



# Mercury accumulation, biomagnification, and relationships to $\delta^{13}\text{C}$ , $\delta^{15}\text{N}$ and $\delta^{34}\text{S}$ of fishes and marine mammals in a coastal Arctic marine food web

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## ABSTRACT

Combining mercury and stable isotope data sets of consumers facilitates the quantification of whether contaminant variation in predators is due to diet, habitat use and/or environmental factors. We investigated inter-species variation in total Hg (THg) concentrations, trophic magnification slope between  $\delta^{15}\text{N}$  and THg, and relationships of THg with  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  in 15 fish and four marine mammal species (249 individuals in total) in coastal Arctic waters. Median THg concentration in muscle varied between species ranging from  $0.08 \pm 0.04 \mu\text{g g}^{-1} \text{dw}$  in capelin to  $3.10 \pm 0.80 \mu\text{g g}^{-1} \text{dw}$  in beluga whales. Both  $\delta^{15}\text{N}$  ( $r^2 = 0.26$ ) and  $\delta^{34}\text{S}$  ( $r^2 = 0.19$ ) best explained variation in log-THg across consumers. Higher THg concentrations occurred in higher trophic level species that consumed more pelagic-associated prey than consumers that rely on the benthic microbial-based food web. Our study illustrates the importance of using a multi-isotopic approach that includes  $\delta^{34}\text{S}$  when investigating trophic Hg dynamics in coastal marine systems.

## 1. Introduction

Humans are hyperkeystone species that are fundamentally changing the geological, biological and chemical components of Earth at unprecedented rates leading to a new geological epoch, The Anthropocene (Lewis and Maslin, 2015; Worm and Paine, 2016). Human industrial activity emits a wide array of chemical pollutants, such as toxic trace metals like mercury (Hg), which can pose toxicological consequences to biota across ecosystems and is of health concern to the global human population (Driscoll et al., 2013; Basu et al., 2022). Anthropogenically emitted Hg is transported to the Arctic from more southern latitudes via the atmosphere, rivers, oceans and migratory species (AMAP, 2021). Legacy mercury is also released into the environment via natural processes and as a result of climate change (e.g. increased permafrost thaw; AMAP, 2021). Furthermore, Hg concentrations in Arctic biota have been

increasing over time (Morris et al., 2022) coinciding with rapidly warming temperatures in the Arctic leading to a wide suite of physical (e.g. sea ice declines) and ecological changes (e.g. inter-specific interactions and primary productivity; Kortsch et al., 2015; Yurkowski et al., 2018; Lewis et al., 2020). Of particular concern to animals is methylmercury (MeHg), which is converted from inorganic Hg by microorganisms and bioaccumulates then biomagnifies with each trophic step up the aquatic food web to longer-lived top predators and be at high risk for adverse effects (Kidd et al., 2012; Dietz et al., 2019; Dietz et al., 2022). In muscle of higher trophic level species, such as predatory fish and marine mammals, total mercury (THg) concentrations are considered equivalent to MeHg concentrations as only MeHg biomagnifies (Campbell et al., 2005; Barst et al., 2022).

Stable isotope ratios are biogeochemical tracers that provide time-integrated information on consumer diet and habitat use (Layman

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et al., 2012). When measured with contaminants, stable isotopes can allow researchers to track chemical pollution through the food web and determine whether contaminant variation in top predators is a result of diet, habitat use, or other environmental factors (Loseto et al., 2008; Clayden et al., 2013). For example, nitrogen stable isotopes ( $\delta^{15}\text{N}$ ) characterize the hierarchical structure of aquatic food webs indicating the consumer trophic position throughout the system (Minagawa and Wada, 1984). Since THg biomagnifies up the food web, relationships between THg and  $\delta^{15}\text{N}$  reveal the extent of Hg biomagnification via an estimate of the trophic magnification slope (TMS) – a measure of the extent to which Hg increases from lower to upper trophic levels in a food web (Lavoie et al., 2013). Other stable isotope ratios, such as  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$ , can be effective predictors of contaminants that are associated with the habitat source of assimilated prey items (e.g., between terrestrial/freshwater and marine resources or benthic and pelagic resources), as both of these stable isotope ratios show little variation between prey and predator compared to endmember sources of both habitat types (McCutchan Jr et al., 2003; Florin et al., 2011).

Sulfates, a form of dissolved sulfur found throughout the water column, are enriched in  $^{34}\text{S}$  compared to sulfides, a form of sulfur that occurs within sediments, as a result of sulfate-reducing bacterial processes (Peterson and Fry, 1987; Connolly et al., 2004). As such, species inhabiting or foraging in benthic habitats, where sulfates are converted to sulfides, have a lower  $\delta^{34}\text{S}$  than their more pelagic-associated counterparts, who have a higher contribution of sulfates in their diet (Brunner and Bernasconi, 2005; Szpak and Buckley, 2020). Concomitantly, sulfate-reducing bacteria also produce MeHg from inorganic Hg but this production can potentially be mediated by the availability of sulfates throughout the water column leading to variation in Hg levels in biota (Jeremiason et al., 2006; Driscoll et al., 2013). This can result in a positive relationship between  $\delta^{34}\text{S}$  and THg, as shown for several seabird species and their prey (Elliott and Elliott, 2016; Górgora et al., 2018), but this relationship has not been investigated across the numerous fish and marine mammal species within Arctic food webs.

The marine waters around Southampton Island, Nunavut, Canada (Fig. 1) are considered a biological hotspot of productivity and

abundance of top predators (Yurkowski et al., 2019), including a core area of Paleo-Inuit occupation for over 2500 years, suggesting this area has continued to have enhanced and reliable productivity over millennia (Clark, 1980). This area provides multiple ecosystem services by supporting Inuit subsistence harvesting (e.g. Atlantic walrus, beluga, narwhal, ringed seals) and local fisheries (i.e. Arctic char), therefore this region has recently been identified by Fisheries and Oceans Canada as an Area of Interest for marine conservation (Loewen et al., 2020). The presence of Hg across the Arctic may diminish the value of ecosystem services thus necessitating a study to increase knowledge of Hg levels in the marine waters of Southampton Island. In this study, we investigated the THg concentrations, biomagnification, and relationships to  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  for 15 fish and four marine mammal species in coastal waters around Southampton Island. Specifically, our objectives were to: 1) quantify differences in the THg concentrations between sampled fish and marine mammal species in the Southampton Island food web, 2) determine the THg trophic magnification slope and intercept across lower and upper trophic level fish and marine mammals in the Southampton Island food web, and 3) quantify relationships between  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  values and THg concentrations across all sampled species in the context of variability in habitat type.

## 2. Materials and methods

### 2.1. Sample collection

Fish samples were collected in August and September 2016, 2018 and 2019 on research expedition cruises in the marine waters around Southampton Island, Nunavut, Canada aboard the RV William Kennedy and the MV Nulijuk, as well as from opportunistic collections from the breeding ledges of thick-billed murres (*Uria lomvia*) at Coats Island (Fig. 1). Ships towed a 3-m pelagic trawl (0.5-cm cod-end mesh) at 2–3 knots for 15 min targeting the subsurface chlorophyll maximum depth, and a 3-m benthic trawl (0.5-cm cod-end mesh) at 2–3 knots for 15 min on the bottom following approved research licenses and animal care protocols (see Acknowledgements for specific details). Ninety muscle

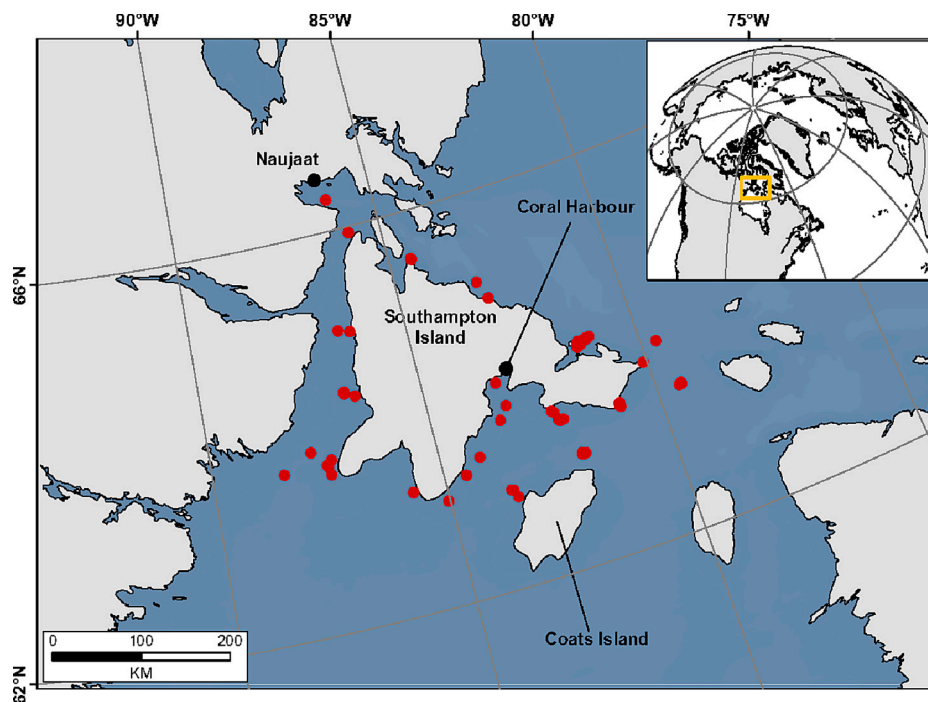


Fig. 1. Study area around Southampton Island highlighting where fish were collected from scientific trawls (red circles) and the Inuit communities of Naujaat and Coral Harbour where marine mammals were collected as part of community-based monitoring programs (black circles). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

samples were collected from 14 fish species, including Arctic cod (*Boreogadus saida*), polar cod (*Arctogadus glacialis*), fourline snakeblenny (*Eumesogrammus praecisus*), Arctic staghorn sculpin (*Gymnocanthus triscuspis*), daubed shanny (*Leptoclinus maculatus*), slender eelblenny (*Lumpenus fabricii*), capelin (*Mallotus villosus*), Arctic sculpin (*Myoxocephalus scorpioides*), shorthorn sculpin (*Myoxocephalus scorpius*), Greenland cod (*Gadus ogac*), banded gunnel (*Pholis fasciata*), Arctic shanny (*Stichaeus punctatus*), moustache sculpin (*Trigllops murrayi*), and ribbed sculpin (*Trigllops pingelii*). Arctic char (*Salvelinus alpinus*) muscle samples were collected opportunistically in August 2019 as part of Inuit subsistence harvests in association with an ongoing community-based monitoring programs with Fisheries and Oceans Canada based in Nauyasat, Nunavut (Fig. 1). Atlantic walrus (*Odobenus rosmarus rosmarus*), ringed seal (*Pusa hispida*), narwhal (*Monodon monoceros*) and beluga (*Delphinapterus leucas*) muscle and liver samples were collected from June to September in 2016, 2018 and 2019 by Inuit hunters from Nauyasat and Coral Harbour, Nunavut as part of their subsistence harvests and ongoing community-based monitoring programs in collaboration with Fisheries and Oceans Canada. Tissue samples were frozen at  $-20\text{ }^{\circ}\text{C}$  and shipped to the Freshwater Institute in Winnipeg, Manitoba before processing.

## 2.2. THg analysis

Prior to THg analysis, fish and marine mammal samples were freeze-dried for 48 h to remove moisture and all THg data is presented in  $\mu\text{g g}^{-1}$  dry weight. Freeze-dried sample were weighed into a tared vessel, thermally decomposed and released THg was quantified THg using a direct Hg analyzer in accordance with USEPA7473 (NIC MA-3000, Nippon Instruments, North America) at McGill University as described in Golzadeh et al. (2020). Quality assurance steps included the analysis of DOLT-5 (dogfish liver) and S3 (Ontario chinook salmon) certified reference materials with each batch of 15 samples. Precision was indicated by relative standard deviation of sets of triplicate samples ( $n = 11$ ) and ranging from 0.96 % to 10.4 % with a mean across all sets of  $<5\%$ . Accuracy was measured by THg concentrations of DOLT-5 ( $0.33 \pm 0.077\ \mu\text{g g}^{-1}$  dw; certified THg value is  $0.44 \pm 0.18\ \mu\text{g g}^{-1}$ ) and S3 ( $0.91 \pm 0.02\ \mu\text{g g}^{-1}$ ; certified THg value is  $0.99 \pm 0.06\ \mu\text{g g}^{-1}$ ) where the mean accuracies were  $74.2 \pm 3.3\%$  and  $92.2 \pm 4.7\%$ , respectively. THg was not detected in the blanks and all samples were above the mean detection limit of  $0.00024\ \mu\text{g}$ .

## 2.3. Stable isotope analysis

Frozen fish muscle and marine mammal muscle and liver samples were freeze-dried for 48 h and then crushed into a fine powder using a mortar and pestle. Due to the presence of lipids in Arctic marine mammal liver and muscle (Yurkowski et al., 2015), and fish muscle (Post et al., 2007), lipids were extracted with 2:1 chloroform:methanol solvent using a modified version of the Bligh and Dyer (1959) method. Stable isotope analysis was performed at the Chemical Tracers Laboratory, Great Lakes Institute for Environmental Research, at the University of Windsor using a Delta V Advantage Mass spectrometer (Thermo Finnigan, San Jose, CA, USA) coupled to a Costech 4010 Elemental Combustion system (Costech, Valencia, CA, USA) and a ConFlo IV gas interface. For  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  analysis, subsamples of 400–600  $\mu\text{g}$  were weighed into tin capsules. For  $\delta^{34}\text{S}$ , 3000–6000  $\mu\text{g}$  of the sample plus 300–500  $\mu\text{g}$  of vanadium pentoxide was encapsulated. All stable isotope ratios are expressed in per mil (‰) in standard delta ( $\delta$ ) notation relative to the international standards Pee Dee Belemnite for carbon, atmospheric  $\text{N}_2$  for nitrogen, and Vienna Cañon Diablo Triolite for sulfur using the following equation:  $\delta X = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^{-3}$ , where X is  $^{13}\text{C}$ ,  $^{15}\text{N}$  or  $^{34}\text{S}$  and R equals  $^{13}\text{C}/^{12}\text{C}$ ,  $^{15}\text{N}/^{14}\text{N}$  and  $^{34}\text{S}/^{32}\text{S}$ . Instrumental accuracy checked throughout the period of time that these samples were analyzed was based on U.S. National Institute of Standards and Technology (NIST) standards 8573, 8547 and 8574 for  $\delta^{15}\text{N}$ , 8542,

8573 and 8574 for  $\delta^{13}\text{C}$  ( $n = 50$  for all), and 8555 and 8529 for  $\delta^{34}\text{S}$  ( $n = 30$  for all). The mean difference from the certified values were  $-0.09$ ,  $0.14$ ,  $-0.06\%$  for  $\delta^{15}\text{N}$ ,  $0.09$ ,  $0.01$  and  $-0.08\%$  for  $\delta^{13}\text{C}$ , and  $0.25$  and  $0.30\%$  for  $\delta^{34}\text{S}$ , respectively. Precision, assessed by the standard deviation of replicate analyses of three standards (NIST1577c, internal lab standard, tilapia muscle), USGS 40 and Urea ( $n = 22$  for all), measured  $\leq 0.25\%$  for  $\delta^{15}\text{N}$  and  $\leq 0.13\%$  for  $\delta^{13}\text{C}$  for all the standards. For  $\delta^{34}\text{S}$ , the precision from standards USGS 42, NIST 8555 and NIST 8529 ( $n = 39$  for all), measured  $\leq 0.43\%$ .

## 2.4. Statistical analysis

All statistical analyses were performed in R v.4.1.2 (R Core Team, 2021) using packages “ggplot2” (Wickham, 2016) and “AICcmodavg” (Mazerolle and Mazerolle, 2017) where  $\alpha$  was set to 0.05. Only muscle samples of fishes and marine mammals were used for analysis due to sample size. Prior to analysis, THg concentrations were log transformed to achieve normality. THg concentrations across fish and marine mammal species were compared using one-way analysis of variance (ANOVA), followed by a Tukey’s post hoc test. Pearson’s correlation was used to evaluate the correlation between log transformed THg concentration and  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  of muscle across species altogether. General linear models were used to investigate relationships between THg concentration (dry weight and log-transformed) as the dependent variable and  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  as the independent variables across all species. Due to differences in sample size among species, we averaged log-THg concentration,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  values for all species except daubed shanny ( $n = 1$ ) prior to correlation, partial correlation and linear regression analyses. Models were evaluated and ranked using Akaike’s Information Criterion adjusted for small sample sizes where the most parsimonious model is one with the lowest  $\Delta\text{AIC}_c$  (Burnham and Anderson, 2004). If models were within two  $\Delta\text{AIC}_c$  of the lowest  $\text{AIC}_c$  model, we considered the model with the fewest number of estimated parameters and with the lowest standard error in the parameter estimate to be the best model following Burnham and Anderson (2004).

Trophic magnification slope was determined as the slope value between mean log THg concentration of muscle and mean  $\delta^{15}\text{N}$  of muscle across all species. Relationships between mean log THg concentration of muscle and mean  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  of muscle across all species were considered to indicate associations with habitat type (i.e., pelagic or benthic). The effect of  $\delta^{15}\text{N}$  on log THg while controlling for habitat type (i.e.,  $\delta^{34}\text{S}$ ) was determined using partial correlation analysis which estimates the unique contribution of an independent variable, in this case  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$ , to the  $r^2$  of the model. We also used residual plots to test linearity in the relationships between log THg,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  (Zuur et al., 2010). To test for differences in the slope and intercept between log THg and  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$ , we used linear analysis of covariance.

## 3. Results

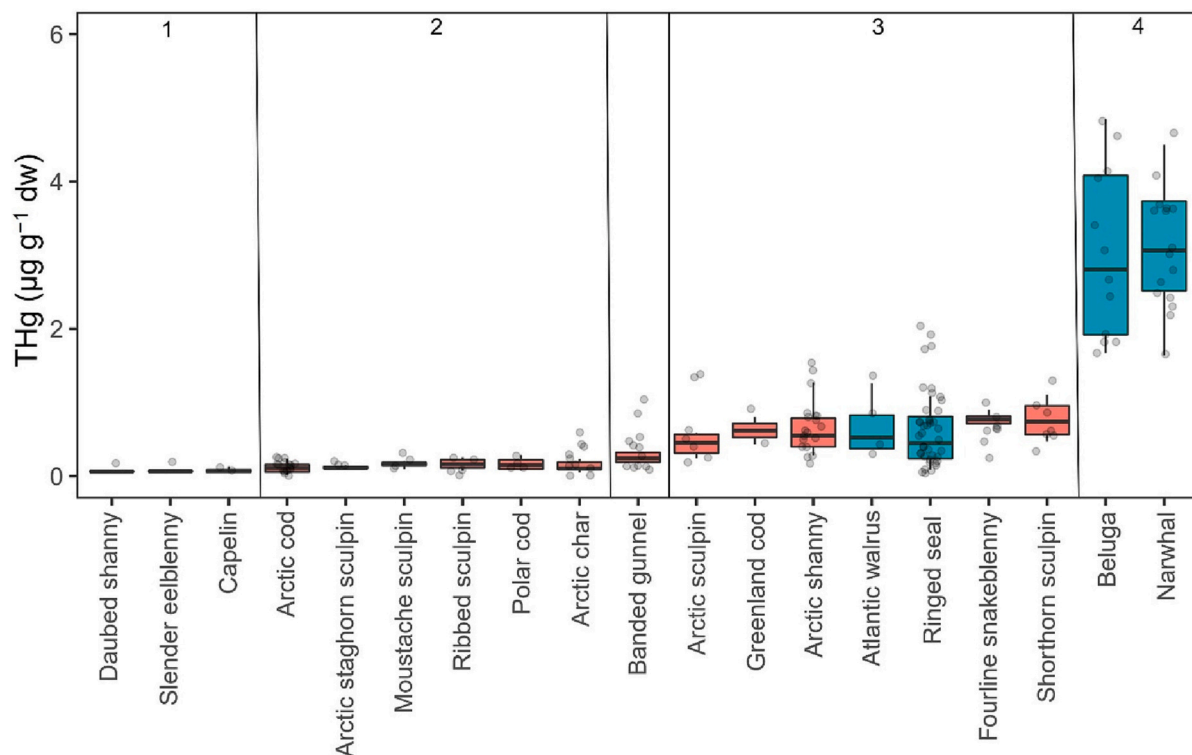
### 3.1. Inter-specific variation in THg concentrations of muscle

Muscle THg concentrations varied widely among fish and marine mammal species, with median concentrations ranging from  $0.08 \pm 0.04$  (median  $\pm$  SD)  $\mu\text{g g}^{-1}$  dw in slender eelblenny and capelin to  $3.10 \pm 0.80\ \mu\text{g g}^{-1}$  dw in beluga (Table 1). One-way analysis of variance (ANOVA) revealed significant differences in muscle THg among species ( $F_{18,162} = 29.95$ ,  $p < 0.001$ ). Tukey’s post-hoc tests revealed four species groups (Fig. 2). The first species group consisted of capelin, slender eelblenny and daubed shanny, for which the THg concentrations were significantly lower than all other species. The second species group, composed of Arctic char, polar cod, ribbed sculpin, moustache sculpin, Arctic staghorn sculpin and Arctic cod, was significantly lower than species in the third species group (i.e., shorthorn sculpin, fourline snakeblenny, ringed seal, Atlantic walrus, Arctic shanny, Greenland cod and Arctic sculpin;  $p < 0.05$ ). The THg concentrations of banded gunnel

**Table 1**

Species, sample size and mean  $\pm$  SD of standard length (cm),  $\delta^{13}\text{C}$  (‰),  $\delta^{15}\text{N}$  (‰), C:N,  $\delta^{34}\text{S}$  (‰) and total mercury (THg;  $\mu\text{g g}^{-1}$  dw) of fishes and marine mammals collected around Southampton Island, Nunavut from 2016 to 2019. Note that all samples were lipid extracted before stable isotope analysis.

Common name	Species name	n	Standard length	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	C:N	$\delta^{34}\text{S}$	THg
<b>Muscle</b>								
Polar cod	<i>Arctogadus glacialis</i>	5	18 $\pm$ 3	-19.5 $\pm$ 0.3	15.2 $\pm$ 0.9	3.1 $\pm$ 0.1	18.5 $\pm$ 0.7	0.17 $\pm$ 0.09
Arctic cod	<i>Boreogadus saida</i>	12	14 $\pm$ 4	-19.4 $\pm$ 0.7	15.1 $\pm$ 1.0	3.1 $\pm$ 0.1	17.7 $\pm$ 0.7	0.11 $\pm$ 0.06
Fourline snakeblenny	<i>Eumesogrammus praecisus</i>	7	14 $\pm$ 2	-18.4 $\pm$ 0.5	16.1 $\pm$ 0.5	3.1 $\pm$ 0.1	16.6 $\pm$ 1.2	0.71 $\pm$ 0.20
Arctic staghorn sculpin	<i>Gymnocanthus tricuspis</i>	3	10 $\pm$ 3	-16.2 $\pm$ 0.2	14.4 $\pm$ 0.6	3.3 $\pm$ 0.1	13.6 $\pm$ 0.3	0.12 $\pm$ 0.03
Daubed shanny	<i>Leptoctinus maculatus</i>	1	12	-19.5	13.8	3.1	15.7	0.1
Slender eelblenny	<i>Lumpenus fabricii</i>	2	11 $\pm$ 2	-16.3 $\pm$ 0.2	14.6 $\pm$ 0.2	3.2 $\pm$ 0.1	14.5 $\pm$ 1.3	0.08 $\pm$ 0.01
Capelin	<i>Mallotus villosus</i>	5	11 $\pm$ 1	-19.4 $\pm$ 0.7	13.8 $\pm$ 0.4	3.1 $\pm$ 0.1	17.2 $\pm$ 0.5	0.08 $\pm$ 0.04
Arctic sculpin	<i>Myoxocephalus scorpioides</i>	6	19 $\pm$ 9	-19.5 $\pm$ 0.8	16.1 $\pm$ 0.8	3.1 $\pm$ 0.1	19.1 $\pm$ 0.3	0.56 $\pm$ 0.41
Shorthorn sculpin	<i>Myoxocephalus scorpius</i>	7	21 $\pm$ 10	-19.0 $\pm$ 0.9	16.5 $\pm$ 0.9	3.1 $\pm$ 0.1	18.6 $\pm$ 0.3	0.77 $\pm$ 0.24
Greenland cod	<i>Gadus ogac</i>	2		-20.7 $\pm$ 0.2	15.4 $\pm$ 0.4	3.2 $\pm$ 0.1	18.3 $\pm$ 0.4	0.62 $\pm$ 0.26
Banded gunnel	<i>Pholis fasciata</i>	10	20 $\pm$ 2	-19.6 $\pm$ 0.6	14.7 $\pm$ 1.0	3.1 $\pm$ 0.1	18.7 $\pm$ 0.3	0.32 $\pm$ 0.21
Arctic char	<i>Salvelinus alpinus</i>	10		-19.4 $\pm$ 1.2	13.8 $\pm$ 1.7	3.2 $\pm$ 0.1	15.0 $\pm$ 1.7	0.17 $\pm$ 0.15
Arctic shanny	<i>Stichaeus punctatus</i>	18	13 $\pm$ 1	-19.2 $\pm$ 0.4	15.9 $\pm$ 0.6	3.1 $\pm$ 0.1	17.7 $\pm$ 0.6	0.64 $\pm$ 0.32
Moustache sculpin	<i>Triglops murrayi</i>	6	10 $\pm$ 3	-19.0 $\pm$ 0.9	14.6 $\pm$ 0.5	3.1 $\pm$ 0.1	17.8 $\pm$ 1.4	0.16 $\pm$ 0.04
Ribbed sculpin	<i>Triglops pingelii</i>	6	13 $\pm$ 2	-19.3 $\pm$ 0.8	15.2 $\pm$ 0.2	3.2 $\pm$ 0.1		0.17 $\pm$ 0.07
Atlantic walrus	<i>Odobenus rosmarus rosmarus</i>	4		-18.5 $\pm$ 0.3	12.9 $\pm$ 1.0	3.2 $\pm$ 0.1	18.1 $\pm$ 0.4	0.67 $\pm$ 0.42
Narwhal	<i>Monoceros monodon</i>	15	400 $\pm$ 30	-18.1 $\pm$ 0.3	15.4 $\pm$ 1.2	3.1 $\pm$ 0.1	17.5 $\pm$ 0.8	3.10 $\pm$ 0.80
Beluga	<i>Delphinapterus leucas</i>	20	346 $\pm$ 44	-18.2 $\pm$ 0.3	16.0 $\pm$ 0.5	3.1 $\pm$ 0.1	17.5 $\pm$ 0.6	3.05 $\pm$ 1.14
Ringed Seal	<i>Pusa hispida</i>	41	111 $\pm$ 17	-18.3 $\pm$ 0.4	17.6 $\pm$ 1.3	3.2 $\pm$ 0.1	17.4 $\pm$ 0.5	0.70 $\pm$ 0.99
<b>Liver</b>								
Atlantic walrus	<i>Odobenus rosmarus rosmarus</i>	4		-19.3 $\pm$ 0.2	13.0 $\pm$ 0.8	3.9 $\pm$ 0.4	18.0 $\pm$ 0.4	63.5 $\pm$ 34.2
Narwhal	<i>Monoceros monodon</i>	12	400 $\pm$ 30	-18.4 $\pm$ 0.2	16.4 $\pm$ 0.3	3.3 $\pm$ 0.1	17.9 $\pm$ 0.7	65.1 $\pm$ 50.7
Beluga	<i>Delphinapterus leucas</i>	8	346 $\pm$ 44	-18.4 $\pm$ 0.4	17.0 $\pm$ 0.5	3.4 $\pm$ 0.1	18.2 $\pm$ 0.8	30.9 $\pm$ 54.4
Ringed Seal	<i>Pusa hispida</i>	45	111 $\pm$ 17	-18.2 $\pm$ 0.5	18.4 $\pm$ 1.1	3.4 $\pm$ 0.2	17.7 $\pm$ 0.6	28.7 $\pm$ 64.8



**Fig. 2.** Boxplots of total mercury (THg) in muscle samples from 15 fish species and four marine mammal species collected around Southampton Island, Nunavut, 2016–2019. The box length represents the upper and lower interquartiles, the bars represent the ranges and the bold horizontal lines are the median values. Grey points represent each individual from that sampled species. Vertical lines separate the four species groupings revealed by Tukey's posthoc tests. The THg concentrations of banded gunnel were not significantly different from any species in the second or third clusters ( $p > 0.05$ ).

were not significantly different from any species in the second or third clusters ( $p > 0.05$ ). The fourth species group consisted of narwhal and beluga, which had a significantly higher THg concentrations compared to all other species ( $p < 0.001$ ).

### 3.2. THg relationships with $\delta^{13}\text{C}$ , $\delta^{15}\text{N}$ and $\delta^{34}\text{S}$

There was a positive correlation between muscle  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  ( $t_{137} = 3.52, p < 0.001, r = 0.29$ ) and  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  ( $t_{178} = 4.97, p < 0.001, r$

= 0.35), but there was no correlation between  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  ( $t_{137} = -1.50$ ,  $p = 0.13$ ). All three isotopes were positively correlated with log THg ( $\delta^{13}\text{C}$ :  $t_{178} = 3.77$ ,  $p < 0.001$ ,  $r = 0.27$ ;  $\delta^{15}\text{N}$ :  $t_{178} = 3.33$ ,  $p = 0.001$ ,  $r = 0.24$ ;  $\delta^{34}\text{S}$ :  $t_{137} = 2.48$ ,  $p = 0.01$ ,  $r = 0.21$ ).

There were four models with a  $\Delta\text{AIC}_c < 2.0$  that best explained variation in log THg of muscle across species and mainly included  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$ , suggesting that both variables are important in explaining THg variation in the Southampton Island marine ecosystem (Table 2). The best-supported model only included  $\delta^{15}\text{N}$  which was positively associated with log THg (slope = 0.227,  $t_{17} = 2.427$ ,  $p$ -value  $< 0.03$ ,  $r^2 = 0.26$ ; Tables 2 and 3; Fig. 3), followed by the second-best supported model which included both  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  (Table 2). The remaining models with a  $\Delta\text{AIC}_c < 2.0$  included  $\delta^{34}\text{S}$  which was also positively associated with log THg (Tables 2 and 3; Fig. 3). Partial correlation analysis revealed that  $\delta^{15}\text{N}$  explained 27 % of the variation in log THg while controlling for  $\delta^{34}\text{S}$  (i.e. habitat type), while  $\delta^{34}\text{S}$  explained 7 % of the variation in log THg while controlling for  $\delta^{15}\text{N}$ . There was no difference in the slope between log THg and  $\delta^{15}\text{N}$  when controlling for and not controlling for the influence of  $\delta^{34}\text{S}$  ( $t_{16} = 1.06$ ,  $p = 0.28$ ). There was also no difference in y-intercepts between the best and second-best supported models (y-intercept  $\pm$  standard error;  $\delta^{15}\text{N}$  model =  $-4.0 \pm 1.4$ ;  $\delta^{15}\text{N} + \delta^{34}\text{S} = -5.0 \pm 1.6$ ).

#### 4. Discussion

This study provides the first characterization of mercury concentration, its relationship with  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$ , and the level of biomagnification across numerous fish and marine mammal species representing a portion of an Arctic coastal marine food web. As such, this study can provide a reference point to assess future trends in stable isotope and mercury dynamics of fishes and marine mammals in northern Hudson Bay. Fish species showed wide variation in THg concentrations with the highest occurring in shorthorn sculpin, fourline snakeblenny, Arctic shanny and Arctic sculpin whose THg concentrations (mean of 0.56–0.77  $\mu\text{g g}^{-1}$  dw) were comparable to their conspecifics from other locations in the Canadian Arctic (see Loseto et al., 2008; Braune et al., 2015; McMeans et al., 2015; Pedro et al., 2017; Barst et al., 2022). The mean THg concentrations of Atlantic walrus, ringed seal, beluga and narwhal around Southampton Island (mean of 0.67–3.10  $\mu\text{g g}^{-1}$  dw) were also similar to their conspecifics at other locations across the Canadian Arctic and in west Greenland (Hansen et al., 1990; Atwell et al., 1998; Rig  t et al., 2007; Loseto et al., 2008; Braune et al., 2015; Brown et al., 2016). Moreover, both  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  were positively associated with THg concentration across all sampled fish and marine mammal species in the Southampton Island marine food web, providing some support for the influence of trophic position and habitat type (related to the sulfate availability hypothesis), consistent with results from other studies on seabirds and their prey (Elliott and Elliott, 2016; G  ngora et al., 2018).

##### 4.1. Inter-specific variation in THg

In general, some of the known benthic-associated fish species (i.e.,

**Table 2**

Candidate models of total mercury (THg) variation of muscle across fishes and marine mammals in the Southampton Island food web ranked by  $\text{AIC}_c$ .

Model	k	$\text{AIC}_c$	$\Delta\text{AIC}_c$	$\text{AIC}_c$ weight
$\delta^{15}\text{N}$	3	28.95	0.00	0.30
$\delta^{15}\text{N} + \delta^{34}\text{S}$	4	30.12	1.17	0.17
$\delta^{13}\text{C} + \delta^{34}\text{S}$	4	30.12	1.17	0.17
$\delta^{34}\text{S}$	3	30.60	1.65	0.13
$\delta^{13}\text{C} + \delta^{15}\text{N} + \delta^{34}\text{S}$	5	31.60	2.66	0.08
Null	2	31.75	2.80	0.07
$\delta^{13}\text{C} + \delta^{15}\text{N}$	4	32.20	3.26	0.06
$\delta^{13}\text{C}$	3	34.60	5.65	0.02

**Table 3**

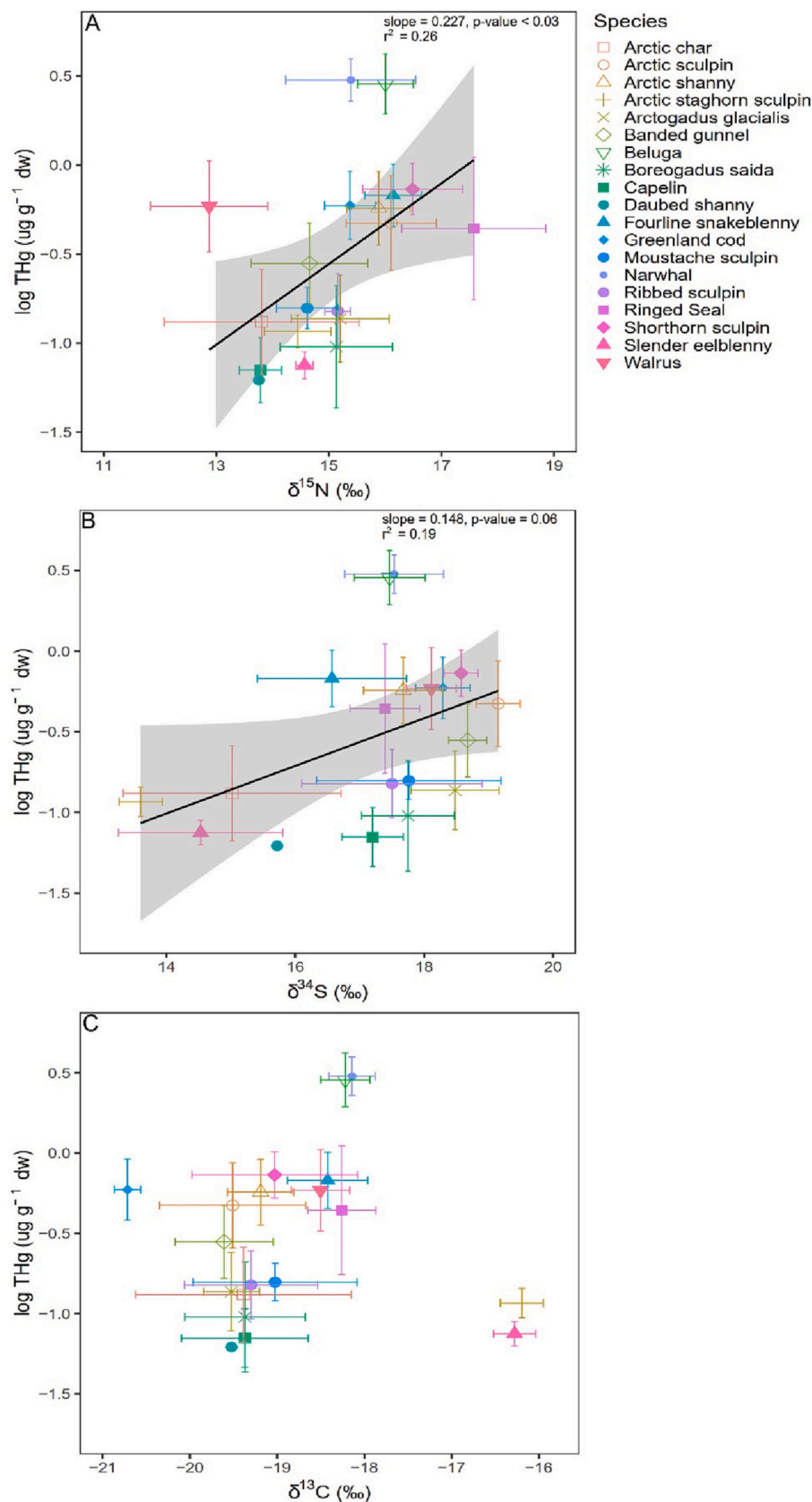
Results from the top four candidate models explaining THg variation of muscle across fish and marine mammal species around Southampton Island. SE: standard error.

Model	Parameter	Estimate	SE	t-value	p-value
$\delta^{15}\text{N}$	Intercept	-3.956	1.414	-2.98	0.01
	$\delta^{15}\text{N}$	0.227	0.093	2.43	<0.03
$\delta^{15}\text{N} + \delta^{34}\text{S}$	Intercept	-4.99	1.576	-3.17	<0.01
	$\delta^{15}\text{N}$	0.181	0.098	1.84	0.08
	$\delta^{34}\text{S}$	0.100	0.074	1.38	0.19
$\delta^{13}\text{C} + \delta^{34}\text{S}$	Intercept	-0.646	1.767	-0.37	0.72
	$\delta^{13}\text{C}$	0.237	0.127	1.86	0.08
	$\delta^{34}\text{S}$	0.265	0.094	2.84	0.01
$\delta^{34}\text{S}$	Intercept	-3.076	1.278	-2.41	<0.03
	$\delta^{34}\text{S}$	0.148	0.074	2.00	0.06

shorthorn sculpin, fourline snakeblenny, Arctic shanny and Arctic staghorn sculpin) showed the highest THg concentrations as well as the highest  $\delta^{15}\text{N}$  values and in turn, trophic position among fish species. These benthic fishes are understudied in the Arctic but have been documented to exhibit a generalist feeding strategy by consuming a wide variety of invertebrate (i.e. copepods, amphipods, decapods and polychaetes) and fish species from both the pelagic and benthic trophic pathways (Keats et al., 1993; Giraldo et al., 2016; Landry et al., 2018). Furthermore, larger-sized benthic fishes, such as shorthorn sculpin, can consume Arctic cod and cannibalize smaller conspecifics due to their relatively larger gape size which likely contributes to higher dietary exposure to Hg (Landry et al., 2018; Hilgendag et al., 2022). Thus, shorthorn sculpin, along with other sculpin species, typically have a relatively higher THg concentration among fishes, which has also been observed in the Beaufort Sea (Loseto et al., 2008), West Greenland (Rig  t et al., 2007), in Frobisher Bay (Hilgendag et al., 2022) and several other areas across the Eastern Canadian Arctic (Pedro et al., 2017, 2019). Despite the relatively higher THg concentrations observed in shorthorn sculpin, fourline snakeblenny, Arctic shanny and Arctic sculpin, these four species are at low risk for sublethal toxic effects of Hg, while all other fish species from this study are considered at no risk for Hg toxicity based on risk categories and thresholds described in Barst et al. (2022).

The mean THg concentrations of key mid-trophic level forage fish species, such as Arctic cod, polar cod and capelin were all  $< 0.2 \mu\text{g g}^{-1}$  dw, which is generally similar to observations from other Arctic systems including Arctic cod near Resolute and Clyde River, Nunavut ( $0.03 \mu\text{g g}^{-1}$  ww; Pedro et al., 2019), Arctic cod in the Beaufort Sea ( $0.16 \mu\text{g g}^{-1}$  dw; Loseto et al., 2008), capelin in Cumberland Sound ( $0.02 \mu\text{g g}^{-1}$  ww; McMeans et al., 2015), western Hudson Bay ( $0.01 \mu\text{g g}^{-1}$  ww; Pedro et al., 2019) and in west Greenland ( $0.07 \mu\text{g g}^{-1}$  dw; Rig  t et al., 2007) further highlighting the ubiquitous trophic role these species play across Arctic food webs. Though many higher trophic level Arctic predators are switching diets and now consuming more capelin, a pelagic-associated species, and less Arctic cod, an ice-associated species, than in previous decades (Gaston et al., 2003; Watt et al., 2016; Yurkowski et al., 2017; McKinney et al., 2022), similarities in THg concentrations between both fish species observed in this study and in Pedro et al. (2017) suggest a limited impact on Hg uptake by Arctic predators. The mean THg concentration of Greenland cod, a species that principally consumes capelin and larger invertebrates, was considerably higher than observed in conspecifics from west Greenland ( $0.03 \mu\text{g g}^{-1}$  ww; Hansson et al., 2020) suggesting relatively higher consumption of fishes than invertebrates in the Southampton Island area. The THg concentration of anadromous Arctic char, which also consume a wide variety of marine forages fishes, marine invertebrates and freshwater insects, was low and indicated no risk to Hg toxicity – a similar result to their conspecifics collected at numerous locations across the Arctic (see Barst et al., 2022).

The mean THg concentrations of narwhal and beluga muscle ( $3.10$  and  $3.05 \mu\text{g g}^{-1}$  dw) were over four times higher compared to Atlantic walrus and ringed seals ( $0.67$  and  $0.70 \mu\text{g g}^{-1}$  dw), where species



**Fig. 3.** Relationships of mean logarithm<sub>10</sub>-transformed total mercury (THg) with (A) mean  $\delta^{15}\text{N}$ , (B) mean  $\delta^{34}\text{S}$ , and (C) mean  $\delta^{13}\text{C}$  of muscle for fish and marine mammal species collected around Southampton Island, Nunavut. Error bars represent standard deviation. Only 1 sample of Daubed shanny was collected. Linear regressions (black lines) with 95 % confidence intervals (grey) were performed on mean values of each species. There was no relationship between mean logarithm<sub>10</sub>-transformed THg with mean  $\delta^{13}\text{C}$ .

differences in maximum life expectancy may be playing a role since THg concentration accumulates with age and because pinnipeds are able to offload Hg through hair growth and moulting (Dietz et al., 2006). Based on previously reported body length – age relationships of narwhal, beluga (Kelley et al., 2015) and ringed seals (Kovacs et al., 2021), most individuals harvested from each species were adults. Narwhals have a maximum life expectancy of approximately 100 years (Garde et al., 2015), while age estimates for belugas have exceeded 70 years (Luque and Ferguson, 2010), which doubles the life expectancy estimated for ringed seals and Atlantic walrus (approximately 40 years of age; Lydersen, 2018; Hammill, 2009).

The  $\delta^{15}\text{N}$  of narwhal and beluga were similar and suggests trophic omnivory by consuming a mix forage fishes (e.g. Arctic cod and capelin) and invertebrates, such as squid (Louis et al., 2021; Watt et al., 2013). Ringed seals had the highest  $\delta^{15}\text{N}$  among species in this study and suggests a high level of piscivory in the Southampton Island area. Though ringed seals had the highest  $\delta^{15}\text{N}$  compared to other species yet a relatively low THg concentration, it is important to note that the ringed seals were sampled near the end of their spring moult fasting period. This can lead to increased  $\delta^{15}\text{N}$  values that reflect catabolism of endogenous protein stores (Young and Ferguson, 2013).

Mean THg concentrations in marine mammal liver were ten to a hundred times higher than in muscle in this study likely due to the liver being an organ that detoxifies methylmercury through demethylation in mammals which results in continual accumulation of inorganic Hg (Braune et al., 2015). Overall, there is no or low risk for adverse health effects or sublethal Hg toxicity in muscle on all four marine mammal species based on risk categories and thresholds described in Dietz et al. (2022).

#### 4.2. Biomagnification of THg

The y-intercepts (baseline THg) and slopes (trophic magnification slope) between the best ( $\delta^{15}\text{N}$ ) and second-best ( $\delta^{15}\text{N} + \delta^{34}\text{S}$ ) supported models were not significantly different suggesting no difference in baseline THg concentrations or trophic magnification slopes between the benthic and pelagic-associated species in this food web. The trophic magnification slope in this study (0.227) was similar to several other marine areas across the Arctic (average slope of 0.21 across eight systems, Lavoie et al., 2013), including the Beaufort Sea and Amundsen Gulf areas (0.232 to 0.255; Loseto et al., 2008), and Cumberland Sound (0.231; McMeans et al., 2015). The slope we report here, however, was higher than that in the marine food web of West Greenland (0.183; Rigét et al., 2007), Lancaster Sound (0.200; Atwell et al., 1998), the North Water Polynya (0.197; Campbell et al., 2005) and a polynya system near Nasaruaalik Island, Nunavut (0.095; Clayden et al., 2015). Although some of these trophic magnification slopes were derived separately by different habitat types (e.g. benthic, pelagic and epibenthic) in those studies, pelagic-benthic habitats are highly interconnected in that benthic-associated species also acquire resources from the pelagic or sympagic carbon pathways in the form of detritus (e.g., filter-feeding clams, Sun et al., 2009; Amiraux et al., 2021). As well, the mean  $\delta^{15}\text{N}$  values of the pelagic-feeding *Calanus hyperboreus* and the benthic grazing sea urchin (*Strongylocentrotus droebachiensis*) were similar (9.5 ‰ versus 8.7 ‰, respectively; Amiraux et al., 2023b), suggesting that the  $\delta^{15}\text{N}$  baselines were generally similar between habitat types. Benthic top predators can also exert top-down trophic control on benthic-associated prey that more pelagic- or sympagic-associated consumers also use (Amiraux et al., 2023a). In addition, demersal fishes, such as several sculpin species, feed on a variety of resources from both pelagic and benthic habitats in the Arctic (Landry et al., 2018) and therefore would show some variability in  $\delta^{34}\text{S}$  value across species, which aligns with our results where Arctic sculpin had the highest  $\delta^{34}\text{S}$  value (19.1 ‰) and Arctic staghorn sculpin having the lowest  $\delta^{34}\text{S}$  value (13.6 ‰). Furthermore, most mobile species, especially those at a higher trophic level, are generalist consumers that exhibit habitat coupling by foraging

on numerous prey items across benthic, pelagic or sympagic habitats in response to varying resource availability (Rooney et al., 2006; McMeans et al., 2013; Young and Ferguson, 2013).

Eco-physiological species traits can also influence trophic and THg dynamics within and across benthic, pelagic and benthopelagic habitats in aquatic systems (Lavoie et al., 2013; Vainio et al., 2022). Despite homeotherms (e.g., marine mammals) having a higher energy demand than poikilotherms (e.g., fishes) due to higher food consumption rates and thus a higher potential intake of Hg, their influence on estimated biomagnification factors and trophic magnification slopes across habitat type is varied from no effect on the trophic magnification slope (Lavoie et al., 2013) to a high effect (Fisk et al., 2001; Borgå et al., 2012). However, Vainio et al. (2022) found that the addition of homeothermic birds, who can consume resources across habitat types, to their trophic magnification models that included ectotherms did not show different trophic magnification factors between benthic and pelagic habitats, which was in contrast to their models excluding birds. Since Arctic ecosystems consist of numerous species with different metabolic strategies that also consume resources from multiple carbon sources or habitats, it is likely that both foraging and thermoregulatory strategies of species within and across habitat types influences trophic contaminant dynamics in an Arctic ecosystem.

#### 4.3. Relationships between $\delta^{13}\text{C}$ , $\delta^{34}\text{S}$ and THg of fishes and marine mammals

When incorporating several habitat source variables, such as  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$ , in models investigating THg variation across a food web, we found that  $\delta^{34}\text{S}$  had a higher range and standard deviation than  $\delta^{13}\text{C}$  across species (5.5 versus 4.5, and 1.1 versus 1.5, respectively) and was a better predictor of THg variation than  $\delta^{13}\text{C}$ , as has been observed in other aquatic studies (Evans and Crumley, 2005; Elliott and Elliott, 2016; Willacker et al., 2017; Elliott et al., 2021). In many coastal areas across the Arctic, the  $\delta^{13}\text{C}$  values of pelagic consumers can be higher than benthic consumers (Szpak and Buckley, 2020) or can largely overlap and the average difference in  $\delta^{13}\text{C}$  values can be small (e.g., within 4 ‰; Hobson et al., 2002; McMeans et al., 2013; Søreide et al., 2013; Linnebjerg et al., 2016; Amiraux et al., 2023b), likely due to the tight sympagic-pelagic-benthic coupling that drives energy flow between these habitats and is key to Arctic ecosystem functioning (Wassmann et al., 2004). This small difference in  $\delta^{13}\text{C}$  between benthic and pelagic consumers supports the use of a multi-isotopic approach that includes  $\delta^{34}\text{S}$  as opposed to solely using  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  when investigating Hg trophic dynamics in a food web.

Total mercury concentrations across species increased with  $\delta^{34}\text{S}$ . This supports the sulfate availability hypothesis which states that the supply of sulfate is mediated by sulfate-reducing bacteria in the  $^{34}\text{S}$ -enriched water column. Therefore, species with higher THg concentrations are likely foraging on prey in habitats richer in sulfates within the water column and on prey that are outside of the benthic microbial-based food web and which are higher in  $^{34}\text{S}$ -depleted sulfides (Peterson and Fry, 1987). Similar to this study, the origin of sulfur also drove THg variation across several seabird species and their prey in the Pacific Ocean (Elliott and Elliott, 2016), as well as in small forage fishes inhabiting different estuarine wetland systems (Willacker et al., 2017). In contrast, Góngora et al. (2018) found a negative correlation between  $\delta^{34}\text{S}$  and THg among thick-billed murre prey species around Southampton Island, suggesting higher levels of sulfides in prey items. The difference between studies could be due to some of the different prey fish species collected and the inclusion of invertebrates in Góngora et al. (2018) that may be a part of the benthic microbial food web, whereas this study included more top predator fishes and marine mammals.

A few caveats could slightly affect our data interpretation. First, variation in protein content (e.g., cysteine) and protein quality of a predators' diet can influence their  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  discrimination which could vary slightly among species in this study (McCutchan Jr

et al., 2003; Caut et al., 2009). However, the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  discrimination factors of marine fishes ( $\Delta^{13}\text{C}$ : 1.7 ‰;  $\Delta^{15}\text{N}$ : 3.7 ‰; Canseco et al., 2021), is comparable to that of piscivorous marine mammals ( $\Delta^{13}\text{C}$ : 1.3 ‰;  $\Delta^{15}\text{N}$ : 2.4 ‰; Hobson et al., 1996), and slight differences between the two species groups in  $\Delta^{13}\text{C}$  (0.4 ‰) and  $\Delta^{15}\text{N}$  (1.3 ‰) is much lower than the ranges of  $\delta^{13}\text{C}$  (4.5 ‰) and  $\delta^{15}\text{N}$  (5.5 ‰) observed across species in this study. In addition,  $\Delta^{34}\text{S}$  can significantly vary among consumers of a higher protein diet versus a lower protein diet by approximately 2.5 ‰ (Matthews and Ferguson, 2015), however this value is much lower than the range in  $\delta^{34}\text{S}$  among species in this study (5.5 ‰). Second, though fish and marine mammal samples were collected from different years, we could not examine inter-annual variability in trophic interactions in relation to environmental metrics due to low sample sizes for most species. However, the influence of inter-annual variability on consumer stable isotope values is likely negligible since standard deviations of  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  for each species were generally low (<1.0 ‰). Sea surface temperatures can influence baseline  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values and although surface temperature can vary by up to 8 °C over the upper 30 m around Southampton Island (Castro de la Guardia et al., 2023), water temperatures at the depth of the chlorophyll max (averaged 27 m; Kitching, 2022) and below where many of these species were collected did not vary more than a few degrees Celsius (Castro de la Guardia et al., 2023). Third, terrestrial or freshwater inputs around Southampton Island could slightly influence relationships between THg and  $\delta^{34}\text{S}$  across species since terrestrial and freshwater resources are typically depleted in  $^{34}\text{S}$  (McCutchan Jr et al., 2003). Southampton Island is found at the confluence of water masses from the Pacific Ocean, Atlantic Ocean, and the Hudson Bay Complex, which receives large amounts of riverine input during the open-water period, but this primarily occurs in southwestern Hudson Bay and James Bay (Stewart and Barber, 2010). Summer circulation of seawater around Southampton Island originates from Foxe Basin to the north and Hudson Strait to the east where both areas are comprised of cold, saline water (~32 PSU) suggesting minimal freshwater input in this area of Hudson Bay (Stewart and Barber, 2010). Nonetheless, some of the species studied are mobile or migratory and therefore their THg and  $\delta^{34}\text{S}$  values could be influenced from other regions in which they feed during other seasons.

## 5. Conclusions

This study provided new insights into the mercury and trophic dynamics for part of the coastal marine food web around Southampton Island: (1) THg concentrations were highest in beluga and narwhal among marine mammals and in shorthorn sculpin, fourline snakeblenny, Arctic shanny, Greenland cod and Arctic sculpin among fishes, (2) THg concentrations for each species were generally similar to their conspecifics from other areas across the Arctic, (3) the trophic magnification slope was comparable to several other marine areas across the Arctic, and (4)  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  were better predictors of THg variation across the food web than  $\delta^{13}\text{C}$ , highlighting the importance of a multi-isotopic approach that includes  $\delta^{34}\text{S}$  when investigating the trophic dynamics of mercury. Our study provides a marine ecosystem assessment of THg concentrations and its relationship to  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and  $\delta^{34}\text{S}$  for 19 species inhabiting marine waters around Southampton Island that can be used to monitor future changes. With climate change continuing to have pronounced effects on sea ice dynamics, primary productivity, and species interactions in the Arctic (Post et al., 2013), and likely on mercury in the Arctic (Chételat et al., 2022; McKinney et al., 2022), long-term monitoring of mercury and food web dynamics across coastal marine systems is imperative to inform national and international conservation and management activities.

## CRedit authorship contribution statement

D.J.Y. and M.A.M. conceived and designed the study. D.J.Y.

conducted the data analysis and drafted the manuscript with input from M.A.M. E.M. conducted the THg analysis under the supervision of N.B. and M.A.M. A.T.F. conducted the stable isotope analysis. All co-authors contributed samples and input at all stages, and approved the final version.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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## References

- AMAP, 2021. AMAP Assessment 2021: Mercury in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Tromsø, Norway, p. 324.
- Amiriaux, R., Archambault, P., Moriceau, B., Lemire, M., Babin, M., Memery, L., Massé, G., Tremblay, J.E., 2021. Efficiency of sympagic-benthic coupling revealed by analyses of n-3 fatty acids, IP25 and other highly branched isoprenoids in two filter-feeding Arctic benthic molluscs: *Mya truncata* and *Serripes groenlandicus*. *Org. Geochem.* 151, 104160 <https://doi.org/10.1016/j.orggeochem.2020.104160>.
- Amiriaux, R., Yurkowski, D.J., Archambault, P., Pierrejean, M., Mundy, C.J., 2023a. Top predator sea stars are the benthic equivalent to polar bears of the pelagic realm. *Proc. Natl. Acad. Sci.* 120 <https://doi.org/10.1073/pnas.2216701112> (p. e2216701120).
- Amiriaux, R., Mundy, C.J., Pierrejean, M., Niemi, A., Hedges, K.J., Brown, T.A., Ehns, J. K., Elliott, K.H., Ferguson, S.H., Fisk, A.T., Gilchrist, G.H., Harris, L.N., Iken, K., Jacobs, K., Johnson, K.F., Kuzyk, Z.Z., Limoges, A., Loewen, T.N., Love, O.P., Matthews, C.J.D., Ogloff, W.R., Rosenberg, B., Søreide, J., Watt, C.A., Yurkowski, D. J., 2023b. Tracing Carbon Flow and Trophic Structure of a Coastal Arctic Marine Food Web Using Highly Branched Isoprenoids, and Carbon, Nitrogen and Sulfur Stable Isotopes. *Ecol. Indic.* Accepted.
- 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/f98-001>.
- Barst, B.D., Chételat, J., Basu, N., 2022. Toxicological risk of mercury for fish and invertebrate prey in the Arctic. *Sci. Total Environ.* 836, 155702 <https://doi.org/10.1016/j.scitotenv.2022.155702>.
- Basu, N., Abass, K., Dietz, R., Kruemmel, E., Rautio, A., Weihe, P., 2022. The impact of mercury contamination on human health in the Arctic: a state of the science review. *Sci. Total Environ.* 831, 154793 <https://doi.org/10.1016/j.scitotenv.2022.154793>.
- Bligh, E.G., Dyer, W.J., 1959. A rapid method of total lipid extraction and purification. *Can. J. Biochem. Physiol.* 37, 911–917. <https://doi.org/10.1139/o59-099>.
- Borgå, K., Kidd, K.A., Muir, D.C., Berglund, O., Conder, J.M., Gobas, F.A., Kucklick, J., Malm, O., Powell, D.E., 2012. Trophic magnification factors: considerations of ecology, ecosystems, and study design. *Integr. Environ. Assess. Manag.* 8, 64–84. <https://doi.org/10.1002/ieam.244>.
- Braune, B., Chételat, J., Amyot, M., Brown, T., Clayden, M., Evans, M., Fisk, A., Gaden, A., Girard, C., Hare, A., Kirk, J., Lehnerr, I., Letcher, R., Loseto, L.,



- Macdonald, R., Mann, E., McMeans, B., Muir, D.C.G., 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, 67–90. <https://doi.org/10.1016/j.scitotenv.2014.05.133>.
- Brown, T.M., Fisk, A.T., Wang, X., Ferguson, S.H., Young, B.G., Reimer, K.J., Muir, D.C.G., 2016. Mercury and cadmium in ringed seals in the Canadian Arctic: influence of location and diet. *Sci. Total Environ.* 545, 503–511. <https://doi.org/10.1016/j.scitotenv.2015.12.030>.
- Brunner, B., Bernasconi, S.M., 2005. A revised isotope fractionation model for dissimilatory sulfate reduction in sulfate reducing bacteria. *Geochim. Cosmochim. Acta* 69, 4759–4771. <https://doi.org/10.1016/j.gca.2005.04.015>.
- Burnham, K.P., Anderson, D.R., 2004. Multimodel inference: understanding AIC and BIC in model selection. *Sociol. Methods Res.* 33, 261–304. <https://doi.org/10.1177/0049124104268644>.
- Campbell, L.M., Norstrom, R.J., Hobson, K.A., Muir, D.C., Backus, S., Fisk, A.T., 2005. Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay). *Sci. Total Environ.* 351, 247–263. <https://doi.org/10.1016/j.scitotenv.2005.02.043>.
- Canseco, J.A., Niklitschek, E.J., Harrod, C., 2021. Variability in  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  trophic discrimination factors for teleost fishes: a meta-analysis of temperature and dietary effects. *Rev. Fish Biol. Fish.* 32, 313–329. <https://doi.org/10.1007/s11160-021-09689-1>.
- Castro de la Guardia, L., Filbee-Dexter, K., Reimer, J., MacGregor, K.A., Garrido, I., Singh, R.K., Bélanger, S., Konar, B., Iken, K., Johnson, L.E., Archambault, P., Sejr, M. K., Søreide, J.E., Mundy, C., 2023. Increasing depth distribution of Arctic kelp with increasing number of open water days with light. *Elem. Sci. Anth.* 11, 00051.
- Caut, S., Angulo, E., Courchamp, F., 2009. Variation in discrimination factors ( $\Delta^{15}\text{N}$  and  $\Delta^{13}\text{C}$ ): the effect of diet isotopic values and applications for diet reconstruction. *J. Appl. Ecol.* 46, 443–453. <https://doi.org/10.1111/j.1365-2664.2009.01620.x>.
- Chételat, J., McKinney, M.A., Amyot, M., Dastoor, A., Douglas, T.A., Heimbürger-Boavida, L.E., Kirk, J., Kahilainen, K.K., Outridge, P.M., Pelletier, N., Skov, H., 2022. Climate change and mercury in the Arctic: abiotic interactions. *Sci. Total Environ.* 824, 153715.
- Clark, B., 1980. The Lake site (KkHh-2), Southampton Island, NWT and its position in Sadlermiut prehistory. *Can. J. Archaeol.* 4, 53–81. <https://www.jstor.org/stable/41102220>.
- Clayden, M.G., Kidd, K.A., Wyn, B., Kirk, J.L., Muir, D.C., 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., Arsenault, L.M., Kidd, K.A., O'Driscoll, N.J., Mallory, M.L., 2015. Mercury bioaccumulation and biomagnification in a small Arctic polynya ecosystem. *Sci. Total Environ.* 509, 206–215. <https://doi.org/10.1016/j.scitotenv.2014.07.087>.
- Connolly, R.M., Guest, M.A., Melville, A.J., Oakes, J.M., 2004. Sulfur stable isotopes separate producers in marine food-web analysis. *Oecologia* 138, 161–167. <https://doi.org/10.1007/s00442-003-1415-0>.
- Dietz, R., Riget, F., Born, E.W., Sonne, C., Grandjean, P., Kirkegaard, M., Olsen, M.T., Asmund, G., Renzoni, A., Baagø, H., Andreasen, C., 2006. Trends in mercury in hair of greenlandic polar bears (*Ursus maritimus*) during 1892–2001. *Environ. Sci. Technol.* 40, 1120–1125. <https://doi.org/10.1021/es051636z>.
- Dietz, R., Letcher, R.J., Desforges, J.P., Eulaers, I., Sonne, C., Wilson, S., Andersen-Ranberg, E., Basu, N., Barst, B.D., Bustnes, J.O., Bytingsvik, J., 2019. Current state of knowledge on biological effects from contaminants on arctic wildlife and fish. *Sci. Total Environ.* 696, 133792. <https://doi.org/10.1016/j.scitotenv.2019.133792>.
- Dietz, R., Letcher, R.J., Aars, J., Andersen, M., Boltunov, A., Born, E.W., Ciesielski, T.M., Das, K., Dastnai, S., Derocher, A.E., Desforges, J.P., 2022. A risk assessment review of mercury exposure in Arctic marine and terrestrial mammals. *Sci. Total Environ.* 829, 154445. <https://doi.org/10.1016/j.scitotenv.2022.154445>.
- 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, 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., Braune, B.M., Elliott, J.E., 2021. Beyond bulk  $\delta^{15}\text{N}$ : combining a suite of stable isotopic measures improves the resolution of the food webs mediating contaminant signals across space, time and communities. *Environ. Int.* 148, 106370. <https://doi.org/10.1016/j.envint.2020.106370>.
- Evans, D.W., Crumley, P.H., 2005. Mercury in Florida Bay fish: spatial distribution of elevated concentrations and possible linkages to Everglades restoration. *Bull. Mar. Sci.* 77, 321–346.
- Fisk, A.T., Hobson, K.A., Norstrom, R.J., 2001. Influence of chemical and biological factors on trophic transfer of persistent organic pollutants in the Northwater Polynya marine food web. *Environ. Sci. Technol.* 35, 732–738. <https://doi.org/10.1021/es001459w>.
- Florin, S.T., Felicetti, L.A., Robbins, C.T., 2011. The biological basis for understanding and predicting dietary-induced variation in nitrogen and sulphur isotope ratio discrimination. *Funct. Ecol.* 25, 519–526. <https://doi.org/10.1111/j.1365-2435.2010.01799.x>.
- Garde, E., Hansen, S.H., Ditlevsen, S., Tvermosegaard, K.B., Hansen, J., Harding, K.C., Heide-Jørgensen, M.P., 2015. Life history parameters of narwhals (*Monodon monoceros*) from Greenland. *J. Mammal.* 96, 866–879. <https://doi.org/10.1093/jmammal/gyv110>.
- Gaston, A.J., Woo, K., Hipfner, J.M., 2003. Trends in forage fish populations in northern Hudson Bay since 1981, as determined from the diet of nestling thick-billed murres *Uria lomvia*. *Arctic* 227–233. <https://www.jstor.org/stable/40512539>.
- Giraldo, C., Stasko, A., Choy, E.S., Rosenberg, B., Majewski, A., Power, M., Swanson, H., Loseto, L., Reist, J.D., 2016. Trophic variability of Arctic fishes in the Canadian Beaufort Sea: a fatty acids and stable isotopes approach. *Polar Biol.* 39, 1267–1282. <https://doi.org/10.1007/s00300-015-1851-4>.
- Golzadeh, N., Barst, B.D., Basu, N., Baker, J.M., Auger, J.C., McKinney, M.A., 2020. Evaluating the concentrations of total mercury, methylmercury, selenium, and selenium: mercury molar ratios in traditional foods of the Bigstone Cree in Alberta, Canada. *Chemosphere* 250, 126285. <https://doi.org/10.1016/j.chemosphere.2020.126285>.
- Góngora, E., Braune, B.M., Elliott, K.H., 2018. Nitrogen and sulfur isotopes predict variation in mercury levels in Arctic seabird prey. *Mar. Pollut. Bull.* 135, 907–914. <https://doi.org/10.1016/j.marpolbul.2018.07.075>.
- Hammill, M.O., 2009. Ringed seal: *Pusa hispida*. In: *Encyclopedia of Marine Mammals*. Academic Press, pp. 972–974.
- Hansen, C.T., Nielsen, C.O., Dietz, R., Hansen, M.M., 1990. Zinc, cadmium, mercury and selenium in minke whales, belugas and narwhals from West Greenland. *Polar Biol.* 10, 529–539. <https://doi.org/10.1007/BF002233702>.
- Hansson, S.V., Desforges, J.P., van Beest, F.M., Bach, L., Halden, N.M., Sonne, C., Mosbech, A., Søndergaard, J., 2020. Bioaccumulation of mining derived metals in blood, liver, muscle and otoliths of two Arctic predatory fish species (*Gadus ogac* and *Myoxocephalus scorpius*). *Environ. Res.* 183, 109194. <https://doi.org/10.1016/j.envres.2020.109194>.
- Hilgendag, I.R., Swanson, H.K., Lewis, C.W., Ehrman, A.D., Power, M., 2022. Mercury biomagnification in benthic, pelagic, and benthopelagic food webs in an Arctic marine ecosystem. *Sci. Total Environ.* 841, 156424. <https://doi.org/10.1016/j.scitotenv.2022.156424>.
- Hobson, K.A., Schell, D.M., Renouf, D., Noseworthy, E., 1996. Stable carbon and nitrogen isotopic fractionation between diet and tissues of captive seals: implications for dietary reconstructions involving marine mammals. *Can. J. Fish. Aquat. Sci.* 53, 528–533. <https://doi.org/10.1139/f95-209>.
- Hobson, K.A., Fisk, A., Karnovsky, N., Holst, M., Gagnon, J.M., Fortier, M., 2002. A stable isotope ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ) model for the North Water food web: implications for evaluating trophodynamics and the flow of energy and contaminants. *Deep Sea Res. Part II: Top. Stud. Oceanogr.* 49, 5131–5150. [https://doi.org/10.1016/S0967-0645\(02\)00182-0](https://doi.org/10.1016/S0967-0645(02)00182-0).
- Jeremiason, J.D., Engstrom, D.R., Swain, E.B., Nater, E.A., Johnson, B.M., Almendinger, J.E., Monson, B.A., Kolka, R.K., 2006. Sulfate addition increases methylmercury production in an experimental wetland. *Environ. Sci. Technol.* 40, 3800–3806. <https://doi.org/10.1021/es0524144>.
- Keats, D.W., Steele, D.H., Green, J.M., Martel, G.M., 1993. Diet and population size structure of the Arctic shanny, *Stichaeus punctatus* (Pisces: Stichaeidae), at sites in eastern Newfoundland and the eastern Arctic. *Environ. Biol. Fish.* 37, 173–180.
- Kelley, T.C., Stewart, R.E., Yurkowski, D.J., Ryan, A., Ferguson, S.H., 2015. Mating ecology of beluga (*Delphinapterus leucas*) and narwhal (*Monodon monoceros*) as estimated by reproductive tract metrics. *Mar. Mamm. Sci.* 31, 479–500.
- Kidd, K.A., Clayden, M., Jardine, T.D., 2012. Bioaccumulation and biomagnification of mercury in food webs. In: Liu, G., Cai, Y., O'Driscoll, N. (Eds.), *Environmental Chemistry and Toxicology of Mercury*. John Wiley & Sons, Inc, Hoboken, NJ, pp. 453–499.
- Kitching, E., 2022. Physical Processes Driving Phytoplankton Production Around Southampton Island, Nunavut in Late Summer 2018 and 2019. MSc Thesis.. University of Manitoba <http://hdl.handle.net/1993/36865>.
- Kortsch, S., Primicerio, R., Fosheim, M., Dolgov, A.V., Aschan, M., 2015. Climate change alters the structure of arctic marine food webs due to poleward shifts of boreal generalists. *Proc. R. Soc. B Biol. Sci.* 282, 20151546. <https://doi.org/10.1098/rspb.2015.1546>.
- Kovacs, K.M., Citta, J., Brown, T., Dietz, R., Ferguson, S., Harwood, L., Houde, M., Lea, E. V., Quakenbush, L., Riget, F., Rosing-Asvid, A., 2021. Variation in body size of ringed seals (*Pusa hispida hispida*) across the circumpolar Arctic: evidence of morphs, ecotypes or simply extreme plasticity? *Polar Res.* 40, 5753. <https://doi.org/10.33265/polar.v40.5753>.
- Landry, J.J., Fisk, A.T., Yurkowski, D.J., Hussey, N.E., Dick, T., Crawford, R.E., Kessel, S. T., 2018. Feeding ecology of a common benthic fish, shorthorn sculpin (*Myoxocephalus scorpius*) in the high arctic. *Polar Biol.* 41, 2091–2102. <https://doi.org/10.1007/s00300-018-2348-8>.
- Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A., Campbell, L.M., 2013. Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. *Environ. Sci. Technol.* 47, 13385–13394. <https://doi.org/10.1021/es403103t>.
- Layman, C.A., Araujo, M.S., Boucek, R., Hammerschlag-Peyer, C.M., Harrison, E., Jud, Z. R., Matich, P., Rosenblatt, A.E., Vaudo, J.J., Yeager, L.A., Post, D.M., 2012. Applying stable isotopes to examine food-web structure: an overview of analytical tools. *Biol. Rev.* 87, 545–562. <https://doi.org/10.1111/j.1469-185X.2011.00208.x>.
- Lewis, S.L., Maslin, M.A., 2015. Defining the Anthropocene. *Nature* 519, 171–180. <https://doi.org/10.1038/nature14258>.
- Lewis, K.M., Van Dijken, G.L., Arrigo, K.R., 2020. Changes in phytoplankton concentration now drive increased Arctic Ocean primary production. *Science* 369, 198–202. <https://doi.org/10.1126/science.aag838>.
- Linnebjerg, J.F., Hobson, K.A., Fort, J., Nielsen, T.G., Møller, P., Wieland, K., Born, E.W., Rigét, F.F., Mosbech, A., 2016. Deciphering the structure of the West Greenland marine food web using stable isotopes ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ). *Mar. Biol.* 163, 1–15. <https://doi.org/10.1007/s00227-016-3001-0>.
- Loewen, T.N., Hornby, C.A., Johnson, M., Chambers, C., Dawson, K., MacDonell, D., Bernhardt, W., Gnanapragasam, R., Pierrejean, M., Choy, E., 2020. Ecological and Biophysical Overview of the Southampton Proposed Area of Interest for the Southampton Island Ecologically and Biologically Significant Area. DFO Can. Sci. Adv. Sec. Res. Doc. 2020/032. vi + 96 p.

- Loseto, L.L., Stern, G.A., Deibel, D., Connelly, T.L., Prokopowicz, A., Lean, D.R.S., Fortier, L., Ferguson, S.H., 2008. Linking mercury exposure to habitat and feeding behaviour in Beaufort Sea beluga whales. *J. Mar. Syst.* 74, 1012–1024. <https://doi.org/10.1016/j.jmarsys.2007.10.004>.
- Louis, M., Skovrind, M., Garde, E., Heide-Jørgensen, M.P., Szpak, P., Lorenzen, E.D., 2021. Population-specific sex and size variation in long-term foraging ecology of belugas and narwhals. *R. Soc. Open Sci.* 8, 202226 <https://doi.org/10.1098/rsos.202226>.
- Luque, S.P., Ferguson, S.H., 2010. Age structure, growth, mortality, and density of belugas (*Delphinapterus leucas*) in the Canadian Arctic: responses to environment? *Polar Biol.* 33, 163–178. <https://doi.org/10.1007/s00300-009-0694-2>.
- Lydersen, C., 2018. Walrus: *Odobenus rosmarus*. In: *Encyclopedia of Marine Mammals*. Academic Press, pp. 1045–1048.
- Matthews, C.J., Ferguson, S.H., 2015. Seasonal foraging behaviour of Eastern Canada-West Greenland bowhead whales: an assessment of isotopic cycles along baleen. *Mar. Ecol. Prog. Ser.* 522, 269–286. <https://doi.org/10.3354/meps11145>.
- Mazerolle, M.J., Mazerolle, M.M.J., 2017. Package 'AICcmodavg'. R package, 281.
- McCutchan Jr., J.H., Lewis Jr., W.M., Kendall, C., McGrath, C.C., 2003. Variation in trophic shift for stable isotope ratios of carbon, nitrogen, and sulfur. *Oikos* 102, 378–390. <https://doi.org/10.1034/j.1600-0706.2003.12098.x>.
- McKinney, M.A., Chételat, J., Burke, S.M., Elliott, K.H., Fernie, K.J., Houde, M., Kahilainen, K.K., Letcher, R.J., Morris, A.D., Muir, D.C.G., Routti, H., Yurkowski, D. J., 2022. Climate change and mercury in the Arctic: biotic interactions. *Sci. Total Environ.* 834, 155221 <https://doi.org/10.1016/j.scitotenv.2022.155221>.
- McMeans, B.C., Rooney, N., Arts, M.T., Fisk, A.T., 2013. Food web structure of a coastal Arctic marine ecosystem and implications for stability. *Mar. Ecol. Prog. Ser.* 482, 17–28. <https://doi.org/10.3354/meps10278>.
- McMeans, B.C., Arts, M.T., Fisk, A.T., 2015. Impacts of food web structure and feeding behavior on mercury exposure in Greenland sharks (*Somniosus microcephalus*). *Sci. Total Environ.* 509, 216–225. <https://doi.org/10.1016/j.scitotenv.2014.01.128>.
- Minagawa, M., Wada, E., 1984. Stepwise enrichment of  $^{15}\text{N}$  along food chains: further evidence and the relation between  $\delta^{15}\text{N}$  and animal age. *Geochim. Cosmochim. Acta* 48, 1135–1140. [https://doi.org/10.1016/0016-7037\(84\)90204-7](https://doi.org/10.1016/0016-7037(84)90204-7).
- Morris, A.D., Wilson, S.J., Fryer, R.J., Thomas, P.J., Hudelson, K., Andreassen, B., Blévin, P., Bustamante, P., Chastel, O., Christensen, G., Dietz, R., Evans, M., Evensen, A., Ferguson, S.H., Fort, J., Gamberg, M., Grémillet, D., Houde, M., Letcher, R.J., Loseto, L., Muir, D.C.G., Pinzone, M., Poste, A., Routti, H., Sonne, C., Stern, G., Rigét, F., 2022. Temporal trends of mercury in Arctic biota: 10 more years of progress in Arctic monitoring. *Sci. Total Environ.* 839, 155803 <https://doi.org/10.1016/j.scitotenv.2022.155803>.
- Pedro, S., Fisk, A.T., Tomy, G.T., Ferguson, S.H., Hussey, N.E., Kessel, S.T., McKinney, M. A., 2017. Mercury and persistent organic pollutants in native and invading forage species of the Canadian Arctic: consequences for food web dynamics. *Environ. Pollut.* 229, 229–240. <https://doi.org/10.1016/j.envpol.2017.05.085>.
- Pedro, S., Fisk, A.T., Ferguson, S.H., Hussey, N.E., Kessel, S.T., McKinney, M.A., 2019. Limited effects of changing prey fish communities on food quality for aquatic predators in the eastern Canadian Arctic in terms of essential fatty acids, methylmercury and selenium. *Chemosphere* 214, 855–865. <https://doi.org/10.1016/j.chemosphere.2018.09.167>.
- Peterson, B.J., Fry, B., 1987. Stable isotopes in ecosystem studies. *Annu. Rev. Ecol. Evol.* 18, 293–320. <https://www.jstor.org/stable/2097134>.
- Post, D.M., Layman, C.A., Arrington, D.A., Takimoto, G., Quattrochi, J., Montana, C.G., 2007. Getting to the fat of the matter: models, methods and assumptions for dealing with lipids in stable isotope analyses. *Oecologia* 152, 179–189. <https://doi.org/10.1007/s00442-006-0630-x>.
- Post, E., Bhatt, U.S., Bitz, C.M., Brodie, J.F., Fulton, T.L., Hebblewhite, M., Kerby, J., Kutz, S.J., Stirling, I., Walker, D.A., 2013. Ecological consequences of sea-ice decline. *Science* 341, 519–524. <https://doi.org/10.1126/science.1235225>.
- R Core Team, 2021. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria, p. 2012.
- Rigét, F., Møller, P., Dietz, R., Nielsen, T.G., Asmund, G., Strand, J., Larsen, M.M., Hobson, K.A., 2007. Transfer of mercury in the marine food web of West Greenland. *J. Environ. Monit.* 9, 877–883. <https://doi.org/10.1039/B704796G>.
- Rooney, N., McCann, K., Gellner, G., Moore, J.C., 2006. Structural asymmetry and the stability of diverse food webs. *Nature* 442, 265–269. <https://doi.org/10.1038/nature04887>.
- Søreide, J.E., Carroll, M.L., Hop, H., Ambrose Jr., W.G., Hegseth, E.N., Falk-Petersen, S., 2013. Sympagic-pelagic-benthic coupling in Arctic and Atlantic waters around Svalbard revealed by stable isotopic and fatty acid tracers. *Mar. Biol. Res.* 9, 831–850. <https://doi.org/10.1080/17451000.2013.775457>.
- Stewart, D.B., Barber, D.G., 2010. The ocean-sea ice-atmosphere system of the Hudson Bay complex. In: *A Little Less Arctic: Top Predators in the World's Largest Northern Inland Sea, Hudson Bay*, pp. 1–38.
- Sun, M.Y., Clough, L.M., Carroll, M.L., Dai, J., Ambrose Jr., W.G., Lopez, G.R., 2009. Different responses of two common Arctic macrobenthic species (*Macoma balthica* and *Monoporeia affinis*) to phytoplankton and ice algae: Will climate change impacts be species specific? *J. Exp. Mar. Biol. Ecol.* 376, 110–121.
- Szpak, P., Buckley, M., 2020. Sulfur isotopes ( $\delta^{34}\text{S}$ ) in Arctic marine mammals: indicators of benthic vs. pelagic foraging. *Mar. Ecol. Prog. Ser.* 653, 205–216.
- Vainio, R.K., Jormalainen, V., Dietz, R., Laaksonen, T., Schulz, R., Sonne, C., Søndergaard, J., Zubrod, J.P., Eulaers, I., 2022. Trophic dynamics of mercury in the Baltic Archipelago Sea food web: the impact of ecological and ecophysiological traits. *Environ. Sci. Technol.* 56, 11440–11448. <https://doi.org/10.1021/acs.est.2c03846>.
- Wassmann, P., Bauerfeind, E., Fortier, M., Fukuchi, M., Hargrave, B., Moran, B., Noji, T., Nöthig, E.M., Olli, K., Peinert, R., Sasaki, H., Shevchenko, V., 2004. Particulate organic carbon flux to the Arctic Ocean sea floor. In: *The Organic Carbon Cycle in the Arctic Ocean*. Springer, Berlin, Heidelberg, pp. 101–138.
- Watt, C.A., Heide-Jørgensen, M.P., Ferguson, S.H., 2013. How adaptable are narwhal? A comparison of foraging patterns among the world's three narwhal populations. *Ecosphere* 4, 1–15. <https://doi.org/10.1890/ES13-00137.1>.
- Watt, C.A., Orr, J., Ferguson, S.H., 2016. A shift in foraging behaviour of beluga whales *Delphinapterus leucas* from the threatened Cumberland Sound population may reflect a changing Arctic food web. *Endanger. Species Res.* 31, 259–270. <https://doi.org/10.3354/esr00768>.
- Wickham, H., 2016. Data analysis. In: *In ggplot2*. Springer, Cham, pp. 189–201.
- Willacker, J.J., Eagles-Smith, C.A., Ackerman, J.T., 2017. Mercury bioaccumulation in estuarine fishes: novel insights from sulfur stable isotopes. *Environ. Sci. Technol.* 51, 2131–2139. <https://doi.org/10.1021/acs.est.6b05325>.
- Worm, B., Paine, R.T., 2016. Humans as a hyperkeystone species. *Trends Ecol. Evol.* 31, 600–607. <https://doi.org/10.1016/j.tree.2016.05.008>.
- Young, B.G., Ferguson, S.H., 2013. Seasons of the ringed seal: pelagic open-water hyperphagy, benthic feeding over winter and spring fasting during molt. *Wildl. Res.* 40, 52–60. <https://doi.org/10.1071/WR12168>.
- Yurkowski, D.J., Hussey, N.E., Semeniuk, C., Ferguson, S.H., Fisk, A.T., 2015. Effects of lipid extraction and the utility of lipid normalization models on  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in Arctic marine mammal tissues. *Polar Biol.* 38, 131–143. <https://doi.org/10.1007/s00300-014-1571-1>.
- Yurkowski, D.J., Hussey, N.E., Fisk, A.T., Imrie, K.L., Tallman, R.F., Ferguson, S.H., 2017. Temporal shifts in intraguild predation pressure between beluga whales and Greenland halibut in a changing Arctic. *Biol. Lett.* 13, 20170433. <https://doi.org/10.1098/rsbl.2017.0433>.
- Yurkowski, D.J., Hussey, N.E., Ferguson, S.H., Fisk, A.T., 2018. A temporal shift in trophic diversity among a predator assemblage in a warming Arctic. *R. Soc. Open Sci.* 5, 180259 <https://doi.org/10.1098/rsos.180259>.
- Yurkowski, D.J., Auger-Méthé, M., Mallory, M.L., Wong, S.N., Gilchrist, G., Derocher, A. E., Richardson, E., Lunn, N.J., Hussey, N.E., Marcoux, M., Togunov, R.R., Fisk, A.T., Harwood, L.A., Dietz, R., Rosing-Asvid, A., Born, E.W., Mosbech, A., Fort, J., Grémillet, D., Loseto, L., Richard, P.R., Iacozza, J., Jean-Gagnon, F., Brown, T.M., Westdal, K.H., Orr, J., LeBlanc, B., Hedges, K.J., Treble, M.A., Kessel, S.T., Blanchfield, P.J., Davis, S., Maftai, M., Spencer, N., McFarlane-Tranquilla, L., Montevecchi, W.A., Bartzan, B., Dickson, L., Anderson, C., Ferguson, S.H., 2019. Abundance and species diversity hotspots of tracked marine predators across the North American Arctic. *Divers. Distrib.* 25, 328–345. <https://doi.org/10.1111/ddi.12860>.
- Zuur, A.F., Ieno, E.N., Elphick, C.S., 2010. A protocol for data exploration to avoid common statistical problems. *Methods Ecol. Evol.* 1, 3–14.