Mercury (Hg) and organochlorine pesticides (OCP) in mesopelagic and demersal fish of the Labrador Sea and Canadian Arctic

by

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Abstract

Warming polar waters are leading to the decline of sea-ice and the expansion of subarctic species towards higher latitudes. Northward fish movement is shifting the fate of environmental pollutants in Arctic trophic webs. Here I measure contaminant concentrations for total mercury (THg), methylmercury (MeHg), and a range of legacy organochlorine pesticides (OCPs) in mesopelagic and demersal fish in the Labrador Sea and the Canadian Arctic collected between 2016 and 2021. Muscle tissue samples were collected from three pelagic fish species (Arctic cod [*Boreogadus saida*], capelin [*Mallotus villosus*], glacier lanternfish [*Benthosema glaciale*]) and three demersal fish (abyssal grenadier [*Coryphaenoides armatus*], blue hake [*Antimora rostrata*], and Greenland halibut [*Reinhardtius hippoglossoides*]). The objectives were to determine (1) how contaminant concentrations in pelagic and demersal fish vary spatially across the Canadian Arctic and subarctic regions; and (2) how ecology (measured from stable isotope ratios of $\delta^{15}N$ and $\delta^{13}C$) and habitat (pelagic and demersal) influence contaminant concentrations in these fish species. Hg concentrations were best explained by species, trophic level, area, and habitat, while OCP concentrations were explained by species, trophic level, and habitat. These findings can be used to predict future contaminant concentrations in Arctic trophic food webs given climate-driven ecological changes.

General Summary

Mercury and organochlorine pesticides reach even the most remote corners of the planet and the Canadian Arctic is no exception. As the Arctic warms, the loss of habitat and shift towards subarctic species impact contaminants levels in northern food webs. To understand how contaminants will change in the future, we must first understand which factors influence contaminant concentrations today. In this study, I sampled muscle tissue of 620 demersal (living in proximity to the seafloor) and pelagic (living in the water column) fish in the Labrador Sea and the Canadian Arctic from 2016 to 2021. I found (1) that larger fish living on the seafloor generally had higher contaminants than smaller fish living in the water column and (2) that mercury was higher in fish in the Beaufort Sea than the Canadian Arctic Archipelago and Baffin Bay.

Acknowledgements

My sincere thanks to my two co-supervisors, Drs. Maxime Geoffroy and Dave Cote and my committee member Dr. Tanya Brown. I would like to underline the contribution of Dr. Brown and her team at the Marine Mammal and Ecotoxicology lab who were instrumental in the planning, processing, and analysis of my data. I greatly appreciate your collective guidance, support, and diligence on this project. Special thanks to my colleagues in the Center for Fisheries Ecosystem Research, Pierre Priou, Alessandra Gentile, and Jordan Sutton, for their mentorship and support. Thank you to Jennifer Herbig for contributing many hours helping with labwork and for traveling to both the Arctic and Antarctic with me. I thank you also for your patience and your endless encouragement.

Thank to you Brian Burke and Baffin Fisheries for coordinating and supplying Greenland Halibut samples from Baffin Bay. I would also like to thank Andrew Majewski and Krystal Woodward for collecting and processing the muscle tissue from Arctic cod in the Beaufort Sea. Thank you to Mary Denniston who supplied me with capelin from Nain, Labrador. Thank you to Sheena Roul and the longlining crew of the *Clear Cove's Pride* for the collection of the demersal samples. I would also like to thank the coast guard crew of the CCGS *Amundsen* and the scientists who helped with net deployments and fish sorting; Cyril Aubry, Guillaume Blais, and Amy McAllister.

I would like to thank my parents, Manon Ruel and Colin Jacobsen, for their unwavering support throughout my academic journey. To Tami Jacobsen, Keith Williams, and Lauren Calderone, thank you for always believing me and cheering on in this new adventure. Lastly, I would like to thank my close friends, Shanti Thurber, Oonagh Fogarty, Kasey Vader, Samantha Balliet, who always provided encouragement and a listening ear even from across the country.

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Co-authorship statement

I primarily conducted the work in this thesis with the support of my co-supervisors, Drs. Dave Cote and Maxime Geoffroy, as well as my committee member Dr. Tanya Brown. The body of this thesis uses fish from mid-water, benthic bottom trawls, and longlines collected from 2017 to 2021 across the Canadian Arctic and the Labrador Sea. I started my program in January 2021 and completed my fieldwork that same year on the CCGS *Amundsen* in the Labrador Sea and Baffin Bay. Most fish samples had been collected previously by our collaborators and colleagues before the start of my program.

All muscle tissues were removed by Jennifer Herbig, Krystal Woodward, and me. Additional homogenizing of the sample pools was performed by Sofya Reger at Fisheries and Oceans in West Vancouver, British Columbia. Dr. Tanya Brown and her team at the Fisheries and Oceans Canada at the Pacific Sciences Enterprise Center analyzed the muscle tissues for total mercury. Dr. Brown also prepared the muscle tissues and funded the stable isotope analysis with Dr. Aaron Fisk at the University of Windsor. I conducted all the statistical analyses and manuscript preparation with editorial support from my entire committee.

The first chapter includes a literature review and general introduction of topics discussed in the body of the thesis. The core of the thesis (Chapter 2) examines contaminant variation (THg and OCPs) in pelagic and demersal fish from the Labrador Sea and Canadian Arctic. Chapter 2 was written as a manuscript that will be submitted for publication in a peer-reviewed scientific journal. Chapter 3 includes a general conclusion that puts the findings of this work into greater context. Included in the last chapter are also improvements and suggestions for future work.

Chapter 1 - General Introduction

1.1 Climate change in the Arctic

The Arctic is warming nearly four times faster than the global average, which is having profound effects on Arctic ecosystems (Overland & Wang, 2013; Rantanen et al., 2022). Despite there being regional differences in amplification rates across the Arctic, the North Atlantic-Arctic gateway still maintains a warming rate four times above the global average (Rantanen et al., 2022). The primary reason for high rates of amplification is the reduction of cold-season ice cover (i.e. more pronounced in the Barents Sea) and weaker ice-albedo feedback (Onarheim & Årthun, 2017; Isaksen et al., 2022). Arctic ecosystems are not only experiencing later sea ice freeze up, earlier ice breakup, and less snow cover, but also altered nutrient availability (Post, 2002; Kovacs et al., 2011; Wassmann et al., 2011; Post et al., 2013). As such, the decline of sea ice extent and thickness has been considered one of the most impactful effects of global climate change (Meredith et al., 2019) because it acts as a negative feedback loop accelerating the rates of sea surface and bottom water temperature increase (Screen & Simmonds, 2010; Pistone et al., 2014). Sea ice also provides refuge for fish and marine mammals like Arctic cod (*Boreogadus saida*), ringed seals (*Pusa hispida*) and polar bears (*Ursus maritimus*) which have all shown changes in habitat use, feeding ecology, abundance, and/or reproductive success as a result of sea ice loss (Stirling, 2005; Stirling & Parkinson, 2009; McKinney et al., 2009; Regehr et al., 2010; Provencher et al., 2012; Huserbråten et al., 2019; Ferguson et al., 2017) . Increased sea surface temperature and reduced sea ice cover have cascading effects on long-term food availability for Inuit in the Canadian Arctic.

Environmental changes in Arctic ecosystems continue to have direct effects on endemic species. For example, Arctic cod is the most abundant forage fish in the Arctic Ocean (Geoffroy et al. in press). In certain regions of the Arctic Ocean, they are responsible for shuttling more than 70% of the energy from the pelagic food web to endothermic predators (e.g. marine mammals and seabirds) (Bradstreet,1982; Welch et al., 1992; Steiner et al., 2019). Arctic cod rely on sea ice habitats in their early life stages (e.g. eggs, larvae, some immatures). During these early life stages, they use sea ice to shelter from predators and can feed on sympagic (ice-associated) zooplankton nauplii (Bradstreet, 1982; Lønne & Gulliksen, 1989; David et al., 2016; Kohlbach et al., 2016, 2017). Fatty acid profiles have indicated that diatoms associated with sea ice (e.g. ice algae) are the primary carbon source of immature Arctic cod in the European high Arctic (Kohlbach et al., 2017), but not in the Canadian Arctic where they mainly feed on copepods (Hop and Crawford, 1992; McNicholl et al., 2016). However, as Arctic cod age they become less sympagic and more benthopelagic, meaning that their reliance on under sea ice habitats for feeding, protection, and energy maintenance declines (Benoit et al., 2008, 2010; Geoffroy et al., 2011, 2016; Hop & Gjøsæter, 2013). For adult Arctic cod in the Barents Sea (Dupont et al., 2020, 2021) and Canadian Arctic (Bouchard et al., 2018), total biomass was either negatively or not related to sea ice.

1.2 Borealization of Arctic communities

Trophic niches in the Arctic are shifting with climate change (McKinney et al., 2022). The rapidly warming waters are leading to the expansion of subarctic species towards higher latitudes, which is influencing the diet, distribution, and overall fitness of top predators (Laidre et al., 2008; Marcoux et al., 2012; Watt & Ferguson, 2015; Bryndum-Buchholz et al., 2020a,b). Because endemic species have narrower thermal ranges and are temporally adapted to the Arctic (Sunday et al., 2019), they are generally more vulnerable to environmental change (Dulvy et al., 2003; Newsome et al., 2012; Yurkowski et al., 2016; Lavoie et al., 2018) . Yet, Arctic amplification is not the same across the entire Arctic and species-specific responses to warming vary across regions and habitats (Parkinson & Cavalieri, 2002; Hanna, 2003; Stern & Heide-Jørgensen, 2003; Stirling & Parkinson, 2009).

1.3 Contaminants in the Arctic

Despite being isolated from most anthropogenic sources, contaminants still reach the Arctic (Brown et al., 2018). Legacy contaminants (i.e. chemicals once used but then discontinued or banned) and emerging contaminants (i.e. chemicals that are not currently regulated) have been identified in the Arctic and in the subarctic (McKinney et al., 2015; Pedro, 2017; Brown et al., 2018). They persist in soil, biota, and water where they concentrate in fatty tissues which is then passed onto offspring through milk and eggs (Hernández et al., 1994; Zhang et al., 2016; Nowosad et al., 2018). Inuit rely heavily on harvested food. Unfortunately, legacy and emerging contaminants stored in fish tissues have implications for Indigenous Peoples with most notable adverse effects on childbearing women (Binnington et al., 2016; AMAP, 2021).

In the Northwest Atlantic, contaminants enter the Labrador Sea and the Canadian Arctic primarily through long-range transport via atmosphere/water mass exchanges and to a lesser, yet important, extent through biological importation (Blais et al., 2007; Brown et al., 2018). Bioaccumulation is the increase in concentration of contaminants in an individual over time, while biomagnification is the increase in concentration of contaminants up the food web (Figure 1). Physiological interactions between chemicals and biota (e.g., uptake, metabolism, excretion) (Scheuhammer et al., 2012) as well as climate-driven ecological changes (e.g. trophic level) influence the effects of contaminants on biota (Buck et al., 2019; Ordiano-Flores et al., 2021; McKinney et al., 2022).

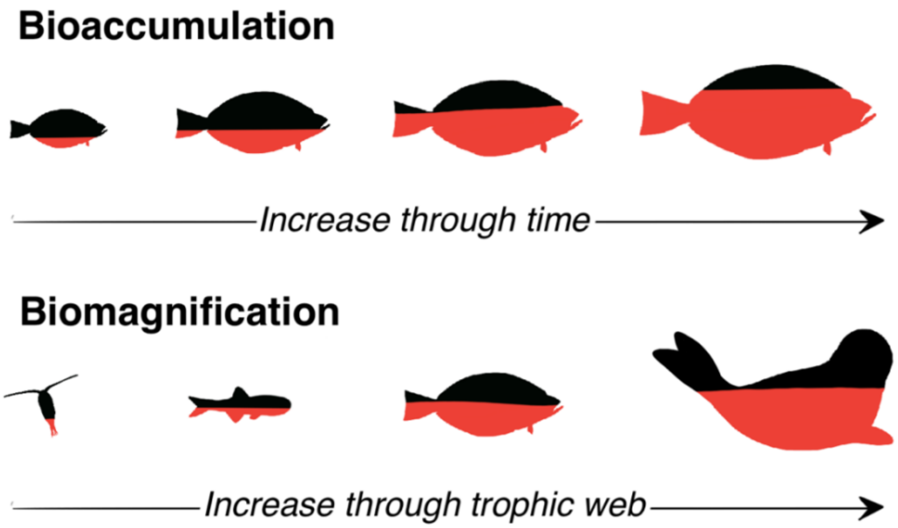


Figure 1. Schematic of bioaccumulation and biomagnification of contaminants increasing in biota through time and across trophic webs. Red represents toxin levels (adapted from Brown 2014).

Studies have reported shifts in tissue contaminant levels of Arctic top predators linked to the increase in abundance of temperate/boreal species (McKinney et al., 2009; Braune et al., 2014a; McKinney et al., 2015). McKinney et al. (2012) found that transient or migratory species in a fjord in the Canadian Arctic (Cumberland Sound) had higher trophic magnification factors (TMFs) than resident species. TMFs represent the average diet-to-consumer transfer of a chemical through the food web. Higher TMFs, calculated from the slope of the regression between the chemical concentration and the trophic level of an aquatic organism, are the result of higher metabolic demands from having to travel larger distances (Figure 2) (McKinney et al., 2012; Borgå et al., 2022). The authors suggested that transient species migrating into the fjord had higher feeding rates, which resulted in more contaminants ingested. Ecosystems that received migratory species were expected to have higher contaminant concentrations because migrating species have higher TMFs (Borgå & Kucklick, 2012). Poleward moving species could have similar tendencies to migratory species with higher rates of Hg bioaccumulation.

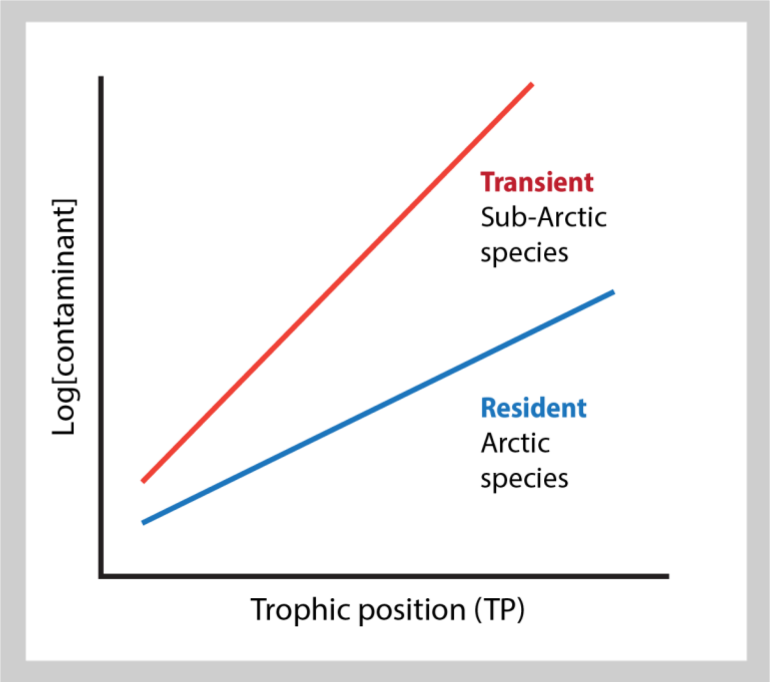


Figure 2. Schematic of the trophic magnification slopes for transient and resident species (adapted from McKinney et al., 2012).

1.4 Mercury and Organochlorine Pesticides

Mercury (Hg) and persistent organic contaminants (POPs) such as organochlorine pesticides (OCPs) reach polar marine ecosystems and accumulate in fish (Borgå et al., 2022; Chételat et al. 2022; McKinney et al., 2022). Fish accumulate organochlorine pesticides (OCPs) and Hg in their lipids and protein-rich tissues, respectively. Top predators like the narwhal (*Monodon monoceros*), beluga whale (*Delphinapterus leucas*), and polar bear (*Ursus maritimus*) are some of the Arctic species most vulnerable to bioaccumulation of mercury and other contaminants because they do not have the physiological capacity to eliminate mercury (Dietz et al., 2013; Meyer et al., 2018).

The use of Hg and OCPs have been reduced under the Minamata Convention and Stockholm Convention, respectively. The Minamata Convention, which entered into force in 2017 as the first global environmental agreement in the 21st millennium, aims to reduce and eliminate the use of mercury. Many mercury-added products have been phased out since 2020, including those in batteries, LED lighting, and cosmetics, while strategies for reducing emissions are being put in place for gold mining, waste incineration, and coal-fire power plants (UN Stockholm, 2019). Many OCPs were banned globally in the 1970s and 1980s. Since then, the Stockholm Convention (an internationally binding agreement adopted in 2004) was formed to further eliminate and restrict OCPs in the environment (UN Stockholm, 2019). However, pesticides like Dichlorodiphenyltrichloroethane (DDT) continue to be used in South America, Africa, and Asia as pest controls to combat malaria.

Mercury is a volatile trace element that exists in three different forms – elemental (Hg^0), inorganic mercuric chloride (Hg^{2+}), and organic methyl mercury (CH_3Hg^+). Hg is primarily introduced into the oceans via riverine input in its dissolved particulate form (Hg^{2+}) and through ocean-atmosphere gas exchange (Hg^0) (Fitzgerald et al., 2007). Sources of elemental and inorganic Hg are the result of both anthropogenic (e.g. residuals from gold mining, fossil fuel combustion, waste incineration) and natural sources (e.g. volcanic eruptions, rock weathering) (UN Environment, 2019) (Figure 3).

While Hg in soil, aquatic sources, and the atmosphere exists in its inorganic form, methyl mercury (MeHg) is bio-transformed by sulphur-reducing anaerobic bacteria found in sediments, wetlands, and the anoxic bottom water and is the major component of dietary Hg in fish and fish-eating wildlife (Bloom, 1992; Van Walleghem et al., 2007; Greenfield & Jahn, 2010). MeHg biomagnifies and bioaccumulates more than elemental and inorganic Hg (Lavoie et al., 2018). MeHg poses the greatest concern to fish and other mid- to upper-trophic level marine organisms. Chronic MeHg exposure in fish can produce birth defects, neurotoxic effects, and reproductive toxicity. Long-term mercury exposure has been associated with altered growth/behaviour and reduced survival of marine fish (Zheng et al., 2019). Hg, which binds to protein-rich tissue (e.g. muscle), can concentrate in piscivorous fish tissue up to 107 times more than in surrounding waters (Wiener et al., 2002). As such, the risk of the methylated form of Hg is especially high in reproducing female seabirds and marine mammals where the neurotoxin can be readily passed to the sensitive developing eggs or embryos and can lead to adverse effects on brain development (Ackerman et al., 2016). Elevated concentrations of MeHg pose a particular threat to humans, fish, and wildlife given its ability to biomagnify across food webs and bioaccumulate in the tissue of long-lived species (Driscoll et al., 2013; Lavoie et al., 2013; Tan et al., 2009). When ingested by humans, MeHg, even under relatively low, chronic exposure, can lead to negative effects on cardiovascular and central nervous systems (Table 1) (UN Environment, 2019). Currently the amount of inorganic Hg removed by the deep ocean is less than the input by the atmosphere and rivers meaning that the deep ocean accumulates Hg (UN Environment, 2019). Emissions remain high for inorganic Hg and in 2015 combustion of coal and artisanal gold mining represented 60% of emissions to the atmosphere (UN Environment, 2019). Long-range transport moves atmospheric emissions to remote regions like the Arctic tundra which has been deemed a globally important sink for mercury (Berg et al., 2003; Ariya et al., 2004; Barkay & Poulain, 2007). In this thesis, Hg will be referred to as total mercury (THg) in the analysis to include its elemental, inorganic, and organic forms.

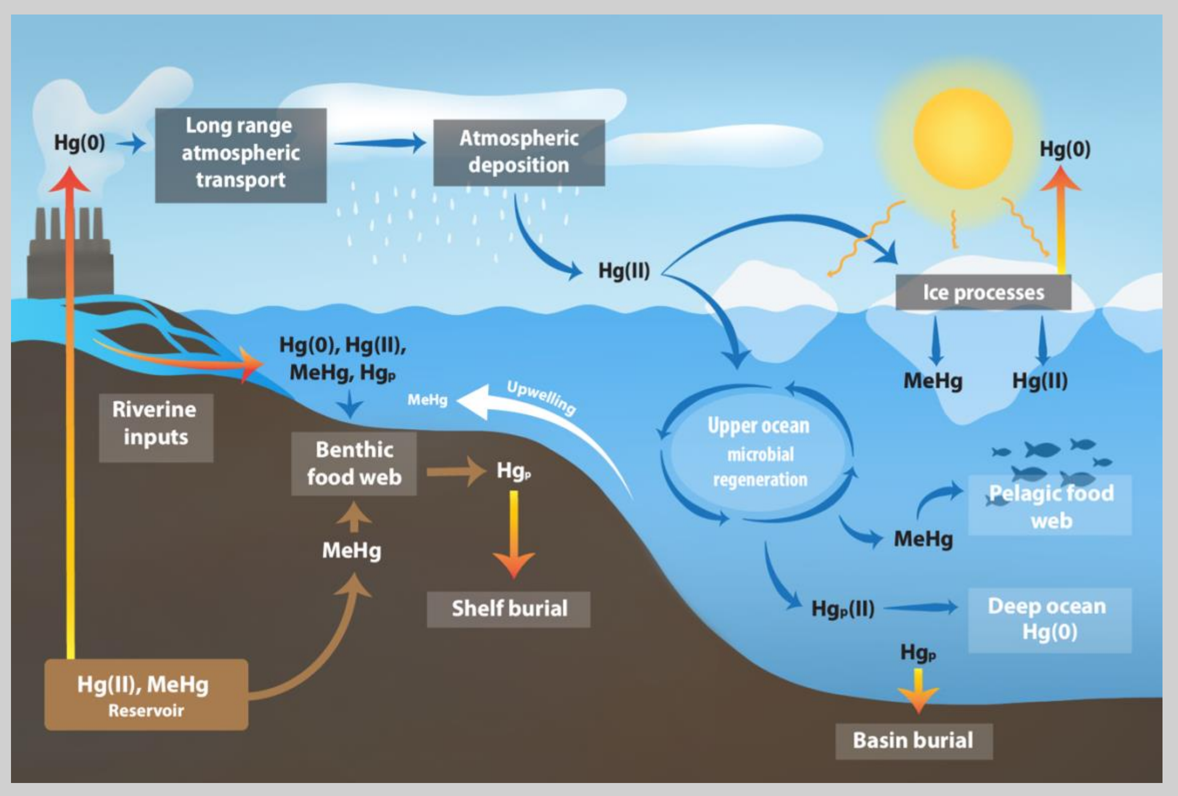


Figure 3. Schematic of mercury cycling in Arctic marine ecosystems (derived from AMAP, 2011).

Organochloride pesticides (OCPs) are synthetic, non-polar, and toxic compounds that are used in agriculture to destroy pests, weeds, and pathogens. They are of concern given their high toxicity and that they persist in soil, water, air, and biota for years after application (Jayaraj et al., 2016). For humans, prolonged exposure OCPs is linked to allergies, hypersensitivity, nervous system damage, reproductive and immune dysfunction, endocrine disruption, and cancer (Table 1) (Jayaraj et al., 2016). Organochlorine compounds are also highly lipophilic and can concentrate in fat-rich food such as meat and milk and be passed onto offspring (e.g. marine mammals) (Hernández et al., 1994). Chronic exposure of fish to sub-lethal concentrations of OCPs can reduce swimming function in the larval stages of fish, inhibit enzymatic activity, and cause growth delays impairing reproduction and survival (Murty, 1986).

Table 1. Glossary of the contaminants detected in >50% of the samples and their effects on humans (US EPA, 2022). The oral reference dose (RfD), which is expressed in units of mg/kg-day, is based on the thresholds that exist for toxic effects (e.g. cellular necrosis) on a daily exposure for human consumption and includes sensitive human sub-groups.

CONTAMINANT SUMMARY AND RISK EXPOSURE

Contaminant	Source	Persistence in the environment	Oral reference dose (RfD)	Human consumption risk
Mercury, elemental	Fossil fuel combustion, waste incineration, volcanic eruptions, rock weathering	Low persistence (half-life: 40 days)	3×10^{-4}	Hand tremor; increases in memory disturbances; slight subjective and objective evidence of autonomic dysfunction
Methyl mercury	Conversion by bacteria, easily taken up by microorganisms because low water solubility	Low persistence (biological half-life: 30-70 days)	1×10^{-4}	Developmental neuropsychological impairment
Dieldrin	Insecticide and impurity from the breakdown product of endrin	High persistence (half-life) (9 months)	5×10^{-5}	Liver lesions
Hexachloro-butadiene	Solvent for other chlorine-containing compounds	Low persistence (half-life: 17 days)	*Oral Slope Factor:* 7.8×10^{-2} per mg/kg-day	Possible human carcinogen
ΣChlorobenzene	Volcanoes, forest fires, crude oil, cigarette smoke, paints, detergents	Low persistence (half-life: 6 days)	4×10^{-3}	Decreased lymphocyte count
ΣChlordane	Insecticide	Low persistence (half-life: 1 year)	5×10^{-4}	Hepatic necrosis
ΣDDT *	Acaricide, insecticide	High persistence (half-life: 2-15 years)	5×10^{-4}	Liver lesions
ΣParlar	Insecticide mixture widely used until the 1980s	High persistence (half-life: 100 days -12 years)	Not found	Undetermined

* Dichlorodiphenyltrichloroethane

1.5 Diversity and species composition of fish in the Arctic

The Canadian Arctic is spatially extensive and has a vast coastline spanning ~173,000 km (Figure 4). The size and oceanographic complexity of the Canadian Arctic is reflected in the complexity of the habitats and the diversity in the fish communities it supports. The most prominent families of fish found across the Canadian Arctic include Zoarchidae (31 species), Salmonidae (17 species), and Cottidae (14 species) (Coad & Reist, 2018). The Canadian Arctic is generally divided into three main regions including the Beaufort Sea, the Canadian Arctic Archipelago, and Baffin Bay. Species diversity of marine fishes tends to increase towards the eastern Canadian Arctic (Baffin Bay) and reflect similar fauna to that in the North Atlantic (Mueter et al., 2013). Increased species diversity in the Baffin Bay-Davis Strait offshore basin could be attributed to the merging of south-flowing Labrador Current and the West Greenland Current which contributes to a highly productive region (Coad & Reist, 2018).

The Beaufort Sea is dominated more by benthic fish (46 species, 11 families) compared to pelagic fish (6 species, 3 families). Arctic cod (a benthopelagic species) was the most abundant fish caught from both benthic and pelagic trawls across the Beaufort Sea and Alaskan waters (Logerwell et al., 2010). The western Canadian Arctic and, specifically, the Amundsen Gulf are thought to be important spawning areas where dense aggregations of adult Arctic cod have been found (Benoit et al., 2008; Geoffroy et al., 2011). In addition, the distinct water masses in the Beaufort Sea (seasonal mixed layer above the Pacific mode water, Atlantic water at the mid-slope layer, and Arctic water in the deep layer) (Figure 5) enables the vertical segregation of Arctic cod and other fish communities (Mueter et al., 2013). Across all months, biomass of small age-0 Arctic cod tended to be higher in the epipelagic (<100m) and biomass of larger age-1+ was highest in bottom areas (<500 m) (Geoffroy et. al 2016; Majewski et al., 2016).

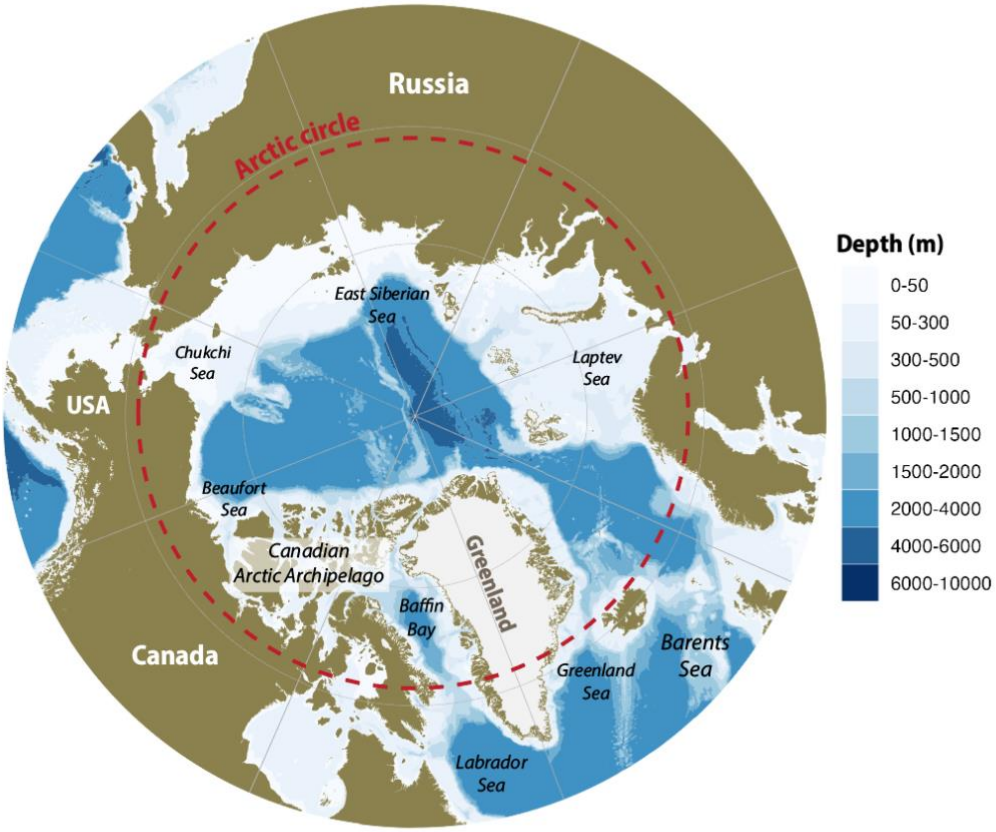


Figure 4. Basins and bathymetry of the circumpolar Arctic.

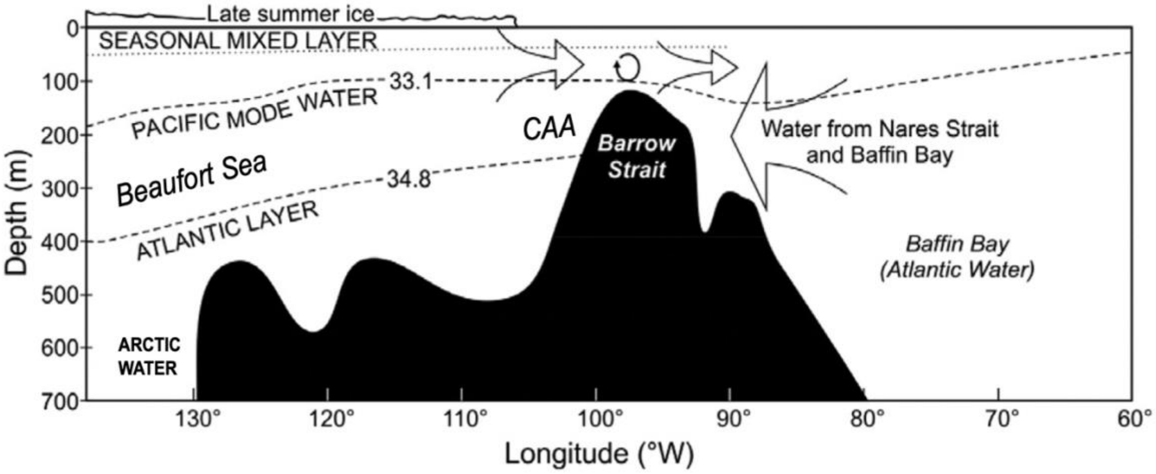


Figure 5. Water masses and bathymetry across the Canadian Arctic Archipelago (Brown et al., 2018).

Fifty one percent of all Arctic marine fish species occupy the Canadian Arctic Archipelago. Four of the main families that account for these species in the Canadian Arctic Archipelago include Zoarcidae (eelpouts), Salmonidae (salmonids), Cottidae (sculpins), and Gadidae (cods) (Mueter et al., 2013). Commercially targeted species in the region include Arctic char (*Salvelinus alpinus)* in the Canadian Arctic Archipelago and Greenland halibut (*Reinhardtius hippoglossoides*) towards the eastern Canadian Arctic. Greenland halibut (or turbot) is the dominant demersal fish caught below 400 m in the eastern margin of the archipelago (i.e. on the west side of Baffin Island) (Bowering & Nedreaas, 1999). Often caught as bycatch in the turbot fishery are the less abundant species: grenadiers (Macrouridae) and skates (Rajidae). The mesopelagic layer (200m – 1000m) in Baffin Bay is dominated by Gadidae (mainly Arctic cod) and juvenile Liparidae (snailfishes) and there is a distinct separation between the mesopelagic communities in Baffin Bay and the Labrador Sea (Chawarski et al., 2022).

The Labrador Sea has four main mesopelagic fish families: Myctophidae (lanternfishes), Stomiidae (dragonfishes), Gonostomatidae (bristlemouths), and Bathylagidae (pencil-smelts) (Chawarski et al., 2022). Glacier lanternfish (*Benthosema glaciale*) of the Myctophidae family dominate the Labrador Sea. Lanternfish are weak swimmers and can be advected into Baffin Bay (Priou et al., submitted), but because of the extreme light regime, alternating from midnight sun to polar night, and lower water temperatures they have low survival rates north of the Labrador Sea (Kaartvedt, 2008; Chawarski et al., 2022). Langbehn et al. (2022) found that in the summer, lanternfish are confined to depths (450m-650m) to avoid visual predation. As a result, they starve in the summer because their prey are shallower in depth. Summer starvation is what reduces survival because it depletes them of their surplus energy needed for reproduction. As such, lanternfish are more abundant below the Arctic circle (i.e. Labrador Sea) where the light regimes are less extreme, and where the light-dependent encounters and temperatures are more favourable (McKelvie, 1985; Chawarski et al., 2022; Langbehn et al., 2022). While the pelagic Labrador Sea is dominated by lanternfish, coastal Labrador is dominated by capelin (*Mallotus villosus*) and sandlance (*Ammodytes* spp) as per Arctic charr diet analyses (Cote et al., 2021). Demersal fish (in waters greater than 500 m) that dominate NAFO fishing zones 2H and 2G (Appendix A) include Sebastidae (deepwater redfish), Pleuronectidae (e.g. Greenland halibut), Moridae (e.g. blue hake [*Antimora rostrata*]), and different species of grenadiers (Cote et al., 2023; Priede et al., 2017).

Species-specific responses related to climate change vary across the Canadian Arctic and subarctic regions. Warming of subarctic and Arctic waters leads to changes in the abundance of pelagic species like Arctic cod and capelin (*Mallotus villosus*). Arctic cod persist in sub-zero Arctic waters, while capelin occur in the subarctic marine environment with some presence in southern Baffin

Bay (Hop & Gjøsæter, 2013). Capelin spawn in the cold waters of the Arctic and subarctic regions and respond to large-scale climate related changes by shifting their distribution northward in response to warming temperatures (Rose, 2005; Carscadden et al., 2013; Ingvaldsen & Gjøsæter, 2013; Ulrich & Tallman, 2021). Capelin respond more quickly to climate warming than Arctic cod because they have faster population growth and a broader thermal range (Rose, 2005). For example, capelin in the Barents Sea have responded to warming waters by a shifting their spawning grounds northward and spawning earlier (Huse & Ellingsen, 2008). The distribution of capelin is influenced by temperature and predator/prey relationships, whereas the distribution of Arctic cod is mainly influenced by temperature (Marsh & Mueter, 2020). Arctic cod are mostly limited to cooler waters as their likelihood of occurrence decreases in southern ranges (Marsh & Mueter, 2020). The Labrador Sea, an Arctic outflow shelf (Carmack & Wassmann, 2006), receives cold water from the Arctic and is a thermal refuge for Arctic cod and acts as a barrier to subarctic fish. In contrast, the Bering Sea, an Arctic inflow shelf (Carmack & Wassmann, 2006), receives warm Pacific water into the Arctic. As a result the Bering Sea experiences accelerated rates of sea surface and bottom temperature warming with a decreased abundance of Arctic cod in the region (Marsh & Mueter, 2020).

Secondary to temperature, competition with boreal species also limits the southward distribution of Arctic fish. Marsh & Mueter (2020) found little evidence of competitive interactions between capelin and Arctic cod in the Labrador Sea. They attribute the co-existence of the two species to the shared predation pressure by mutual predators. When capelin were absent Arctic cod were more regularly preyed upon by species like Atlantic cod (*Gadus morhua*) and Greenland halibut (*Reinhardtius hippoglossoides*). In shallow areas of the Labrador shelf, Greenland halibut abundance were found to be negatively related to Arctic cod where Greenland halibut were smaller and fed on a more pelagic diet (Marsh & Mueter, 2020). After reaching a certain size, Greenland halibut (average length >69 cm) move deeper and switch to a diet dominated by groundfish (Bowering & Lilly, 1992). In those deeper waters, Arctic cod abundance was found to be positively correlated with that of Greenland halibut (Marsh & Mueter, 2020).

Deep sea fish species are often characterized by their slow growth rate, late maturity, low fecundity, and long potential lifespan (Drazen et al., 2012). Their long-lived nature and slow growth rate make them valuable study species for this research because they reflect contaminants that have accumulated in bottom habitats for ~10-50 years (i.e. life span of deep sea fish). Similar to the migrations of pelagic fish, Greenland halibut, especially those in the Northwest Atlantic, are highly migratory (i.e. range from Baffin Bay to the offshore areas of Newfoundland and Labrador [Boje, 2002]) and exhibit benthopelagic feeding strategies (Giraldo et al., 2018). Their long-range migrations, complex life histories, and high lipid content may make them efficient biovectors for contaminants. Abyssal grenadier (*Coryphaenoides armatus*) and blue hake are representative of the deep ocean because they retain site fidelity at depths (Wenner & Musick, 1977). Both species occupy different depth zones in the Labrador Sea but share some overlap between 2000 and 2500m (Murua and Cárdenas, 2005). Abyssal grenadier feed actively on the seafloor and have little upward movement into the water column (Priede et al., 1990), while blue hake have a similar habitat range but live on the continental slope from 400m to 2500m (Wenner & Musick, 1977).

1.6 Human consumption and increased contaminant exposure

Fish and marine mammals (e.g. seals, belugas, narwhals) are an integral to the diet and culture of Indigenous Peoples, specifically those in Inuvialuit (western Canadian Arctic), Nunavut (eastern Canadian Arctic), and Nunatsiavut (Labrador). As such, northern communities have 15 times higher seafood consumption per capita than non-indigenous peoples (AMAP, 2017). For example, Cumberland Sound is a highly productive deep-water basin that supports food supply in Pangnirtung, NU (AMAP, 2017). For communities like Pangnirtung it is not only important to monitor contaminant levels in the directly ingested top predators (i.e. belugas), but also in the prey sources that feed top predators. Increasing our understanding of how ecological variables influence contaminant uptake by demersal and pelagic prey species is needed to better predict how contaminant biomagnification could change with climate warming (UN Environment, 2018).

1.7 Greenland halibut commercial fishing

Indigenous Peoples not only rely on foods harvested from the ocean for sustenance, cultural practices, but also for economic value through their exportation to southern markets (AMAP, 2021). The Greenland halibut fishery is considered one of the most lucrative Atlantic groundfish fisheries. Nunavut fishing enterprises landed on average $90 million per year (2011 and 2017). Regionally, $47 million and $43 million per year in landings originated from the management areas NAFO 0B and 0A (2011 and 2017), respectively (DFO, 2022). NAFO 0A is entirely reserved for Nunavut fisheries (e.g. Baffin Fisheries), while NAFO 0B is shared between Nunavut, Nunavik, Labrador, Newfoundland, and Nova Scotia. In addition, Greenland halibut stocks are transboundary meaning that NAFO Subarea 0 is managed by Canada and Subarea 1 is managed by Denmark (DFO, 2022).

1.8 Research questions

This study aims to quantify and compare THg and OCPs concentrations in six Arctic and subarctic fish species sampled in the Labrador Sea and Canadian Arctic. The objective of this work is to examine the spatial and ecological variability in contaminants for different species with differing dietary niches and habitats. Few studies have focused on demersal fish in the Labrador Sea and little is known about contaminant accumulation in oceanic species like abyssal grenadier and glacier lanternfish. I used pelagic fish (e.g. capelin and glacier lanternfish) to assess bioaccumulation in the water column, demersal fish to assess bioaccumulation in benthic habitats (e.g. abyssal grenadier, blue hake), and benthopelagic fish (e.g. Greenland halibut and Arctic cod) to contrast between the two. Differences of THg in the eastern and western Canadian Arctic were quantified using Arctic cod samples from the Beaufort Sea and Baffin Bay. Comparisons of capelin, deemed the 'replacement species' of Arctic cod (Ulrich & Tallman, 2021), and Arctic cod were conducted to determine whether the northward expansion of capelin (Carscadden et al., 2013) would increase contaminant concentrations in Arctic ecosystems. Glacier lanternfish, occupying a similar trophic level as capelin and Arctic cod, represent the most abundant pelagic fish in the Labrador Sea. Lanternfish tissue was analysed to assess whether the proximity to anthropogenic activities (i.e. more activities in the Labrador Sea than in the rest of the Arctic) influence contaminant concentrations in pelagic fish communities. Previous studies have compared the contaminant concentrations between Arctic and subarctic species (Braune et al., 2014b,c; Pedro et al., 2017; Pedro et al., 2019), but my study covers a much larger geographic range than what was previously covered. This study is also among the first to explicitly compare contaminant concentrations both among and between demersal and pelagic fish. Here I compared their pollutant concentrations with similar sized deep sea fishes that have not been well studied (e.g. abyssal grenadier and blue hake) and who are exposed primarily to THg through sinking particulate and natural geogenic sources (AMAP, 2021). I also studied Greenland halibut, which occupies wide latitudinal and depth ranges and provides an example of a benthopelagic species with a highly migratory behaviour.

Climate change is resulting in a shift from benthic to more pelagic carbon sources (Burton & Koch, 1999; Zhulay et al., 2023). To determine if a shift in benthic to pelagic food webs would shift contaminant biomagnification in fish, we rely on stable isotopes. Nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) are useful dietary markers to describe the ecological changes related to trophic web structure and feeding ecology (Szpak & Buckley, 2020). Because most organisms excrete lighter nitrogen ($\delta\ ^{14}N$) and retain heavier isotopes ($\delta\ ^{15}N$), we can use the increasing ratio $^{15}N/^{14}N$ to reflect this amplification through the food web (Cabana & Rasmussen, 1994; Lavoie et al., 2018; Knickle and Rose 2014). Carbon sources, a proxy for food source (DeNiro & Epstein 1978), are calculated using $\delta^{13}C$ isotopes. For example, a higher $\delta^{13}C$ represents a more benthic feeding strategy. We can use both $\delta^{13}C$ and $\delta^{15}N$ to trace the source of productivity that is fueling food webs (e.g. habitat, taxa, resource type) and find the origin of the individual (Finlay & Kendall, 2007). Considering existing contaminant levels in subarctic species (Dietz et al., 2021; Pedro et al., 2017), their northward migration (Fossheim et al., 2015), and our understanding that fish species could act as biological vectors transporting contaminants into the Arctic (Blais et al., 2005; Krümmel et al., 2003; McKinney et al., 2012), we can make inferences about future contaminant burdens to piscivorous Arctic predators. This project furthers our understanding of how climate-related ecological shifts could influence contaminant bioaccumulation in individuals. It also highlights prey and predator species that might be of concern for human consumption and documents mercury and pesticide concentrations for poorly studied fish - particularly glacier lanternfish, abyssal grenadier, and blue hake.

1.9 Study area

The study area is comprised of the Labrador Sea and three main regions of the Canadian Arctic (Baffin Bay, Canadian Arctic Archipelago, and the Beaufort Sea) (Figure 6).

The Canadian Arctic is known for its complexity with numerous narrow channels and interconnected basins separated by glacial-formed sills (Michel et al., 2006). This complex topography dictates water mass movement throughout the basins and influences biota (Carmack, 2000; Melling, 2000). In addition to the decline of sea-ice, the Canadian Arctic is experiencing major changes in water mass movement including a warming of the Atlantic layer (Carmack et al., 1995) and a shift in the boundary between Atlantic and Pacific water masses (McLaughlin et al., 1996). The Atlantic/Pacific front is moved eastward by a cyclonic circulation mode, which implies more export of Pacific water through the Canadian Arctic Archipelagic (McLaughlin et al., 2002). THg is input into the Arctic Ocean every year through atmospheric deposition (~100t yr^{-1}), advection from the Pacific and Atlantic currents in the dissolved form (~48t yr^{-1}), and riverine inputs and coastal erosion in its particulate form (~60t yr^{-1}) (Fisher et al., 2012; Dastoor & Dunford, 2014; Shang et al., 2015). The western Canadian Arctic (including the Beaufort and the Chukchi Sea) supplies almost all the water into the archipelago and, therefore, acts as a reference point for contaminants entering the Canadian Arctic Archipelago (Brown et al., 2018). Surface waters within the Canadian Arctic have a residence time of ~10 years which means that the last few decades are most important to consider in assessing contaminant accumulations (Brown et al., 2018). However, with climate-induced melting of glaciers, increased riverine runoff, and thawing of permafrost, legacy pollutants and THg that were used in the 1950s and 60s are becoming more available to Arctic biota (Brown et al., 2018). Because there are more glaciers and riverine inputs in the Canadian Arctic compared to the rest of the Arctic Ocean, THg would become more bioavailable in pelagic ecosystems as melting occurs.

The Labrador Sea is connected to the Canadian Arctic by the Labrador Current which extends cold water along the coast of Newfoundland and Labrador and leads to cooler water nearshore and warmer water offshore (Hunt et al., 2016). These colder and lower-salinity waters from the Canadian Arctic are THg-enriched (Cossa et al., 2018). There are several banks and channels throughout the Labrador Shelf that add to the bathymetric complexity of the basin. There is also a cold intermediate layer (<0°C; 100-200m in thickness) that extends 200-300 km offshore in the summer and varies in size seasonally (Petrie et al., 1988). As an Arctic outflow region (Carmack & Wassmann, 2006), polar species like Arctic cod are limited to these cold-water masses in the Labrador Sea (Wyllie-Echeverria & Wooster, 1998). The Labrador Sea is one of the only regions in the world that is dominated by deep water convection thereby delivering oxygenated waters to the deep sea and providing a distinct link between the surface and the deep ocean (Cote et al., 2023). However, with climate change deep sea convection in the Labrador Sea is weakening (Yang et al., 2016).

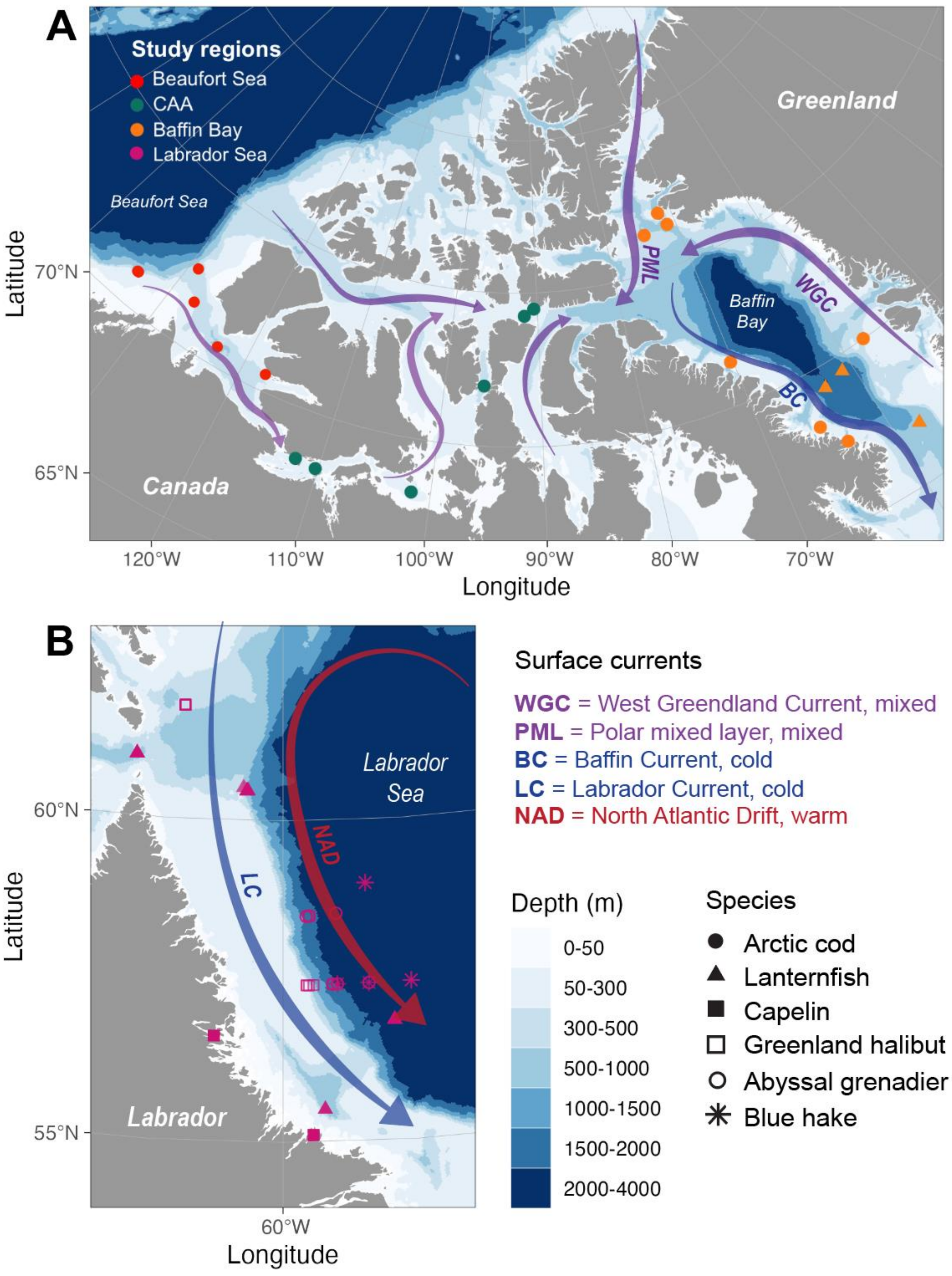


Figure 6. Sampling sites from 2016-2021 with symbols representing the species that were sampled in the (A) the Canadian Arctic and (B) Labrador Sea. Surface currents modified after Rochon et al. (2006) and Sicre et al. (2014).

Chapter 2 – Contaminants in fish across habitats, feeding ecology, and species in the Canadian Arctic and the Labrador Sea

2.1 Introduction

Mercury (Hg) and organochlorine pesticides (OCPs) are a growing concern in Arctic food webs given their high persistence in the environment and their deleterious effects on fish and other wildlife (McKinney et al, 2022). Physical changes brought by climate change will likely further affect the biogeochemistry and cycling of Hg in Arctic ecosystems (Chételat et al., 2022; Stern et al., 2012). Hg in its elemental (Hg^0) and inorganic form (Hg^{2+}) are the result of both anthropogenic activities (e.g. residuals from gold mining, fossil fuel combustion, waste incineration) and natural phenomena (e.g. volcanic eruptions, rock weathering) (AMAP, 2021). The organic form of Hg, methyl mercury (MeHg), is converted by bacteria in oxygen-poor environments (e.g. family Desulfobacteriaceae) (Bloom, 1992; King et al., 2000; Van Walleghem et al., 2007; Greenfield & Jahn, 2010) and is the most toxic form of Hg with the highest level of biomagnification in trophic food webs. Inorganic mercury flows into the ocean through fluvial sources, is converted by the bacteria in the water, and then incorporated into pelagic food webs (WHO, 1990). Mercury exists in low concentrations (<0.01 µg/g) in terrestrial organisms, where most of the mercury is the inorganic, less toxic form (Lavoie et al., 2018). However, marine biota such as fish and shellfish have higher mercury concentrations (<0.04 µg/g), of which 90% is the highly toxic organic methylated form (WHO,1990; Grandjean et al., 2003). Chronic exposure of mercury results in adverse effects on growth, reproduction, and survival of marine fishes (Zheng et al., 2019), as well as adverse effects on humans (Table 1). Hg will hereafter be referred to as total mercury (THg) to include its elemental, inorganic, and organic forms even though the organic form (MeHg) is the main form of mercury in fish.

OCPs, which are characterized as persistent organic pollutants (POPs), were used extensively in agriculture, forestry, and for domestic pest control (Jayaraj et al., 2016). In the 1970s and 1980s many OCPs were restricted or entirely banned in most countries except for South America, Africa, and Asia (Rogan & Cheng, 2005; Thompson et al., 2017; Guo et al., 2019) After discovering these persistent pollutants in remote regions of the Arctic thousands of miles from known sources, the internationally binding Stockholm Convention (adopted in 2001 and put into force in 2004) was put in place to either restrict or entirely ban harmful OCPs like dichlorodiphenyltrichloroethane (DDT). Although pollutants generally appear to be below acute toxicity thresholds, OCP concentrations are still detected in Arctic marine wildlife (Rigét et al., 2007; Letcher et al., 2010; Dietz et al., 2013).

Species in Arctic and subarctic regions shift their distributions as climate changes (Fossheim et al., 2015; McKinney et al., 2022). Arctic waters experience warming at an accelerated rate because of sea ice loss (Pistone et al., 2014; Screen & Simmonds, 2010). Because of amplified changes in the Arctic, the replacement rate of resident species with poleward moving species is five times higher than the global average (Cheung et al., 2009). Fish in Arctic regions face a higher risk of displacement as boreal fish species move northward into their habitat. For example, northward range expansion has been observed in subarctic capelin (*Mallotus villosus*), harp seals (*Pagophilus groenlandicus*), and killer whales (*Orcinus orca*) (Ferguson & Higdon, 2010; Kovacs et al., 2011; Higdon et al., 2012; Carscadden et al., 2013). Arctic species (e.g. polar bears [*Ursus maritimus*]) have shown changes in feeding ecology, reproductive success, and habitat use as a result of boreal species expansion (Stirling et al., 1999; Stirling, 2005; Stirling & Parkinson, 2009; McKinney et al., 2009, 2013). Species moving northward can also act as vectors and transport contaminants into Arctic food webs. During earlier summer ice breakup, polar bears fed more on subarctic species (e.g. harbour seals), which resulted in a faster accumulation of new POPs (McKinney et al., 2009).

Understanding how contaminant concentrations in marine fish vary as a function of feeding ecology and habitat at high latitudes can provide insight into how climate and ecosystems interact to influence contaminant levels in the Arctic (McKinney et al, 2022). These interactions have been well studied in the Canadian Arctic for marine mammals and seabirds (McKinney et al., 2015; Brarune et al., 2014), yet less is known about pelagic and demersal fish communities. In this study, I identify important ecological and spatial factors explaining variability of THg and OCP concentrations among Arctic and subarctic fish.I hypothesize that demersal fish have higher contaminants concentrations than pelagic fish and subarctic fish have higher concentrations compared to Arctic fish, therefore indicating that THg and OCP variation is explained by both spatial and ecological factors.

2.2 Materials and Methods

2.2.1 Study area and design

Fish were collected in the Labrador Sea, Baffin Bay, Canadian Arctic Archipelago, and the Beaufort Sea (Figure 6). A total of 620 fish were collected between 2017 and 2021 (Table 2). Samples of muscle tissue were removed from pelagic fish (capelin, glacier lanternfish [*Benthosema glaciale*], and Arctic cod [*Boreogadus saida*]) and demersal fish (abyssal grenadier [*Coryphaenoides armatus*], blue hake [*Antimora rostrata*], and Greenland halibut [*Reinhardtius* *hippoglossoides*]) across the Labrador Sea (53°N - 66°N; 64°W- 49°W) and Canadian Arctic (66°N – 79°N; 144°W - 53°W) (Table 2). Glacier lanternfish and Arctic cod were collected from the CCGS *Amundsen* using a modified Isaac Kid Midwater Trawl (IKMT, 13.5m^2 rectangular mouth aperture with mesh sizes of 11 mm in the first section and 5 mm in the last section) and bottom beam trawl (3 m^2 rectangular aperture with a headline and a footrope of 4.27 m and a cod-end mesh of 9.5 mm). The IKMT was towed in the mesopelagic sound scattering layer for 20 mins at 3 knots (between 200 m and 1000 m). The bottom beam trawl was deployed for 20 min at 1.5 knots (100 – 200 m). Capelin were collected by community partners in Makkovik and Nain, in Labrador, using a rod and reel and hand-picked from the beach. All demersal fish were collected using longlines. In 2 21, Greenland halibut were collected by Baffin Fisheries in NAFO Subarea 0B (Appendix A) using the same methods. Immediately following their retrieval, all fish from the IKMT, beam trawl, longline, and bottom trawler were identified and placed in storage at -20ºC until muscle extraction.

Table 2. Summary of the samples collected from across the Canadian Arctic and Labrador Sea in the summer (May-September) from 2017 to 2021. Standard length and depth are represented with an average value, the standard deviation (for length), and ranges (for depth). Missing data denoted as *n.d.* (data not available).

	Demersal				**Pelagic**					
	Abyssal grenadier *C. armatus*	Blue hake *A. rostrata*	Greenland halibut *R.hippoglossoides*		Capelin *M.villosus*	Glacier lanternfish *B.glaciale*		Arctic cod *B.saida*		
	Labrador Sea	Labrador Sea	Labrador Sea	Baffin Bay*	Labrador Sea	Labrador Sea	Baffin Bay	Beaufort Sea	CAA	Baffin Bay
Number of fish (f)	6	12	6	10	59	110	122	130	68	97
Total pools (n)	6	12	6	10	20	12	10	12	7	11
Standard length (mm)	208 ± 53 (160-300)	497 ± 53 (430-590)	728 ± 137 (500-800)	313 ± 10 (296–323)	125 ± 15 (105-151)	50 ± 5 (37-64)	55 ± 6 (42-85)	115 ± 25 (52-172)	108 ± 25 (59-182)	103 ± 25 (55-160)
Depth (m)	2756 (2001–3015)	2084 (1738-2504)	1276 (600-1969)	n.d. (1500-3000)	surface	532 (100-834)	484 (400-500)	401 (297-630)	261 (111-366)	329 (90-562)
Sample year	2017	2017	2017	2021	2018, 2021	2019, 2020, 2021	2019, 2020, 2021	2021, 2019	2021, 2019	2021, 2019
Sampling type	Longline	Longline	Longline	Bottom trawler	Rod and line, hand-picking	Mid-water trawl	Mid-water trawl	Beam trawl	Mid-water trawl	Mid-water trawl

*depth estimated by Baffin Fisheries

** CAA is the Canadian Arctic Archipelago

2.2.2 Sample preparation and homogenization

In the lab, dorsal muscle tissue samples (10-20g) were removed from each demersal fish, packaged in 80% acetone-rinsed aluminium foil, and sealed in whirlpacks for contaminant analysis. Pelagic muscle samples were removed following the same extraction and storage technique. Because pelagic fish were smaller, I needed to pool the muscle tissues of several individuals to meet the minimum tissue requirement of 10g. Mercury and OCP concentrations in fish can be influenced by size or age (Storelli et al., 2007). To control for this confounding factor, I selected individuals of the same species and origin with similar morphometrics. All available muscle tissue was removed to create individual species pools which consisted of the following: 3-4 fish for capelin; 6-10 fish for glacier lanternfish; 8-10 fish for Arctic cod. Accordingly, sample sizes (n) for pelagic fishes are pools, while sample sizes for demersal fishes are individuals. For all samples, scales and skin were removed from the muscle and the remaining tissue was homogenized into a paste using acetone-cleaned scissors. Each sample was sub-sampled for their respective analyses (THg: 200mg, MeHg: 1000mg, stable isotopes: 1000mg) and stored at -20ºC. Percent moisture (drying for 48hrs at 70ºC) and lipid content were determined gravimetrically for each pooled sample.

2.2.3 Total mercury analysis

Total mercury concentration in each sample was quantified at the Marine Mammal Ecotoxicology Lab at the Pacific Science Enterprise Center (PSEC) in West Vancouver, B.C. using a Milestone Direct Hg Analyzer following U.S. EPA method 7473. Approximately 0.1g of wet dorsal muscle tissue was analyzed for each sample. Results are expressed at mg/kg wet weight (ww). The method detection limit (IDL) was 0.01ng. Blanks (empty sample boat) were checked at every new sample batch or at one per 20 samples (5%) to confirm the absence of contamination. Blanks less than

10% of the lowest sample concentration were considered acceptable (see point 9.4 of U.S. EPA method 7473 (2007). Every fifth sample (20%) was duplicated to verify the stability and performance of the direct mercury analyser. Any time the difference between duplicates was greater than 10%, the instrument was recalibrated. Matrix spikes, measured at the same frequency of the duplicates and blanks, were checked by spiking samples with a known THg concentration from standard solutions and measuring the recovery of the analyte in the sample. Twenty percent of the spiked precision for both percent recovery and relative percent difference were considered acceptable (see point 3.5 of US EPA Method 7473 [2007]). Standard reference materials (SRMs) used were SRM 2976, DORM-4, and DOLT-5 from the National Research Council of Canada. Percent recovery values for the three SRMS ranged between 96.75-101.74%. SRMs were checked every ten samples to ensure the stability of the instrument calibration where standards were within ±10% of their true value.

2.2.4 Methyl mercury analysis

Methyl mercury concentrations were measured at ALS Environmental in Burnaby, BC following procedures outlined by Liang et al. (1994) and US EPA method 1630 (1998). Homogenized samples were digested in a methanolic potassium hydroxide solution where an aliquot of the digestate was further analyzed by aqueous phase ethylation, purge and trap, desorption, and gas chromatography separation. The separated species were first pyrolyzed to elemental Hg and then quantified by cold vapor atomic fluorescence spectroscopy. Duplicates were run at random within each sampling batch to assess method precision and sample heterogeneity where the acceptable limits were < 9% for MeHg concentration. Method blanks were run for every sample batch where the acceptable limit was less than the MeHg detection limit. Percent recovery values for the three SRMS ranged between 70-130%. All the results were expressed on a wet weight basis.

2.2.5 Organochlorine pesticide analysis

Concentrations of 46 OCP analytes (Appendix B) were analyzed at ALS Environmental in Burlington, ON. Samples were extracted by Soxhlet, prepared by gel-permeation chromatography followed by column chromatography, and analyzed by high resolution gas chromatography and high-resolution mass spectrometry (HRGC/HRMS) following US EPA method 1699 (2007) using isotope dilution and internal standard quantitation techniques. Each batch of samples included a procedural blank, a replicate, and standard reference material (± 2 standard deviations before deemed as acceptable). Where chemicals were detected below the limit, values were replaced with zeroes. OCP values are expressed on a wet weight (ww) basis and were lipid corrected for the analyses using the following equation:

$$Lipid\ normalized\ pesticide = \left[\frac{ng}{g}ww\right] / \left(\frac{\%lipid}{100}\right) \qquad (1)$$

Chemicals that were detected in < 50% of the samples were not included in the calculation. Hereafter, ΣChlorobenzene refers to the sum of 1,2,4,5-Tetrachlorobenzene, 1,2,3,4 – Tetrachlorobenzene, 3,4,5,6-Tetrachloroveratrole, pentachlorobenzene, and hexachlorobezene.. ΣChlordane refers to the sum of heptachlor epoxide B, heptachlor epoxide A, oxychlordane, trans-chlordane, cis-chlordane, trans-nonachlor, and cis-nonachlor. ΣDDT refers to the sum of 2,4’-DDE, 4,4’-DDE, 2,4’-DDD, 4,4’-DDD, 2,4’-DDT, 4,4’-DDMU, and 4,4’-DDT. ΣParlar refers to the sum of parlar 26, parlar 50, and parlar 62.

2.2.6 Stable isotope analysis

Stable isotopes are enriched through the food web (e.g. $\delta^{15}N$) and through carbon sources (e.g. benthic versus pelagic) (e.g. $\delta^{13}C$) which together are used to infer the feeding ecology and habitat of an individual (Choy et al., 2009; Borgå et al., 2022). Stable isotopes were quantified using methods outlined in Yurkowski et al. (2017). Tissues were freeze dried (48hrs at -48°C), ground into a fine powder, and weighed into tin capsules (400 - 600µg). The $\delta^{13}C$ and $\delta^{15}N$ values were measured using a Delta V Advantage Thermoscientific Continuous Flow Mass Spectrometer (Thermo Scientific, Bremen, Germany) coupled to a 4010 Elemental Combustion System (Costech Instruments, Valencia, CA, USA) in the Chemical Tracer Lab at GLIER (University of Windsor, Windsor, ON). Stable isotope ratios are expressed in per mil (‰) in delta notation (δ) described in the following equation: $\delta X = [R_{\text{sample}}/R_{\text{standard}}) - 1] \times 1000$ where X is ^{13}C or ^{15}N and R equals $^{13}C/^{12}C$ or $^{15}N/^{14}N$. The standard materials (R_{standard}) are used were Vienna PeeDee Belemite (PDB) and atmospheric nitrogen (N_2), respectively. The analytical precision of 0.2‰ was measured for both $\delta^{13}C$ and $\delta^{15}N$ in NIST 1577c, tilapia muscle, USGS40 and urea internal standards.

2.2.7 Statistical analysis

To evaluate relationships between contaminant concentrations and ecological factors (e.g. carbon source, trophic level, habitat), I used the non-parametric Spearman correlation matrix (R package *stats*) (Appendix C). I used Shapiro-Wilk to test for normality and Fligner-Kileen to check for homogenity of variances prior to analyses. Length was excluded from the model because length and $\delta^{15}N$ were highly correlated. I used a linear model with fixed effects: trophic feeding (nitrogen), carbon source (carbon), species, and area (equation 2) and log transformed THg to meet normality requirements.

$$\log(THg) = \beta_0 + \beta_1(Nitrogen) + \beta_2(Carbon) + \beta_3(Species) + \beta_4(Location) + \varepsilon \qquad (2)$$

The Akaike information criterion for small sizes (AICc) was used to determine the best fit model with the lowest AICc (R package *AICcmodgav* [Mazerolle, 2020]). Further model assumptions (absence of outliers in residuals, heteroscedasticity, and homogeneity of variance of in the residuals) were confirmed visually using diagnostic plots. All plots were generated using ggplot2 and *ggOceanmaps* in R (Vihtakari, 2022). Statistical analyses were performed in RStudio 2021.9.0.351.

2.3 Results

2.3.1 Dietary variations among demersal and pelagic fish species

Each fish species occupied its own dietary niche, although with partial overlap (Table 3; Figure 7). Arctic cod in the Beaufort Sea and Baffin Bay were most separated compared to other species/locations, with lower trophic feeding in the latter. I observed three distinct trophic niches for pelagic, benthopelagic, and benthic species. Most pelagic species were categorized by lower $\delta^{13}C$ and $\delta^{15}N$ values. Glacier lanternfish had the most distinct dietary niche compared to all other species reflecting its lower trophic-feeding, pelagic life history. Capelin had most overlap with Arctic cod, but Arctic cod had most overlap with Greenland halibut. This reflects the benthopelagic nature of both Arctic cod and Greenland halibut. Blue hake and abyssal grenadier shared the most overlap in their dietary niches of all species reflecting their true benthic feeding ecology.

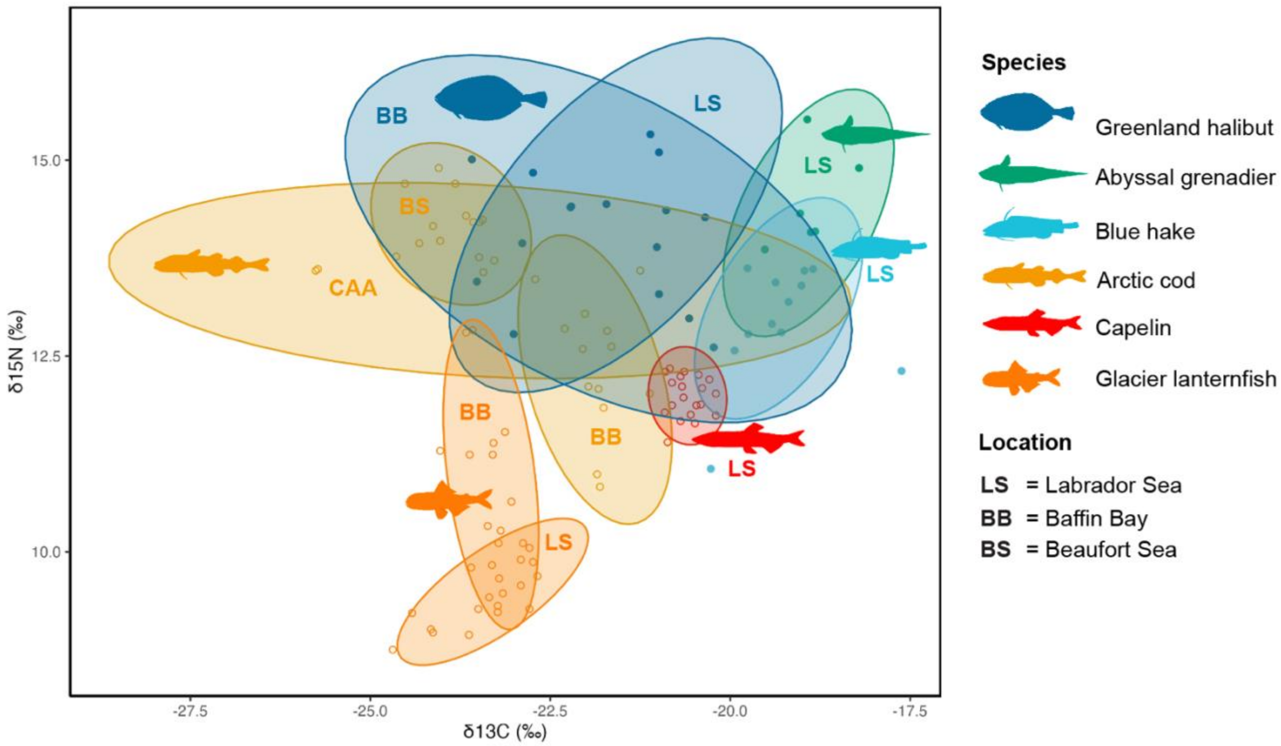


Figure 7. Trophic niches of benthic and pelagic species defined by $\delta^{15}N$ and $\delta^{13}C$ sampled in the Canadian Arctic and Labrador Sea. Higher carbon reflects more benthic feeding and higher nitrogen isotope reflects a species of a higher trophic level. Each species/location combination occupies a distinct dietary niche, but shares overlap with species having similar life histories. Ellipses represent 95% confidence intervals.

2.3.2 Variation in mercury across species, regions, and feeding behaviour

Because THg and MeHg were significantly correlated (R=0.98; Appendix D) and since most of the mercury in fish is MeHg (WHO,1990; Grandjean et al., 2003), I continued with THg in the analyses. Interspecific THg variation (across all species and regions) was best explained by species (type II ANOVA, p-value<0.001) followed by trophic level ($\delta^{15}N$; type II ANOVA, p-value<0.001), area (type II ANOVA, p-value = 0.02), and carbon source ($\delta^{13}C$; type II ANOVA, p-value = 0.04) (Table 4). Even though carbon source ($\delta^{13}C$) significantly impacted THg, the similarity of the top two models suggests that carbon source might be less important than the other variables (Table 4) (Burnham and Anderson, 2002). In addition, one Greenland halibut (0.52 mg/kg) and two abyssal grenadiers (1.82 mg/kg and 0.65 mg/kg) reached concentrations above safe consumption thresholds (above 0.5 mg/kg) set by Health Canada based on the daily human consumption regulations (Health Canada, 2016). I confirmed with diagnostic plots that high THg values were because they were larger fish (Greenland halibut: >800mm, p-value<0.001; abyssal grenadier: 200-300mm, p-value = 0.026) (Figure 8 and 9). The significant difference observed between pelagic and demersal fish could be explained by species' trophic magnification slopes which vary with $\delta^{15}N$ (Figure 10).

Total mercury concentration varied between the Canadian Arctic, Labrador Sea, and Baffin Bay. Across the Canadian Arctic, Arctic cod from the Beaufort Sea had highest mercury concentrations compared to Baffin Bay (Dunn's test, p-value<0.001) and the Canadian Arctic Archipelago (Dunn's test, p-value=0.25) (Figure 11). Comparing the Arctic to the subarctic, the smaller Greenland halibut had lower mercury concentrations in Baffin Bay compared to the larger Greenland halibut in the Labrador Sea (Kruskal-Wallis, p-value<0.001) (Table 3). The opposite was found for pelagic fish where Arctic cod had higher mercury concentrations than capelin (Dunn's test; p-value = $2.82e^{-6}$) (Figure 12). However, capelin had lower on average $\delta^{15}N$ compared to Arctic cod which could explain the higher mercury in Arctic cod.

Table 3. Arithmetic means (mg/kg ww) of methyl mercury, total mercury concentrations (mg/kg ww), and stable isotopes ratios. Sample size (n) represents individual fish for demersal fish and fish pools for pelagic fish. The range is reported in parentheses.

	Demersal				**Pelagic**					
	Abyssal grenadier	Blue hake	Greenland halibut		Capelin	Glacier lanternfish		Arctic cod		
	Labrador Sea	Labrador Sea	Labrador Sea	Baffin Bay	Labrador Sea	Baffin Bay	Labrador Sea	Beaufort Sea	CAA	Baffin Bay
	n = 5	***n = 8***	***n = 6***	***n = 2***	***n = 16***	***n = 8***	***n = 13***	***n = 4***	***n = 2***	***n = 8***
MeHg	419.6 (161-1180)	243.5 (127-351)	278.33 (107-445)	77.35 (76.5-78.2)	41.15 (16.4-73.9)	28.88 (24.2-33.3)	21.75 (15.9-31.5)	49.75 (36-79.2)	27.35 (18.5-36.2)	43.79 (15.4-79.4)
	n = 6	***n = 12***	***n = 6***	***n = 10***	***n = 20***	***n = 10***	***n = 20***	***n = 12***	***n = 7***	***n = 11***
THg	0.6 (0.17-1.82)	0.29 (0.21-0.37)	0.32 (0.13-0.52)	0.11 (0.07-0.15)	0.02 (0.01-0.03)	0.04 (0.02-0.05)	0.03 (0.02-0.04)	0.05 (0.03-0.07)	0.04 (0.01-0.10)	0.03 (0.02-0.06)
	n = 6	***n = 12***	***n = 6***	***n = 10***	***n = 20***	***n = 10***	***n = 20***	***n = 12***	***n = 7***	***n = 11***
δ^{13}C	-18.9 (-19.5- -18.2)	-19.2 (-20.3- -17.6)	-21.7 (-23.0- -20.9)	-21.8 (-23.6- -20.2)	-20.6 (-20.9- -20.2)	-23.4 (-24.0- -23.0)	-23.0 (-23.4- -22.7)	-23.9 (-24.6- -23.3)	-23.5 (-25.8- -21.3)	-22.0 (-23.5- -21.1)
δ^{15}N	14.5 (13.9-15.5)	12.9 (11.1-13.6)	14.2 (12.8-15.3)	14.0 (12.6-15.1)	12.0 (11.4 – 12.3)	11.1 (10.1-12.8)	9.5 (8.8-10.1)	14.2 (13.6-14.9)	13.5 (12.8-14.2)	12.2 (10.8-13.8)

* CAA is the Canadian Arctic Archipelago

Table 4. Akaike's Information Criterion (AIC) with a small sample size results from the linear model. Stepwise model selection favoured the full model ($\delta^{15}N$, $\delta^{13}C$, species, and area) where K is the number of parameters in the model, AICc is the information score of the model, ΔAICc is the difference between the best model and the model being compared, AICcWt is the proportion of the total amount of predictive power provided by the full set of models contained in the model being assessed, and CumWt is the sum of the AICc weights, and LL (loglikelihood) is a measure of how likely it is to see the observed data given the model.

	Predictors	**K**	**AICc**	**ΔAICc**	**AICcWt**	**CumWt**	**LL**
THg	N + C + A + S	12	130.90	0.00	0.74	0.67	-51.75
	N + A + S	11	133.01	2.11	0.26	1.00	-54.08
	A + S	10	145.01	14.11	0.00	1.00	-61.33
	C + A + S	11	145.16	14.26	0.00	1.00	-60.16
	S	7	147.60	16.71	0.00	1.00	-66.22

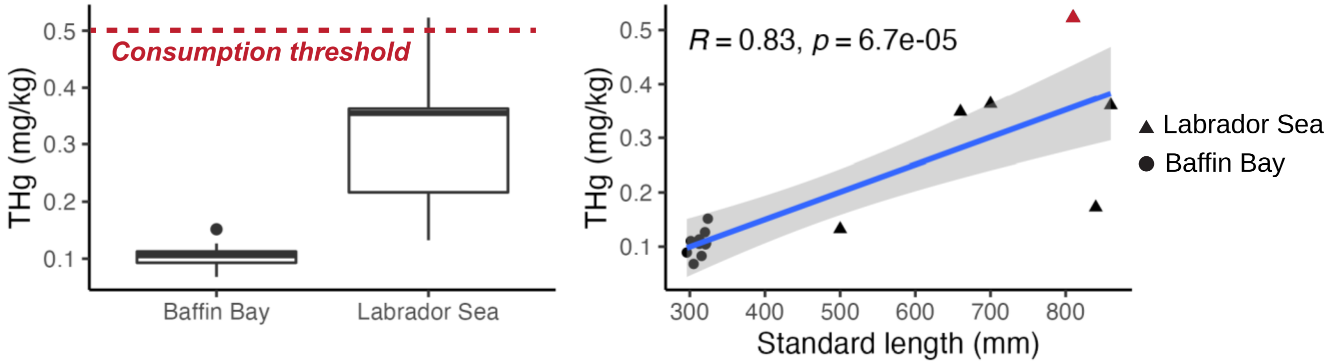


Figure 8. Relationship of length and THg (mg/kg ww) for Greenland halibut in the Labrador Sea and Baffin Bay. (A) Boxplot showing higher THg Greenland halibut in the Labrador Sea and Baffin Bay, and (B) linear regression showing larger fish caught in the Labrador Sea and smaller fish caught in Baffin Bay. The largest Greenland halibut (>800mm) was above the fish toxicity threshold for human consumption located in the Labrador Sea (0.5 mg/kg ww) (red triangle).

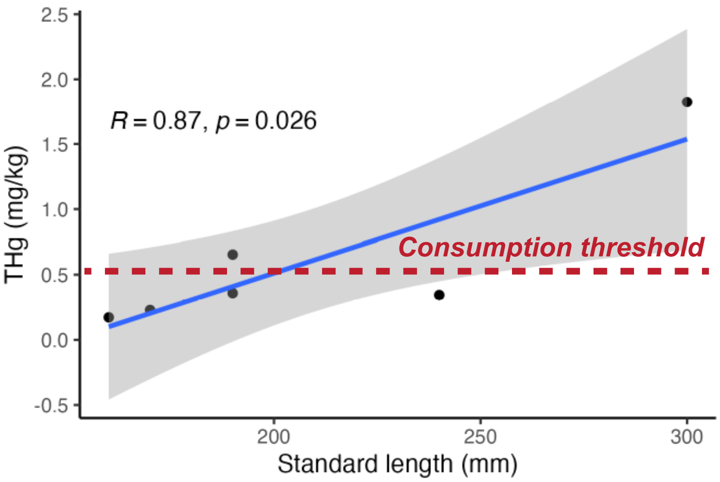


Figure 9. Relationship of the length and THg (mg/kg ww) concentrations for abyssal grenadier. Two abyssal grenadier (~300mm and ~180mm) were higher than the fish toxicity threshold for human consumption (0.5 mg/kg ww) which is displayed in the red trendline.

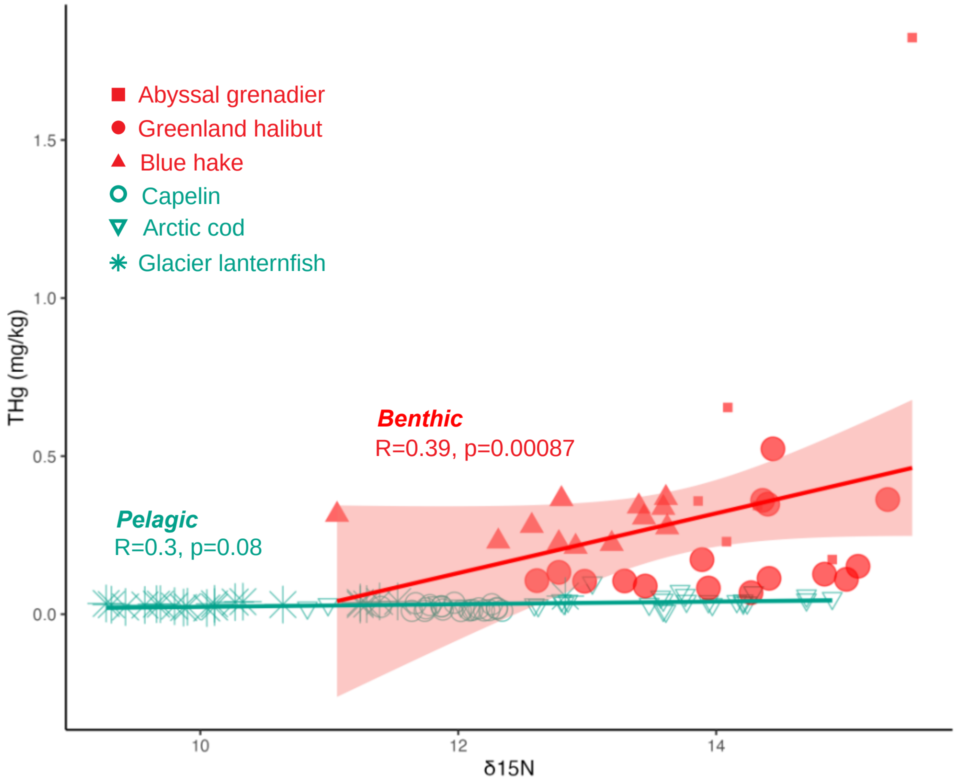


Figure 10. Trophic magnification slopes (TMS) of THg (mg/kg ww) by habitat type. Demersal fish (linear regression; n= 34; p-value<0.001) have a steeper TMS slope compared to pelagic fish (linear regression; n= 77;p-value=0.08).

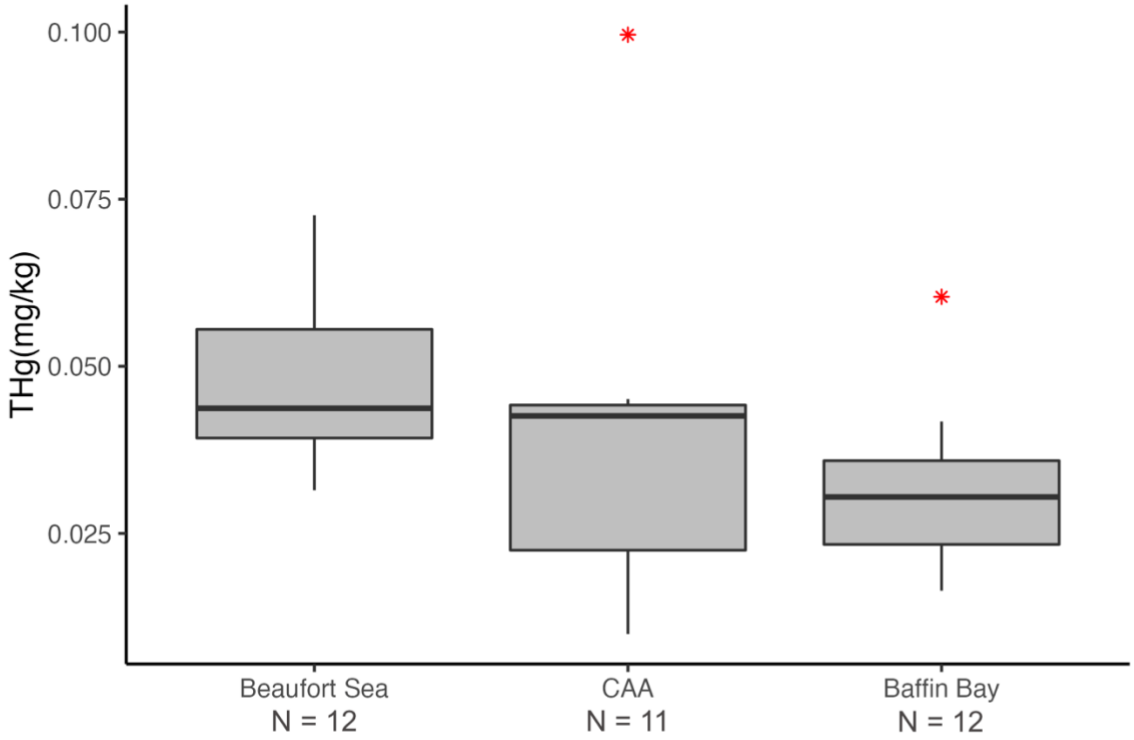


Figure 11. THg (mg/kg ww) in Arctic cod from the Beaufort Sea, the Canadian Arctic, and Baffin Bay. Baffin Bay is significantly different from the Beaufort Sea. The horizontal band inside the boxes represents the median and the lower and upper lines of the boxes represent the first and the third quartiles, respectively. The whiskers represent the maximum and minimum values of the lower and upper quartiles, respectively. Points that are greater than 1.5 times the first/or third quartile multiplied by the interquartile range are outliers and denoted by asterisks (*).

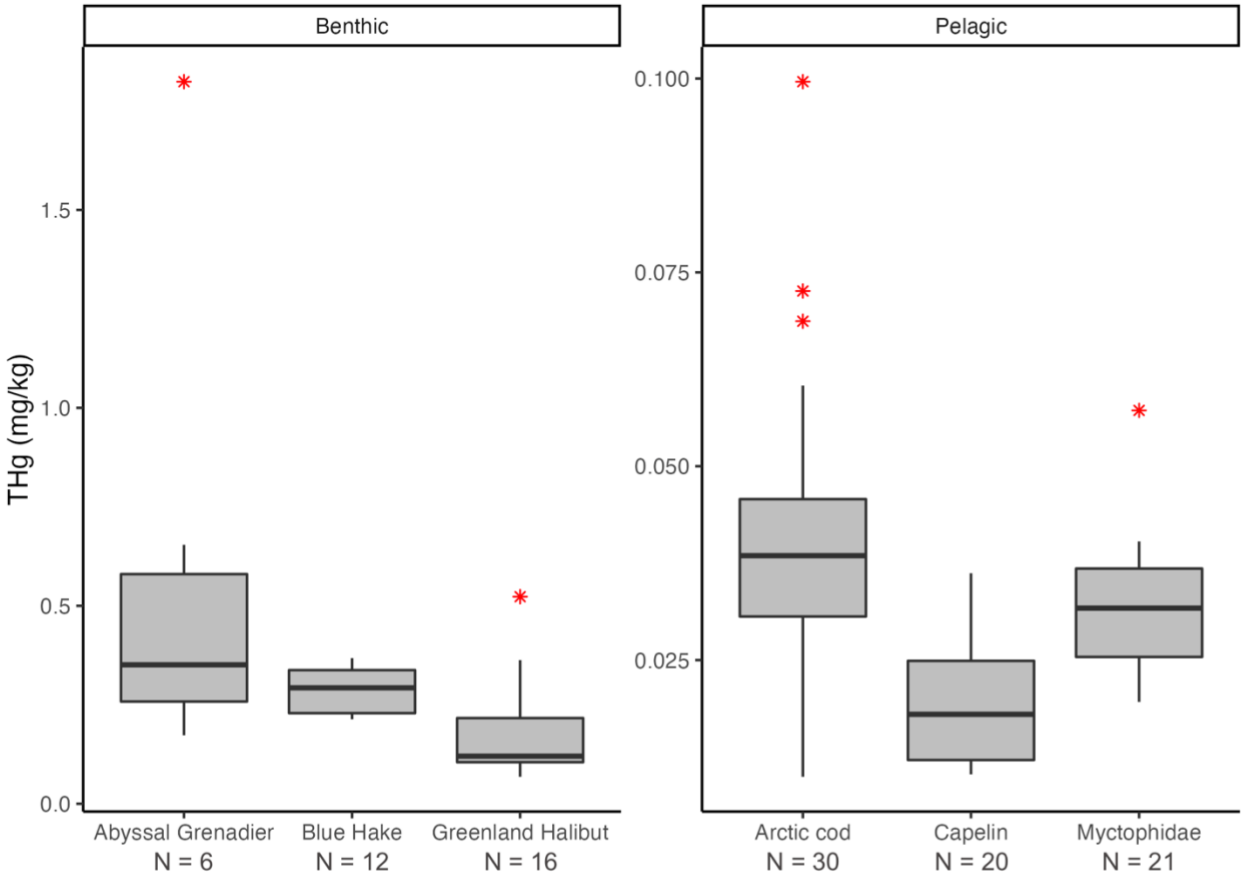


Figure 12. Boxplot comparing mean THg (mg/kg ww) concentrations across all species. Fish include abyssal grenadier and blue hake which were sampled from the Labrador Sea, Greenland halibut sampled from both Baffin Bay and the Labrador Sea, capelin sampled from the Labrador Sea, glacier lanternfish from both the Labrador Sea and Baffin Bay, and Arctic cod sampled from Baffin Bay, the Canadian Arctic Archipelago, and the Beaufort Sea. Total mercury concentrations were higher in demersal fish (0.07-1.82 mg kg^{-1}) compared to pelagic fish (0.01-0.07 mg kg^{-1}) (Kruskal-Wallis; p-value = $2.2e^{-16}$), as demonstrated by the different y-axis scales. The horizontal band inside the boxes represents the median and the lower and upper lines of the boxes represent the first and the third quartiles, respectively. The whiskers represent the maximum and minimum values of the lower and upper quartiles, respectively. Points that are >1.5 times the first/or third quartile multiplied by the interquartile range are outliers and depicted with asterisks (*).

2.3.3 Variation in pesticides across species, regions, and ecology

Species had different concentrations of pesticides. Pesticides that were detected in less than 50% of the samples were removed from further statistical analyses (Table 5). When comparing the wet weight concentrations of Greenland halibut, I found that Greenland halibut had higher on average pesticides concentrations than the other species (Table 5). Significant species differences were observed for ΣChlordane, ΣDDT, and ΣParlar (Figure 13).

To assess the spatial variation of pesticides in the Arctic and subarctic, I compared Greenland halibut and glacier lanternfish that were sampled in both the Labrador Sea and Baffin Bay (Figure 14 and Figure 15). The larger Greenland halibut from the Labrador Sea generally had higher concentrations of pesticides compared to the smaller Greenland halibut in Baffin Bay. However, only Hexachlorobutadiene was significantly higher in Baffin Bay, not the Labrador Sea. This suggests that OCP concentrations were not distinctly different in the Arctic or subarctic. Glacier lanternfish showed higher concentrations of most pesticides in Labrador Sea but only Dieldrin was significant. The lack of significance difference in OCPs for both species and regions is likely because both sampled fish are part of the same populations. For example, lanternfish from Baffin Bay are part of the same population as those from the NW Atlantic because they were likely advected into Baffin Bay.

In addition to species and spatial differences, I identified ecological variation. Four of the six OCPs increased significantly with $\delta^{15}N$. Higher trophic feeders had higher pesticide concentrations (Figure 16). To test this further, I performed a nonmetric multidimensional scaling analysis (NMDS) with habitat, species, and area (Baffin Bay and Labrador Sea) as variables. I found that

OCPs were significantly different in pelagic and demersal habitats (Figure 17) (ANOSIM, p-value=0.04) and in different species (ANOSIM, p-value<0.001) but were not significantly different in the Labrador Sea and Baffin Bay (ANOSIM, p-value=0.10). In other words, some pesticides increased in *concentration* with $\delta^{15}N$ (Figure 16) and differed in *kind* across habitats and species (Figure 17).

Table 5. Summary table of the concentration in OCPs (ng/g ww) that were detected in over 50% of samples. Average values are represented with the ranges in parentheses. Sample size (n) represents individual fish for demersal species and pools for pelagic species. CAA is the Canadian Arctic Archipelago.

	Demersal				**Pelagic**					
	Abyssal grenadier	Blue Hake	Greenland Halibut		Capelin	Glacier lanternfish		Arctic cod		
	Labrador Sea	Labrador Sea	Labrador Sea	Baffin Bay	Labrador Sea	Baffin Bay	Labrador Sea	Beaufort Sea	CAA	Baffin Bay
	n = 6	***n = 12***	***n = 6***	***n = 5***	***n = 6***	***n = 6***	***n = 12***	***n = 0***	***n = 0***	***n = 0***
Lipid (%)	7.8 (6.8 – 9.1)	12.9 (5.7-13.2)	37.4 (31.2-42.4)	7.1 (6.4-8.1)	8.9 (6.2-11.3)	29.3 (24.8-33.0)	20.7 (9.2-30.0)	n.a.	n.a.	n.a.
Dieldrin	2.1 (0-7.4)	0.2 (0-0.9)	10.4 (0-33.4)	1.3 (0-2.5)	0.2 (0-0.5)	0 (0-0)	0.7 (0-1.4)	n.a.	n.a.	n.a.
ΣEndrin	1.3 (0-6.18)	0.1 (0-1.0)	11.0 (4.1-21.8)	0 (0-0)	0.03 (0-0.08)	0.98 (0-2.4)	0.3 (0-1.2)	n.a.	n.a.	n.a.
Hexachloro-butadiene	0.01 (0-0.03)	0.01 (0-0.04)	0.06 (0-0.1)	0.4 (0-1.2)	0.03 (0-0.06)	0.1 (0-0.2)	0.2 (0-0.9)	n.a.	n.a.	n.a.
ΣChlorobenzene	0.9 (0.1-2.4)	0.4 (0.02-0.8)	10.2 (4.6-16.7)	2.1 (0.2-3.3)	0.4 (0-0.7)	1.9 (1.5-2.5)	1.0 (0.8-1.4)	n.a.	n.a.	n.a.
ΣChlordane	3.7 (0.09-16.1)	0.4 (0-0.9)	10.3 (2.7-16.8)	2.8 (0.3-5.2)	0.49 (0-0.1)	0.5 (0-1.1)	0.2 (0-1.1)	n.a.	n.a.	n.a.
ΣDDT	7.5 (1.1-18.7)	0.7 (0-1.9)	36.3 (8.1-80.2)	1.8 (0-4.8)	1.3 (0.6-1.8)	1.9 (0.3-3.4)	1.1 (0.6-2.3)	n.a.	n.a.	n.a.
ΣParlar	4.2 (0-11.9)	0.4 (0-1.5)	33.4 (5.2-67.7)	3.8 (0-7.6)	0.7 (0-1.2)	1.9 (0-5.5)	0.3 (0-1.5)	n.a.	n.a.	n.a.

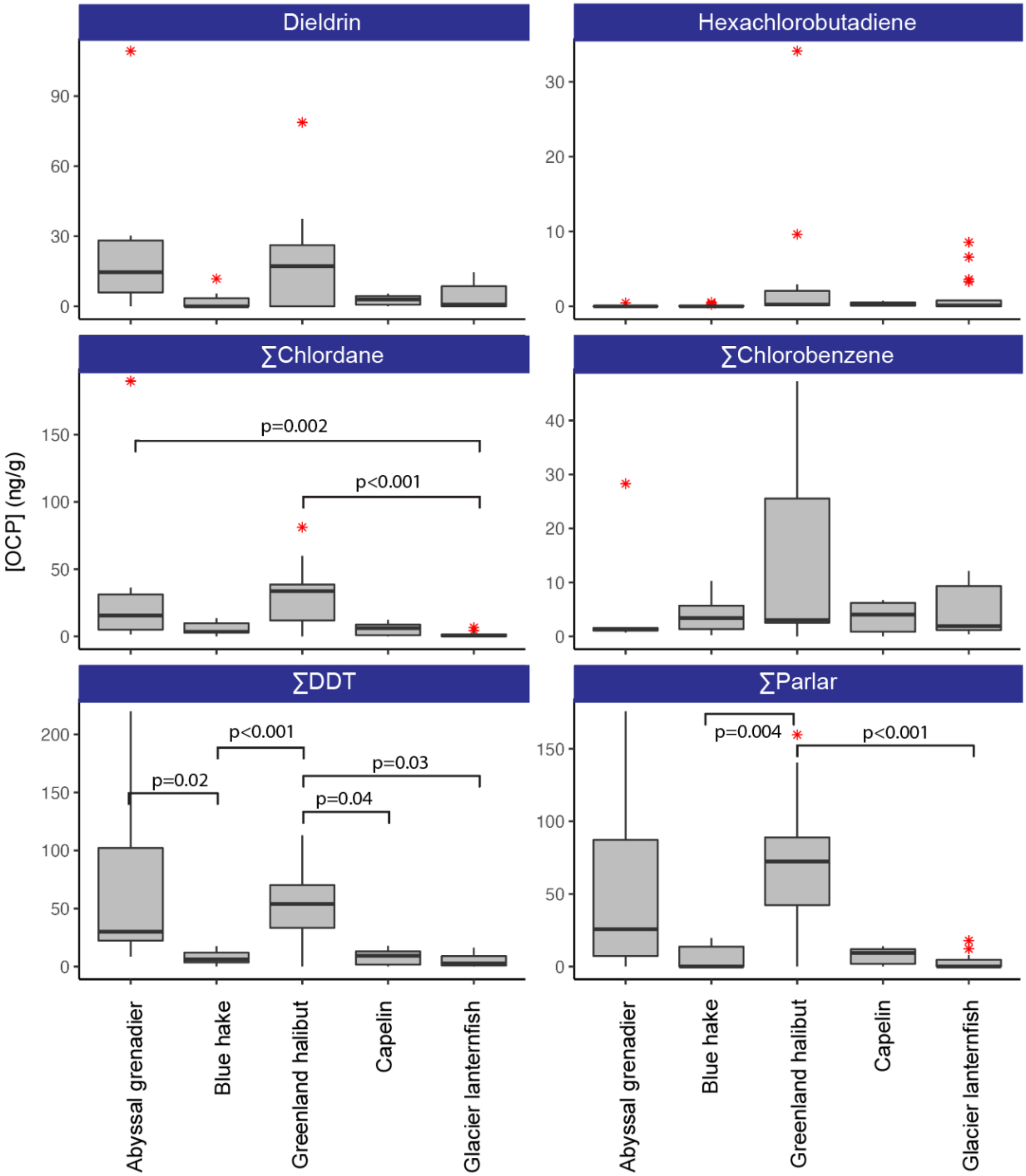


Figure 13. Boxplot of organochlorine pesticide concentrations across species (detected in >50% of samples) (ng/g lw). Greenland halibut had the highest concentrations across all pesticides. Significance was determined using Dunn's test between species pairs. Only significant p-values are displayed. Note that the y-axis has a different scale for each contaminant. Boxes represent the interquartile range, the whiskers represent data within 1.5 times the interquartile range and the asterisks indicate the remaining outlier data (*).

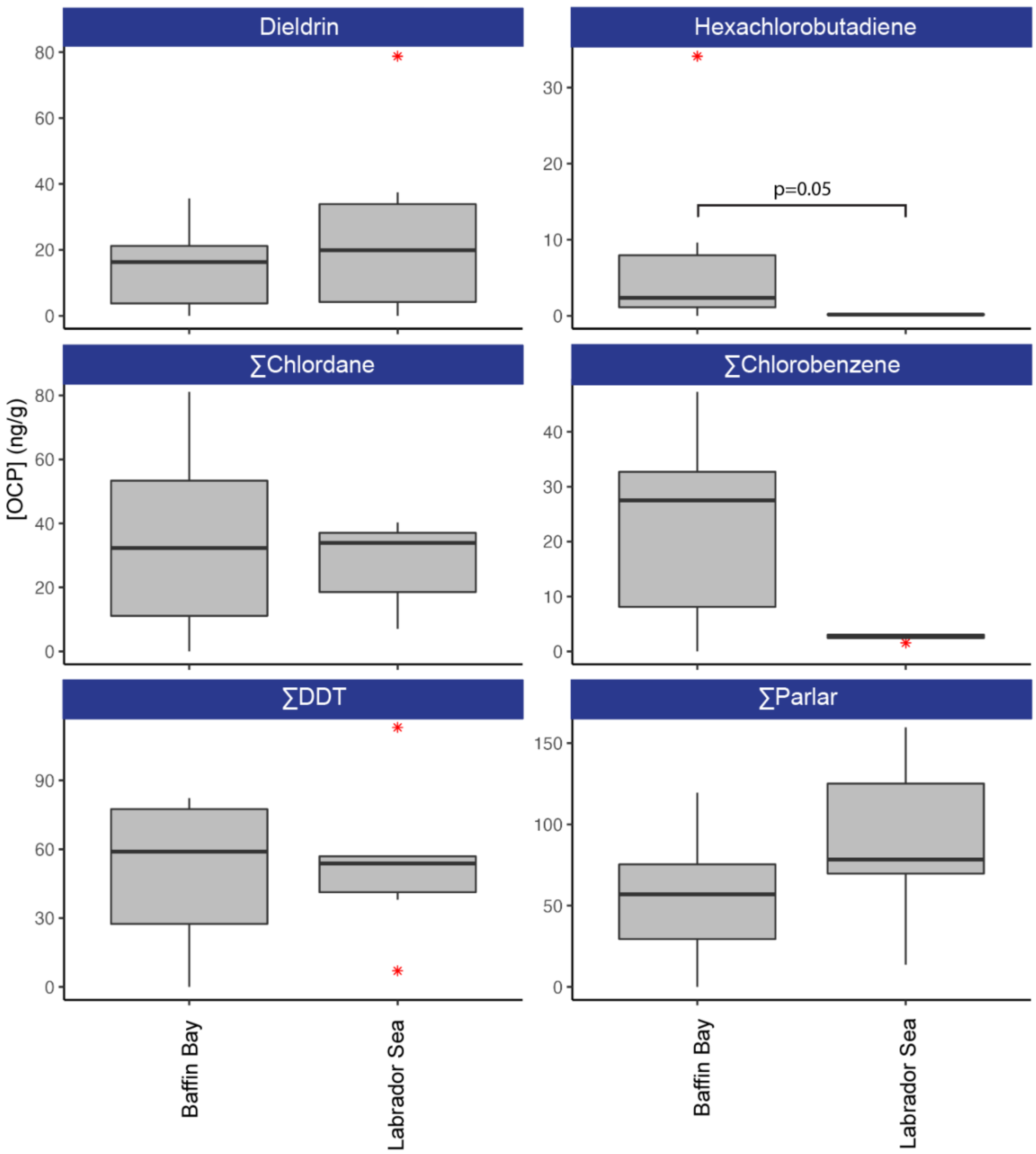


Figure 14. Boxplots comparing pesticides (ng/g lw) between the Labrador Sea and Baffin Bay for Greenland halibut. Significance was determined using Dunn's test between species pairs. Only significant p-values are displayed. Note that the y-axis has a different scale for each contaminant. Boxes represent the interquartile range, the whiskers represent data within 1.5 times the interquartile range and the asterisks indicate the remaining outlier data (*).

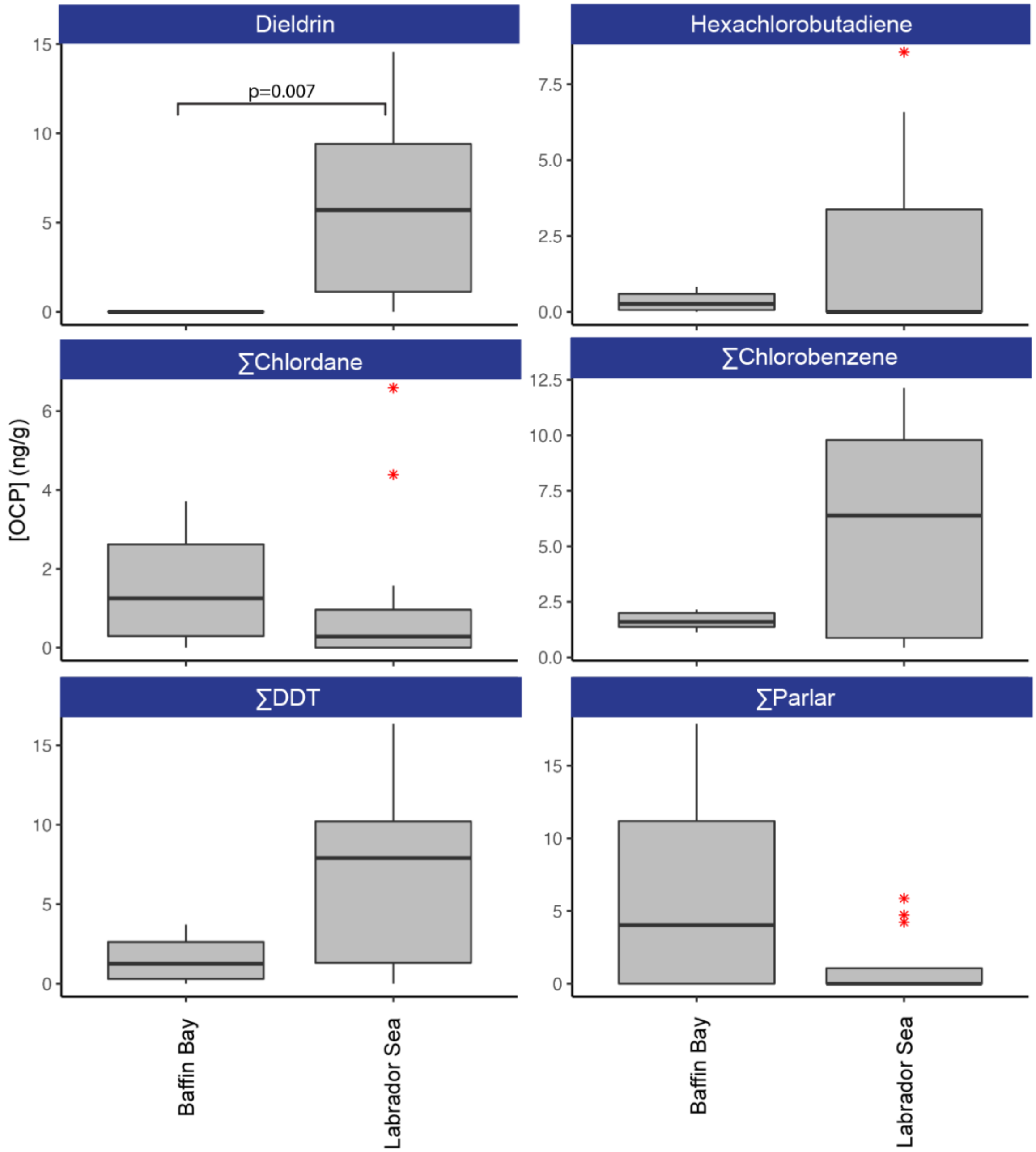


Figure 15. Boxplots comparing pesticides (ng/g lw) between the Labrador Sea and Baffin Bay for glacier lanternfish. Significance was determined using Dunn's test between species pairs. Only significant p-values are displayed. Note that the y-axis has a different scale for each contaminant. Boxes represent the interquartile range, the whiskers represent data within 1.5 times the interquartile range and the asterisks indicate the remaining outlier data (*).

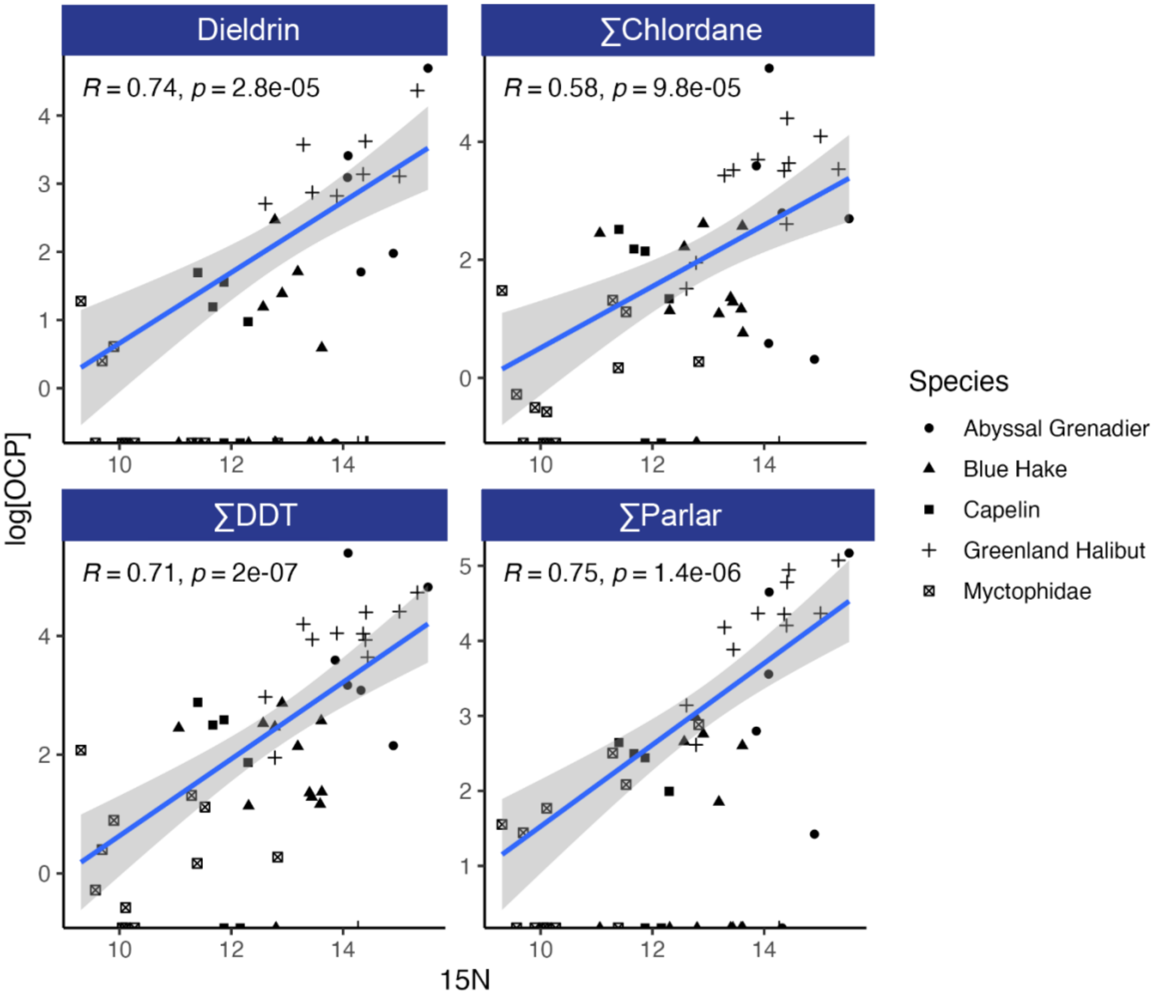


Figure 16. Linear regression of the OCPs (ng/g lw) that were significantly correlated with trophic feeding ($\delta^{15}N$). Four out of six pesticides statistically increased with $\delta^{15}N$ (alpha <0.05) and none with $\delta^{13}C$.

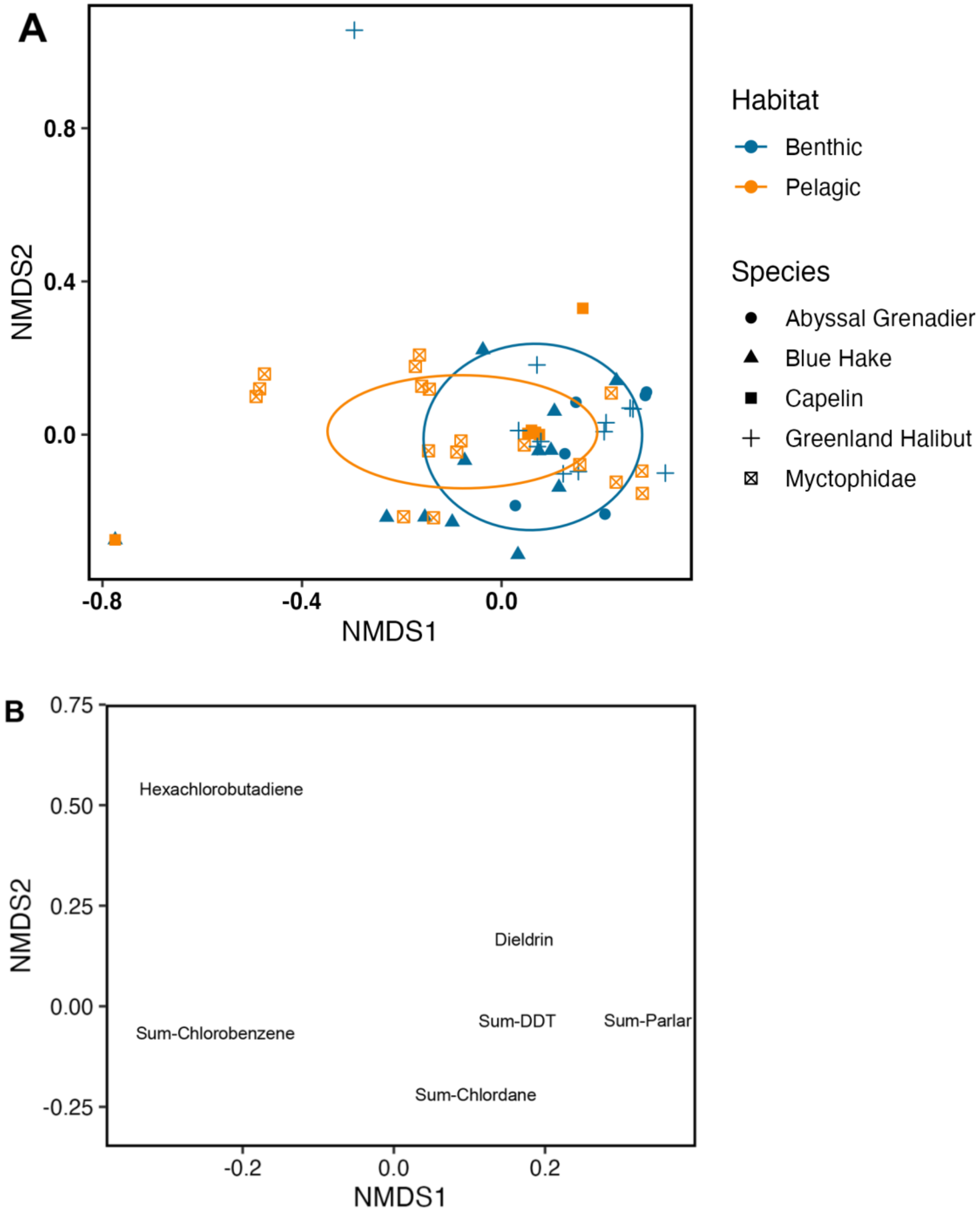


Figure 17. Nonmetric multidimensional scaling analysis (NMDS) of the OCPs (ng/g lw) by species and habitat detected in > 50% of the fish categorized by (A) species and habitat type and (B) scores plot of the OCPs accounting for the separation of the groups. Pesticides are significantly different in different species and in different habitats (species, ANOSIM p-value<0.001; habitat, ANOSIM p-value <0.001). Ellipses represent one standard deviation from the mean.

2.4 Discussion

This study offers novel insights into the ecological patterns which drive contaminant concentrations in Arctic and subarctic fish and, in turn, in northern food webs. The most recent study describing these patterns in the Arctic were conducted by Pedro et al. (2017) and focused on capelin and Arctic cod. By broadening our sampling range into the Labrador Sea and by including benthic fish with different trophic niches I found that mercury significantly varied by species, trophic feeding level, area, and habitat. Organochlorine pesticides varied significantly by species, habitat, trophic feeding, but not location. Our findings suggest that ecological changes driven by climate could influence contaminant uptake by Arctic piscivores. THg reported here were within the same range as previous studies (Barber et al., 1984; Julshamn et al., 2006; Pedro et al., 2017; Grimaldo et al., 2020; Gopakumar et al., 2021) but our pesticide values were orders of magnitude less than all past studies (Berg et al., 1997; Vorkamp et al., 2004; Pedro et al., 2017).

2.4.1 Mercury concentrations compared to literature

Mercury concentrations in glacier lanternfish from this study were consistent with those previously observed in the North Atlantic (Grimaldo et al., 2020) (Table 6). Mercury concentrations in capelin were also consistent with previous studies and there was little variation across the sites (Arviat, the NW Barents Sea, and the Labrador Sea) (Pedro et al., 2017; Gopakumar et al., 2021). Similarly, Arctic cod in Baffin Bay had the same range of concentration as Arctic cod in Clyde River and Arctic cod in the Canadian Arctic Archipelago had the same range as Arctic cod in Resolute Bay (Pedro et al., 2017). THg in Arctic cod was highest in the Beaufort Sea (this study) and lowest in the NW Barents Sea (Gopakumar et al., 2021). Furthermore, THg in Blue hake was consistent with Barber et al. (1984) in the Labrador Sea. Because of knowledge gaps, I could only compare

Greenland halibut from this study to the Barents Sea where they shared overlap in their THg ranges, but mean THg was lower in the Barents Sea (Table 6).

Table 6. Comparison of THg concentrations by species and region with previous literature. Total mercury (THg) is reported in mg/kg ww. CAA is the Canadian Arctic Archipelago.

Species	Location	THg	Source
Lanternfish	North Atlantic	0.039	(Grimaldo et al., 2020)
	Baffin Bay	0.04 (0.02-0.05)	(this study; 2019,2020,2021)
	Labrador Sea	0.03 (0.02-0.04)	(this study; 2019,2020,2021)
Capelin	NW Barents Sea	0.03 (0.01-0.04)	(Gopakumar et al., 2021)
	Arviat	0.01 (0.01-0.02)	(Pedro et al., 2017)
	Labrador Sea	0.02 (0.01-0.03)	(this study; 2018, 2021)
Arctic cod	NW Barents Sea	0.03 (0.02- 0.04)	(Gopakumar et al., 2021)
	Clyde River	0.03 (0.02–0.05)	(Pedro et al., 2017)
	Resolute	0.03 (0.02–0.04)	(Pedro et al., 2017)
	Beaufort Sea	0.05 (0.03-0.07)	(this study; 2016, 2019, 2021)
	CAA	0.04 (0.01-0.10)	(this study; 2016, 2019, 2021)
	Baffin Bay	0.03 (0.02-0.06)	(this study; 2016, 2019, 2021)
Blue hake	Labrador Sea	0.34	(Barber et al., 1984)
	Labrador Sea	0.29 (0.21-0.37)	(this study, 2017)
Greenland halibut	Barents Sea	0.06 (0.02-0.10)	(Julshamn et al., 2006)
	Labrador Sea	0.32 (0.13-0.52)	(this study, 2017)
	Baffin Bay	0.11 (0.07-0.15)	(this study, 2021)

2.4.2 Mercury variation across species explained by ecological dynamics

Mercury was highest in benthic fish, lower in benthopelagic fish, and lowest in pelagic fish. This was consistent with the linear model which returned carbon source ($\delta^{13}C$) as a significant predictor of THg (Table 4). Greenland halibut had the lowest concentration of THg compared to abyssal grenadier and blue hake (Figure 12). Because Greenland halibut are benthopelagic, they spend less of their life foraging on the seafloor and have less exposure to Hg-resuspension by sediments. In contrast to abyssal grenadier which are confined to cold deep waters, Greenland halibut are more abundant in the less saline, higher oxygenated waters above (Murua & de Cárdenas, 2005). Bottom waters with lower pH and anoxic conditions amplify bioaccumulation (Greenfield et al., 2001) which reduce contaminant excretion rates in demersal fish (Trudel & Rasmussen, 1997), and ultimately lead to higher levels of THg. Higher THg in bottom-dwelling fish is consistent with previous studies who found that fish living near sediment had higher mercury concentrations than similar fish living in the water column (Romero-Romero et al., 2022). Furthermore, grenadiers maintain slow aerobic activity to conserve energy at depths and account for food scarcity (Collins 1999) and as such are longer lived (up to 53 years) (Lee et al., 2019) than Greenland halibut (up to 30 years total; sexual maturity: 4-6 males; 9-10 years females) (Dwyer et al., 2013). I propose that higher THg in abyssal grenadier could also be because abyssal grenadier are slower growing and have more time to bioaccumulate THg than Greenland halibut. Note that Hilgendag et al. (2022) found the exact opposite where benthopelagic species had higher THg biomagnification than species in pelagic and benthic food webs. The discrepancies between our findings for fish and those of Hilgendag et al. (2022) could be because they included lower trophic organisms in their analyses (e.g. amphipods). Bioaccumulation rates of lower trophic organisms could be less impacted by changes in pH compared to fish (Watras et al., 1998) and as such, changes pH near sediments would have a greater effect on fish and less on amphipods.

Arctic cod had higher mercury concentrations than capelin. I found two main differences between capelin and Arctic cod which might explain their differences in THg: (1) both are planktivores but capelin feed lower on the trophic web (δ^{15}N) than Arctic cod, and (2) capelin are pelagic whereas Arctic cod are benthopelagic (δ^{13}C). I argue that THg variation between capelin and Arctic cod is explained by their feeding ecology (δ^{15}N) and not their different habitats (δ^{13}C). We confirmed using our model that feeding ecology (δ^{15}N) had a more significant impact on THg variation than habitat (δ^{13}C) (Table 4). I can use capelin and Arctic cod as an example to reinforce that feeding higher on the food chain increases bioaccumulation in Arctic and subarctic fish. Capelin were previously deemed as the replacement species for Arctic cod because of the borealization of Arctic ecosystems (Ullrich and Tallman, 2021). Based on the findings, I predict that THg would decrease in Arctic piscivorous predators with the increased availability of capelin in northern foodwebs. Top predators would have a slower rate of bioaccumulation increase because of lower δ^{15}N in capelin.

2.4.3 Spatial variation in mercury concentrations across the Canadian Arctic

Mercury concentrations were highest in Arctic cod from the Beaufort Sea and lower in other regions. These findings are consistent with previous studies who found higher levels of mercury in ringed seals and polar bears in the western Canadian Arctic compared to the eastern Canadian Arctic (Wagemann et al., 1996; Rigét et al., 2005; Braune et al., 2015) (Figure 11). Higher THg in Arctic cod from the Beaufort Sea compared to the rest of the Canadian Arctic is likely because of

(1) the increased input of THg from the Mackenzie River (Stern & Macdonald, 2005; Andersson et al., 2008; Brown et al., 2018), the largest and longest river in Canada which drains into the Beaufort Sea; (2) closer proximity to Asian atmospheric emissions (Brown et al., 2018), and (3) the increased methylation of inorganic mercury to organic mercury caused by a decreased dissolved oxygen concentration (Wang et al., 2012; Brown et al., 2018). Despite efforts in North America and Europe to reduce Hg emissions, dependencies on coal have led to Asia becoming the most important source of Hg globally (AMAP, 2011). Estuaries receiving higher inputs of atmospheric Hg have higher methylation rates than non-estuarine areas (Barber & Massom, 2007). I anticipate that ongoing emissions and rivers transporting elevated Hg levels from southern latitudes will continue to amplify Hg in in fish-eating wildlife in the western Arctic and exacerbate the difference in Hg concentrations between the western and eastern Canadian Arctic.

2.4.4. Pesticides concentrations compared to literature

I compared pesticides concentrations in this study to past studies (Table 7). Capelin from the Labrador Sea and Greenland halibut from Baffin Bay were lower in Chlorobenzene than what was observed for Chlorobenzene by Vorkamp et al., (2004) in Nuuk, Greenland by two orders of magnitude (Table 7). Compared to capelin in Arviat, Nunavut (33.6 ng/kg), I found lower ΣDDT (1.3 ng/kg) in the Labrador Sea (Pedro et al., 2017) suggesting either higher levels in northern ecosystems or variation in sampling techniques. The largest discrepancy between this study and past studies was the comparison with ΣDDT in blue hake from the Labrador Sea and Baffin Bay (Berg et al., 1997). ΣDDT levels were four orders of magnitude higher in blue hake from Baffin Bay (410-540mm) than blue hake from the Labrador Sea (430-590mm). Because of the large uncertainty in ΣDDT reported by Berg et al. (1997), Greenland halibut from Baffin Bay were comparable to our findings for Greenland halibut in the Labrador Sea. In contrast, values of Greenland halibut in studies that were based in Baffin Bay were different. Berg et al. (1997) reported higher values (102 ng/kg) than our observations (1.8 ng/kg). Similar results were observed for ΣChlordane which were higher (238 ng/kg) in blue hake in Baffin Bay in 1997 compared to our values for blue hake (0.4 ng/kg) for the Labrador Sea.

Table 7. Comparison with the pesticide concentrations by species (all muscle tissue) and region with previous literature. Organochlorine pesticides are reported as ng/g lw. Arctic cod were not included in the table because they were not analyzed for OCPs. Abyssal grenadier and glacier lanternfish were not included because of missing data for comparisons.

Species	Location	ΣChlorobenzene	ΣDDT	ΣChlordane	Source
Capelin	Nuuk	51 (37-79)			(Vorkamp et al., 2004)
	Arviat		33.6 (17.0–54.0)		(Pedro et al., 2017)
	Labrador Sea	0.4 (0-0.07)	1.3 (0.6-1.8)		(this study, 2018, 2021)
Blue Hake	Baffin Bay		1446 (± 415)	238 (+- 72)	(Berg et al., 1997)
	Labrador Sea		0.7 (0-1.9)	0.4 (0-0.9)	(this study, 2017)
Greenland halibut	Baffin Bay		102 (± 87)	173 (+-104)	(Berg et al., 1997)
	Nuuk	51 (21-62)			(Vorkamp et al., 2004)
	Labrador Sea	10.2 (4.6-16.7)	36.3 (8.1-80.2)	10.3 (2.7-16.8)	(this study, 2017)
	Baffin Bay	2.1 (0.2-3.3)	1.8 (0-4.8)	2.8 (0.3-5.2)	(this study, 2021)

2.4.5 Organochlorine pesticide variation by species explained by ecological dynamics

Pesticides varied by species and habitat. I found that OCPs were significantly different in pelagic and demersal habitats (Figure 17) and demersal fish had higher pesticide concentrations than pelagic fish (Figure 13; Table 5). Higher levels of pesticides in species closer to the bottom could be explained by two processes; (1) because they feed higher in trophic webs; and (2) because benthic habitats have elevated levels of OCPs linked to the biological pump. The biological pump sequesters atmospheric CO_2 and POPs via air-water exchange to the deep ocean (Dachs et al., 2002). OCPs have a low solubility in water is why they bind strongly to sinking organic particulate and act as a sink in the benthos (EPA, 2009). This is particularly true in Arctic ecosystems where most of the productivity in the water column and marginal ice zones is ungrazed and settles on the seafloor (Olli et al., 2002).This is why previous studies found that fish occupying similar trophic levels but using benthic rather than pelagic sources have higher POP concentrations because of sinking OCPs (Hebert & Haffner, 1991; Borgå et al., 2004). Sinking particulate with bound pesticides pose a particular risk to benthic species (like abyssal grenadier and blue hake) that live in close contact with bottom sediments and to the predators feeding on them.

2.4.6 Spatial variation of OCPs concentrations across the Arctic and subarctic

OCPs were higher in the Labrador Sea for Greenland halibut and glacier lanternfish. I found no evidence of different OCP signatures in subarctic and Arctic ecosystems (Figure 17), but trends showed the Labrador Sea had higher concentrations than Baffin Bay (with the exception of one pesticide for Greenland halibut which was higher in Baffin Bay [Hexachlorobutadiene]). Overall, there were only two significant pesticides in either location indicating little variation for Greenland halibut and lanternfish between both regions. Greenland halibut are highly migratory and thus the groups sampled in Baffin Bay (albeit smaller than those sampled in the Labrador Sea) are part of the same population. I expect that any differences in concentration between either region is likely a reflection of the different sizes of fish sampled not elevated environmental pesticides. Lanternfish also showed little variation in the two regions and only one pesticide was significantly different in lanternfish from both regions. The little variability for lanternfish is likely explained by currents where populations from the Labrador Sea are advected into Baffin Bay. All lanternfish were the same size and part of the same populations which explains similar OCP concentrations in both locations. Based on our findings for Greenland halibut and lanternfish, I am unable to conclude that there are distinct OCP signatures in Baffin Bay and the Labrador Sea for pelagic and demersal fish.

2.4.7 Future pollutant fields in the Canadian Arctic due to climate driven ecological changes

Mercury will likely decrease in benthic habitats. Benthic habitats are tightly coupled to surface production and are fueled by pelagic algae and macrophytes (Grebmeier et al., 2015; Renaud et al., 2015). With the reduction of sea ice and the warming of the Canadian Arctic, marine fauna are shifting from benthic to more pelagic trophic webs (Grebmeier et al., 2006). Although pelagic food webs are more efficient at transferring mercury (Hilgendag et al., 2022), this shift will result in a more pelagic fish-dominated ecosystem which is lower in Hg than a demersal fish-dominated ecosystem. I anticipate that a climate-induced shift towards pelagic food webs would result in a net decrease in THg in Arctic food webs.

Subarctic capelin moving northward into the Arctic will decrease or increase THg depending on the Arctic piscivore. Stomach content analysis of Arctic char in Cumberland Sound found that when available, Arctic char preferentially fed on capelin (Ulrich & Tallman, 2021). In 2011, capelin accounted for ~75% of the stomach contents of Arctic char whereas the year prior they were entirely absent from their diet. This is consistent with other studies who have found increased abundance of capelin in seabird diets across the Arctic (Gaston et al., 2012; Provencher et al., 2012). The authors suggested that this surge in capelin could be an episodic event or the beginning of a permanent change with capelin expanding their northern range. Although not conclusive, they observed that Arctic char feeding on aggregations of capelin showed positive correlations with growth. Arctic char are opportunistic feeders which is why they show preference towards capelin when available (Cote et al., 2021), but their diet primarily consists of amphipods (48-96%). Because of their switch from lower trophic prey to capelin, Arctic char feeding on high density aggregations of capelin would increase their THg exposure. Florko et al. (2021) found that capelin biomass will continue to increase while the biomass of Arctic cod decreases in the Hudson Bay. In contrast to Arctic char, this shift could benefit top predators like beluga whales because capelin are staples in their summer diet (Arctic cod is their staple in the winter) (Florko et al., 2021). The trade-off is that Arctic cod are more energetically rich compared to capelin, but they are also higher in THg. For Arctic char, capelin invasions will increase their THg exposure and benefit their growth (e.g. capelin are higher energetically than amphipods), but for belugas, capelin invasions will decrease their THg exposure and impact their growth (e.g. capelin are lower energetically than arctic cod) (Florko et al., 2021; Ulrich & Tallman, 2021). I conclude that the interaction between capelin and piscivorous predators is complex and there are energetic and contaminant trade-offs to be considered for each predator.

Pesticides will increase in Arctic top predators. Despite many OCPs being banned or heavily restricted, I predict that OCPs will continue to increase in the northern food webs because of decreased sea ice habitats (Brown et al., 2017). For example, McKinney et al. (2013) found that in warmer years polar bears fed more on subarctic prey (hooded seals) than Arctic prey (ringed seals). Polar bears had more access to prey with higher burdens of OCPs and as a result had higher OCPs bioaccumulated. In addition, large Greenland halibut in the Labrador Sea had higher OCPs than small Greenland halibut in the Arctic. Greenland halibut spawn in Baffin Bay and if their spawning grounds expand northward, it will introduce larger, higher-in-contaminants Greenland halibut further into the high Arctic. Subsequently, Greenland halibut would accelerate pesticide biomagnification on top predators (e.g. narwhals) harvested by Inuit (referred to as Maktaaq). Traditional Inuit diet relies on healthy fats and protein which have high nutritional value but bioaccumulate higher lipophilic pesticides (Hernández et al., 1994). Top predators like narwals feeding on large, contaminated Greenland halibut would accelerate pesticide accumulation in northern diets and have cascading impacts on the health of Indigenous Peoples.

Chapter 3 - General Conclusions

Overall, this study showed that ecological climate-driven changes can influence contaminant concentrations. The current trend of decreased sea ice extent and earlier sea ice breakup is projected to continue in the Canadian Arctic and have consequences for contaminant bioaccumulation by shifting trophic structures and increasing the availability of subarctic prey (McKinney et al., 2013; Brown et al., 2017). For example, in the Beaufort Sea, shorter ice-free seasons have led to a lower survival of young, less contaminated Arctic cod leaving older, more contaminant Arctic cod as the predominant prey (Gaden et al., 2009). I had hypothesized that subarctic species could act as biological vectors transporting elevated contaminant levels from southern latitudes but our comparisons with subarctic and arctic species were skewed by mismatched fish sizes. Instead, what this told us was that size/age was a more important driver of the contaminant differences. I proposed that the relative size of incoming fish moving into the polar regions is more indicative of future contaminant burdens than its proximity to anthropogenic activity. This aligned with the main finding in our study which is that THg and OCPs variability was best explained by species. Species-specific variables like size/age, physiology, and uptake and excretion rates of contaminants are likely just as important if not more to consider than ecological influences.

3.1 Future work

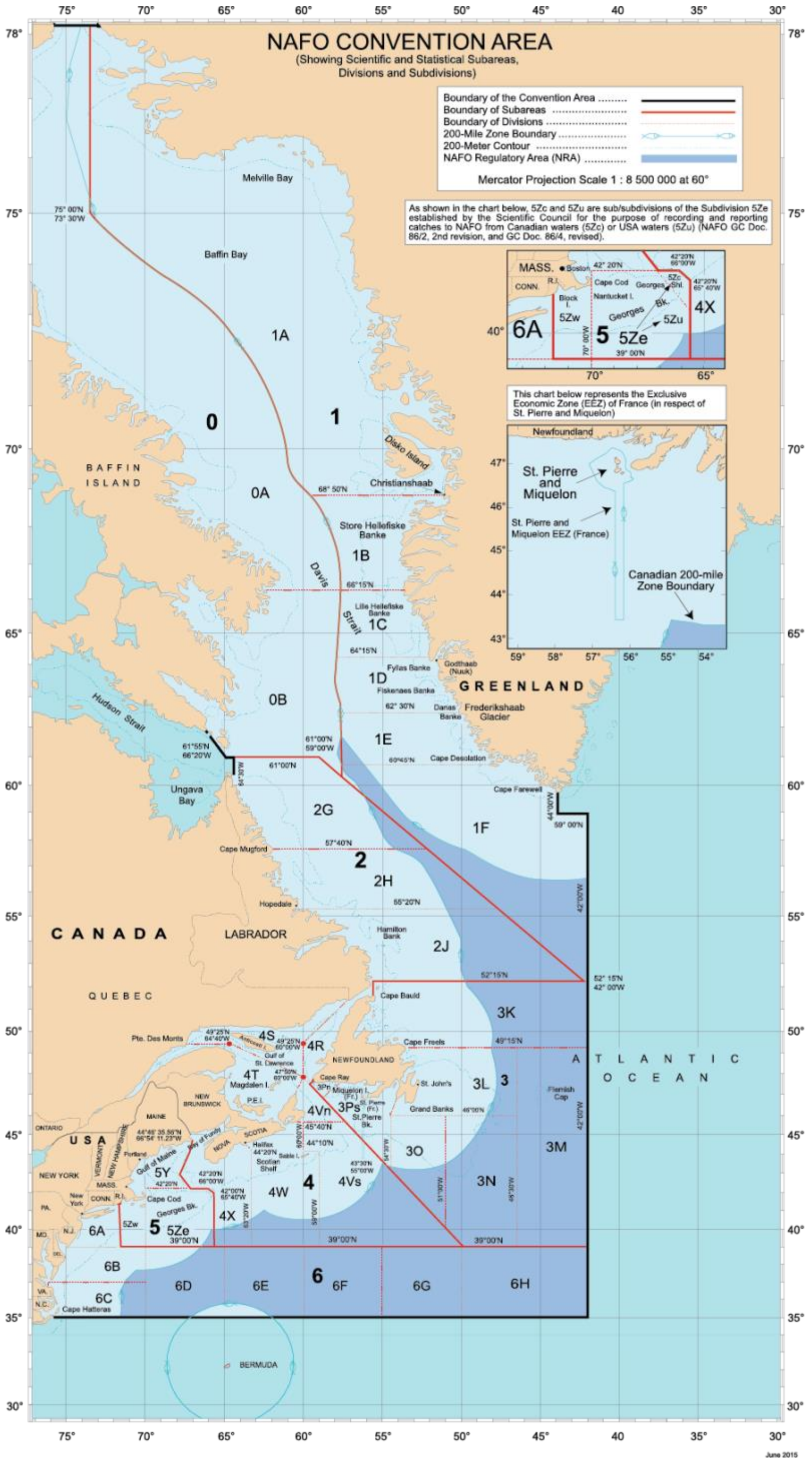
As mentioned above, one of the main takeaways from this research is that *species* was the best variable to explain THg and OCP variability. To predict how invading prey will affect contaminant bioaccumulation, we must know their initial concentrations. Little is still known about distribution of lanternfish in the North Atlantic and even less is known about their contaminant concentrations. Future contaminant models relying on trophic structures should include glacier lanternfish where their northern range overlaps with Arctic ecosystems (e.g. Baffin Bay) (Priou et al., submitted). In addition, distinguishing the normal migration patterns for fish like lanternfish as opposed to their northward expansion is needed. Even though spatial differences were found to be less important in our study, understanding where fish migrate to and from could help anticipate the type of contaminants they could be introducing. Cote et al. (2021) speculated that invasions in the Canadian Arctic might come from the NE Atlantic rather than Labrador. If that is the case, those fish would have different contaminant profiles to those in the Labrador Sea.

One variable that I did not study was the influence of coastal and offshore feeding ecology. THg accumulates in terrestrial and freshwater ecosystems where methylation is higher than oceanic ecosystems (Gamberg et al., 2005). Declining fatty acid carbon ratios ($\delta^{13}C$) have shown shifts from benthic/nearshore/ice-associated prey to pelagic/off-shore/open-water-associated prey (McKinney et al., 2013). Including anadromous fish like Arctic char as a study species would provide valuable insights into contaminant bioaccumulation in both habitats. I suggest using sulphur ($\delta^{34}S$) as it is useful in distinguishing between marine versus freshwater resources and can quantify the differences of offshore and nearshore prey (Fry and Chumchal, 2011; Connolly et al., 2004).

In this study, I assumed that shifts in trophic structures and feeding behaviour were attributed to environmental changes (McKinney et al., 2022). I did not consider the possibility that trophic changes were the result of endocrine disrupting contaminants altering foraging behaviour (e.g.

Tartu et al., 2014). Letcher et al. (2010) highlighted the importance of studying the interactive effects of climate change, anthropogenic emissions, habitat alteration, and contaminant-induced behavioural changes to provide a holistic understanding of contaminant concentrations in biota. Future work could include physiological-based pharmacokinetic modelling which has been successful in disentangling these interactions (Sonne, 2010; Dietz et al., 2015). This study provides an extra piece to the puzzle by highlighting the importance of species-specific and ecological variables in those models to better assess the fate of contaminants in Arctic marine food webs.

Appendix



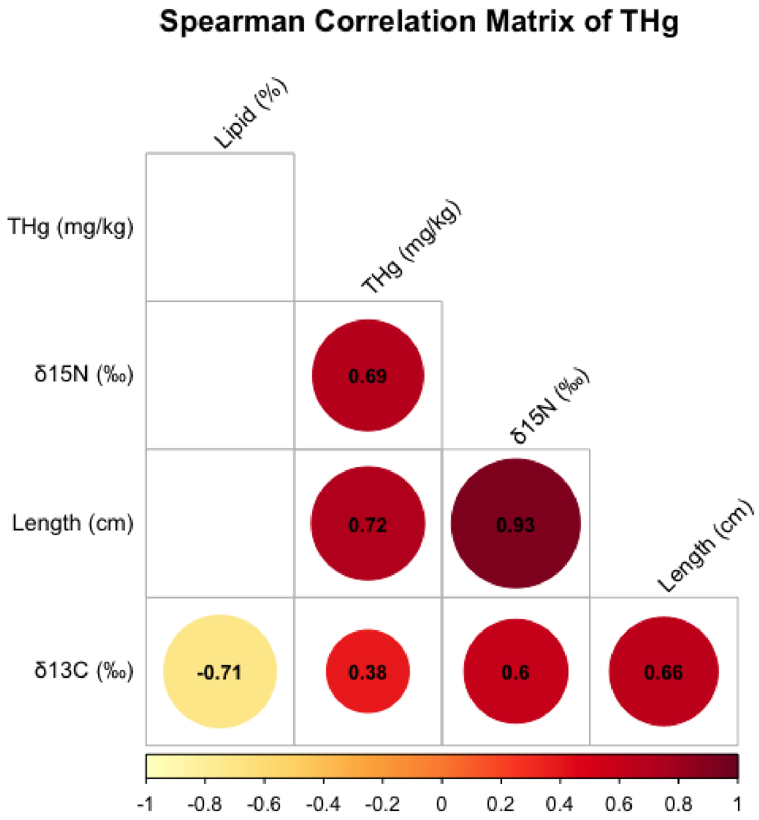
As shown in the chart below, 5Zc and 5Zu are sub/subdivisions of the Subdivision 5Ze established by the Scientific Council for the purpose of recording and reporting catches to NAFO from Canadian waters (5Zc) or USA waters (5Zu) (NAFO GC Doc. 86/2, 2nd revision, and GC Doc. 86/4, revised).

This chart below represents the Exclusive Economic Zone (EEZ) of France (in respect of St. Pierre and Miquelon)

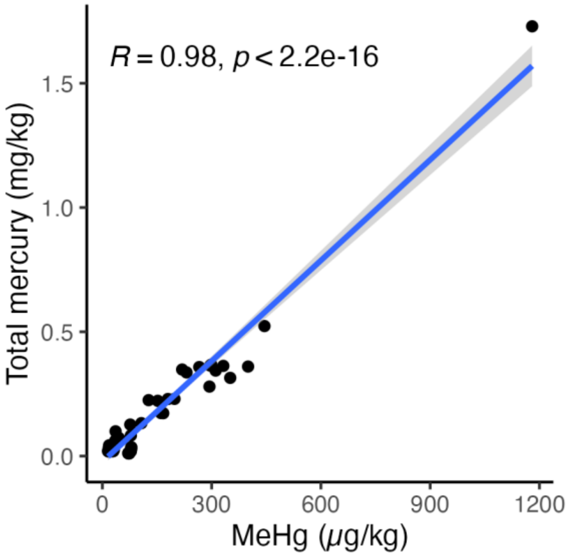
Appendix A: Northwest Atlantic Fishing Organization (NAFO) fisheries management areas. *©Northwest Atlantic Fisheries Organization.*

Hexachlorobutadiene
1,2,4,5-Tetrachlorobenzene
1,2,3,4-Tetrachlorobenzene
Pentachlorobenzene
Hexachlorobenzene
3,4,5,6-Tetrachloroveratrole
Pentachloroanisole
alpha-BHC
beta-BHC
gamma-BHC
delta-BHC
Pentachloronitrobenzene
Heptachlor
4,4'-DDNU
Parlar 62
Endosulfan Sulfate
Dacthal
Chlorpyrifos
Isodrin
Octachlorostyrene
Heptachlor Epoxide B
Heptachlor Epoxide A
Oxychlordane
4,4'-DDMU
trans-Chlordane
cis-Chlordane
trans-Nonachlor
Dieldrin
Endrin
cis-Nonachlor
Endosulfan I
Endosulfan II
Aldrin
2,4'-DDE
4,4'-DDE
2,4'-DDD
4,4'-DDD
2,4'-DDT
4,4'-DDT
Endrin Aldehyde
Endrin Ketone
Methoxychlor
Dicofol
Mirex
Parlar 26
Parlar 50

Appendix B. Total list of 46 OCPs analyzed by ALS Environmental.



Appendix C. Spearman correlation plot showing that length and nitrogen are highly correlated.



Appendix D. Linear regression of THg and MeHg in fish muscle tissues where the relationship between the two is significantly correlated (1:1).

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