, pp. 221–238. https://doi.org/10.1201/9780824751999.ch9.

O'Driscoll, N.J., Canário, J., Crowell, N., Webster, T., 2011. Mercury speciation and distribution in coastal wetlands and tidal mudflats: relationships with sulphur speciation and organic carbon. Water Air Soil Pollut. 220, 313–326. https://doi.org/10.1007/s11270-011-0756-2.

Overman, N.C., Parrish, D.L., 2001. Stable isotope composition of walleye: ^{15}N accumulation with age and area-specific differences in $\delta^{13}C$. Can. J. Fish. Aquat. Sci. 58, 1253–1260. https://doi.org/10.1139/cjfas-58-6-1253.

Parks, J.M., Johs, A., Podar, M., Bridou, R., Hurt Jr., R.A., Smith, S.D., Tomanicek, S.J., Qian, Yun, Brown, S.D., Brandt, C.C., Palumbo, A.V., Smith, J.C., Wall, J.D., Elias, D.A., Liang, L., 2013. The genetic basis for bacterial mercury methylation. Science (80-.) 339, 1332–1335. https://doi.org/10.1126/science.1230667.

Persson, L.M., Breitholtz, M., Cousins, I.T., de Wit, C.A., MacLeod, M., McLachlan, M.S., 2013. Confronting unknown planetary boundary threats from chemical pollution. Environ. Sci. Technol. 47, 12619–12622. https://doi.org/10.1021/es402501c.

Peterson, B.J., Fry, B., 1987. Stable isotopes in ecosystem studies. Annu. Rev. Ecol. Syst. 18, 293–320. https://doi.org/10.1146/annurev.es.18.110187.001453.

Point, D., Sonke, J.E., Day, R.D., Roseneau, D.G., Hobson, K.a., Vander Pol, S.S., Moors, A.J., Pugh, R.S., Donard, O.F.X., Becker, P.R., 2011. Methylmercury photodegradation influenced by sea-ice cover in Arctic marine ecosystems. Nat. Geosci. 4, 188–194. https://doi.org/10.1038/ngeo1049.

Pollman, C.D., Axelrad, D.M., 2014. Mercury bioaccumulation and bioaccumulation factors for everglades mosquitofish as related to sulfate: a re-analysis of Julian II (2013). Bull. Environ. Contam. Toxicol. 93, 509–516. https://doi.org/10.1007/s00128-014-1384-5.

Pućko, M., Burt, A., Walkusz, W., Wang, F., Macdonald, R.W., Rysgaard, S., Barber, D.G., Tremblay, J.-É., Stern, G.A., 2014. Transformation of mercury at the bottom of the Arctic food web: an overlooked puzzle in the mercury exposure narrative. Environ. Sci. Technol. 48, 7280–7288. https://doi.org/10.1021/es404851b.

Ramos, R., Ramírez, F., Jover, L., 2013. Trophodynamics of inorganic pollutants in a wide-range feeder: the relevance of dietary inputs and biomagnification in the Yellow-legged gull (Larus michahellis). Environ. Pollut. 172, 235–242. https://doi.org/10.1016/j.envpol.2012.09.014.

Rigét, F., Braune, B., Bignert, A., Wilson, S., Aars, J., Born, E., Dam, M., Dietz, R., Evans, M., Evans, T., Gamberg, M., Gantner, N., Green, N., Gunnlaugsdóttir, H., Kannan, K., Letcher, R., Muir, D., Roach, P., Sonne, C., Stern, G., Wiig, Ø., 2011. Temporal trends of Hg in Arctic biota, an update. Sci. Total Environ. 409, 3520–3526. https://doi.org/10.1016/j.scitotenv.2011.05.002.

Rockström, J., Steffen, W., Noone, K., Persson, Å., Chapin, F.S., Lambin, E.F., Lenton, T.M., Scheffer, M., Folke, C., Schellnhuber, H.J., Nykvist, B., de Wit, C.A., Hughes, T., van der Leeuw, S., Rodhe, H., Sörlin, S., Snyder, P.K., Costanza, R., Svedin, U., Falkenmark, M., Karlberg, L., Corell, R.W., Fabry, V.J., Hansen, J., Walker, B., Liverman, D., Richardson, K., Crutzen, P., Foley, J.A., 2009. A safe operating space for humanity. Nature 461, 472–475. https://doi.org/10.1038/461472a.

Schaefer, J.K., Rocks, S.S., Zheng, W., Liang, L., Gu, B., Morel, F.M.M., 2011. Active transport, substrate specificity, and methylation of Hg(II) in anaerobic bacteria. Proc. Natl. Acad. Sci. 108, 8714–8719. https://doi.org/10.1073/pnas.1105781108.

Schartup, A.T., Ndu, U., Balcom, P.H., Mason, R.P., Sunderland, E.M., 2015. Contrasting effects of marine and terrestrially derived dissolved organic matter on mercury speciation and bioavailability in seawater. Environ. Sci. Technol. 49, 5965–5972. https://doi.org/10.1021/es506274x.

Schell, D., Barnett, B., Vinette, K., 1998. Carbon and nitrogen isotope ratios in zooplankton of the Bering, Chukchi and Beaufort seas. Mar. Ecol. Prog. Ser. 162, 11–23. https://doi.org/10.3354/meps162011.

Schmitt, C.J., Stricker, C.A., Brumbaugh, W.G., 2011. Mercury bioaccumulation and biomagnification in Ozark stream ecosystems. Ecotoxicol. Environ. Saf. 74, 2215–2224. https://doi.org/10.1016/j.ecoenv.2011.08.008.

Selin, N.E., 2009. Global biogeochemical cycling of mercury: a review. Annu. Rev. Environ. Resour. 34, 43–63. https://doi.org/10.1146/annurev.environ.051308.084314.

Skov, H., Christensen, J.H., Goodsite, M.E., Heidam, N.Z., Jensen, B., Wåhlin, P., Geernaert, G., 2004. Fate of elemental mercury in the arctic during atmospheric mercury depletion episodes and the load of atmospheric mercury to the Arctic. Environ. Sci. Technol. 38, 2373–2382. https://doi.org/10.1021/es030080h.

St. Pierre, K.A., Chételat, J., Yumvihoze, E., Poulain, A.J., 2014. Temperature and the Sulfur Cycle Control Monomethylmercury Cycling in High Arctic Coastal Marine Sediments from Allen Bay, Nunavut, Canada. Environ. Sci. Technol. 48, 2680–2687. https://doi.org/10.1021/es405253g.

Vo, A.-T.E., Bank, M.S., Shine, J.P., Edwards, S.V., 2011. Temporal increase in organic mercury in an endangered pelagic seabird assessed by century-old museum specimens. Proc. Natl. Acad. Sci. U. S. A. 108, 7466–7471. https://doi.org/10.1073/pnas.1013865108.