New insight into short-chain chlorinated paraffin accumulation and contaminant-climate change interactions in a key Arctic monitoring species

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General Abstract

Many legacy persistent organic pollutants (POPs) have been extensively studied in Arctic wildlife, however, newer POPs such as short-chain chlorinated paraffins (SCCPs) have received considerably less attention in Arctic contaminant monitoring studies. In Chapter 3, I quantified the tissue concentrations of SCCPs (C_{10}-C_{13}) in Arctic marine fish, invertebrates, and ringed seals (*Pusa hispida*) from western Hudson Bay, Canada and evaluated the biomagnification potential of SCCPs within this food web. Average concentrations of ΣSCCPs in these biotas varied from 38.3 to 687 ng g^{-1} lipid weight. Despite sometimes high concentrations in fish, trophic-adjusted biomagnification factors of SCCP congeners ranged from just 0.07 to 0.55 for small pelagic fishes to ringed seals, suggesting limited biomagnification potential of SCCPs in this food web.

Across all key environmental contaminants of the Arctic, further understanding of how climate-related ecological changes can impact their concentrations in Arctic wildlife life is needed. In Chapter 4, I compared feeding ecology and contaminant concentrations (legacy and new POPs, and trace metals) in ringed seals from two Canadian Arctic sites (Resolute Bay, Sachs Harbour) and two sub-Arctic sites (Arviat, Nain). Climate-related feeding changes have been documented in ringed seals at the sub-Arctic locations, while less change has been reported at the Arctic locations, suggesting current sub-Arctic conditions may aid in predicting future Arctic conditions. Using blubber fatty acids (FA) and stable isotopes (SI) as dietary tracers, we found distinct FA signatures between seals from Resolute Bay and both sub-Arctic locations, likely related to higher consumption of invading southern species in the sub-Arctic but continued feeding on native species in Resolute Bay. The Sachs Harbour seals showed overlapping FA signatures with all three other sites, suggesting a mixed diet of native and invading species.

Using generalized linear models, SI best-explained contaminant variation for thirteen contaminant models while the FA variables were only significant in two models. If the future diet of ringed seals from Resolute Bay becomes similar to Arviat seals, findings suggest a potential for climate-induced diet-driven increases in some of the newer contaminants, the perfluoroalkyl substances (PFASs), but decreases in total mercury (THg) and legacy POPs, like dichlorodiphenyltrichloroethanes (ΣDDT), as well as in polybrominated diphenyl ether (ΣPBDE) flame retardants, although additional time trends studies are needed. Ultimately, these two studies serve to better understand contamination in ringed seals of the Canadian Arctic to protect this species of high cultural, nutritional, and economic importance.

Résumé Général

De nombreux polluants organiques persistants (POPs) ont fait l'objet d'études approfondies dans la faune arctique. Cependant, de nouveaux POPs tels que les paraffines chlorées à chaîne courte (PCCC) ont reçu beaucoup moins d'attention dans les études de surveillance des contaminants de l'Arctique. Dans le chapitre 3, j'ai quantifié les concentrations tissulaires de PCCC (C_{10} -C_{13}) chez les poissons marins arctiques, les invertébrés et les phoques annelés (*Pusa hispida*) de l'ouest de la baie d'Hudson, au Canada. De plus, j'ai évalué le potentiel de bioamplification des PCCC au sein de ce réseau trophique. Les concentrations moyennes de ΣPCCC dans ces espèces variaient de 38,3 à 687 ng g^{-1} de poids lipidique. Malgré des concentrations parfois élevées chez les poissons, les facteurs de bioamplification ajustés au niveau trophique des congénères de PCCC variaient de seulement 0,07 à 0,55 pour les petits poissons pélagiques jusqu'aux phoques annelés. Ainsi, mes résultats suggèrent un potentiel de bioamplification limité des PCCC dans ce réseau trophique.

Il est nécessaire de mieux comprendre comment les changements écologiques liés au climat peuvent influencer les concentrations de tous les principaux contaminants environnementaux de l'Arctique. Surtout pour les prédateurs comme le phoque annelé, qui accumulent généralement des concentrations plus importantes de ces contaminants. Pour mon chapitre 4, j'ai comparé l'écologie alimentaire et les concentrations de contaminants (anciens et nouveaux POPs ainsi que les métaux traces) chez les phoques annelés de deux sites arctiques canadiens (Resolute Bay et Sachs Harbour) et de deux sites subarctiques (Arviat et Nain). J'ai observé une plus grande quantité de changements d'alimentation liés au climat chez les phoques annelés aux sites subarctiques en comparaison à Resolute Bay, ce qui suggère que les conditions subarctiques actuelles pourraient nous aider à prédire les conditions futures de l'Arctique. En mesurant les acides gras contenus dans leur graisse (AG) et les isotopes stables (IS) contenus dans leur muscles (tous deux des traceurs alimentaires), j'ai noté la présence d'AG distincts entre les phoques de Resolute Bay et les deux sites subarctiques. Cette différence est probablement liée à une consommation plus élevée d'espèces envahissantes du sud dans l'environnement subarctique comparé à Resolute Bay, ou les phoques semble se nourrir plutôt d'espèces indigènes à leur environnement Arctique. Les phoques de Sachs Harbour en revanche, contenaient des AG distincts qui concordent avec les trois autres sites, suggérant un régime alimentaire mixte d'espèces indigènes et envahissantes. À l'aide de modèles linéaires généralisés, j'ai découvert que les IS expliquaient mieux la variation des contaminants pour treize modèles de contaminants, tandis que les variables d'AG n'étaient significatives que dans deux modèles. Si le futur régime alimentaire des phoques annelés de Resolute Bay devient similaire à celui des phoques d'Arviat, mes résultats suggèrent un potentiel d'augmentation de certains des nouveaux contaminants, comme les substances perfluoroalkylées (PFAS). Parallèlement, ces résultats présagent une diminution du mercure total (THg) et les anciens POPs, comme les dichlorodiphényltrichloroéthanes (ΣDDT), ainsi que les retardateurs de flamme polybromodiphényléthers (ΣPBDE). Finalement, ces deux études permettent de mieux comprendre la contamination chez les phoques annelés de l'Arctique canadien afin de protéger cette espèce d'une grande importance culturelle, nutritionnelle et économique.

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Contribution of Authors

Chapter 3: Nadia Facciola and Sara Pedro co-authored this chapter. They both analyzed the data and wrote the initial version of the manuscript. Specifically, Nadia performed all the analysis and writing related to the ringed seals and the Monte Carlo biomagnification factor for the fish and ringed seals. Nadia also wrote the introduction and addressed all the feedback from the co-authors and the journal. Sara performed the writing and SCCP analysis for the fishes and amphipods and compared those levels to legacy POP. All authors contributed to subsequent drafts of the manuscript. Melissa McKinney, Magali Houde and Arron Fisk designed the study. Melissa McKinney, Magali Houde, Steven Ferguson and Derek Muir coordinated with communities the collections of fish, invertebrate and seal samples in Western Hudson Bay, Nunavut. Helena Steer performed the SCCP instrumental analysis.

Chapter 4: Nadia Facciola is the primary author of this chapter. Nadia Facciola performed the fatty acid analysis, analyzed all the data, and wrote the initial version of the manuscript. Melissa McKinney and Magali Houde contributed to subsequent drafts of the manuscript. Nadia Facciola, Melissa McKinney and Magali Houde designed the study. Magali Houde, Steven Ferguson and Derek Muir coordinated with communities the collection of ringed seals from the Arviat, Nain, Resolute Bay and Sachs Harbour in the Canadian Arctic. Magali Houde supplied the ringed seal persistent organic pollutant data and stable isotope data. Derek Muir supplied the ringed seal heavy metal data.

List of Abbreviations

POPs: persistent organic pollutants

SCCPs: short chain chlorinated paraffins

PCBs: polychlorinated biphenyls

Σ_{10}PCB: ten major congeners of PCBs

CHL: chlordanes

HCH: hexachlorocyclohexanes

HCB : hexachlorobenzene

DDT: dichlorodiphenyltrichloroethanes

PBDE: polybrominated diphenyl ether flame retardants

PFSA: perfluorosulfonic acid

PFCA: perfluoroalkyl carboxylates

PFOS: perfluorooctane sulfonate

PFOA: perfluorooctanoic acid

Hg: mercury or THg: total mercury

Pb: lead

Cd: cadmium

SI: stable isotopes

FA: fatty acid

SFT: space for time substitution

CSIA: compound specific isotope analysis

QFASA: quantitative fatty acid signature analysis

Chapter 1: General Introduction

Persistent organic pollutants (POPs) are synthetic chemicals that are resistant to degradation, bioaccumulative, subject to long-range transport and recognized as having adverse health effects on humans, wildlife, and the environment (Stockholm Convention 2001). While the production and use of many pesticides, industrial chemicals, and chemical by-products have been controlled through international regulations such as the UN Stockholm Convention, these POPs continue to cycle in the environment due to their persistence (Riget et al. 2019). Through oceanic, fluvial, and atmospheric transport, these volatile or semi-volatile chemicals can travel far distances and be deposited into cold northern regions such as the Canadian Arctic (Lohmann et al. 2007). Once deposited in the ice, snow and water, POPs can enter the marine food web, bioaccumulate and magnify throughout the food web, often leading to top predators having the highest concentrations (Dietz et al. 2019). This is of concern because, at high concentrations, certain contaminants have known negative health effects on humans and wildlife, including neurological, reproductive, endocrine, and carcinogenic effects (AMAP 2021b).

There are two main categories of POPs. The first class are “legacy” contaminants; this includes the twelve POPs initially listed under the Stockholm Convention in 2001, such as polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethanes (DDTs). Many of these POPs have been extensively studied and monitored for several decades in Arctic wildlife (Houde et al. 2019, Riget et al. 2019). The second class are “new” contaminants; these are POPs that have only recently been added to the Stockholm Convention or recently been monitored such as short-chain chlorinated paraffins (SCCPs) and perfluoroalkyl substances (PFASs). For many of these newer POPs, their impacts on the Arctic environment are not fully understood (AMAP 2021a). For example, SCCPs show similar characteristics to legacy POPs including chemical stability, volatility, capacity to undergo long-range transport and evidence of accumulation in food webs, however, have received considerably less attention in Arctic monitoring programs to date (van Mourik et al. 2016). In addition to POPs, naturally occurring trace metals, such as mercury (Hg), cadmium (Cd), and lead (Pb) have also been documented to have high environmental levels in the Arctic, partly due to anthropogenic emissions (Fant et al. 2001, McConnell and Edwards 2008). Like POPs, these metals have known health effects, and Hg and Cd have been shown to bioaccumulate and/or biomagnify in food webs (Morel et al. 1998, Woshner et al. 2001, AMAP 2021b). Regardless of the type of contaminant (legacy, new or trace metal), a key uncertainty that has been identified is how climate change will impact the fate of these contaminants in Arctic marine life.

As a result of climate change, the Arctic environment has undergone drastic changes and is now considered to be in a new climatic state (IPCC 2021). Physical changes to the Arctic environment have led to widespread ecosystem alterations, including the northward invasion of southern species (Fossheim et al. 2015, Oziel et al. 2020). Invading forage fish can impact the prey available for native Arctic species including seabirds, whales, and seals (Chambellant 2010, Fossheim et al. 2015). Climate-induced changes in the Arctic food web may have an impact on bioaccumulating and biomagnifying contaminant concentrations in upper trophic level native Arctic species such as the ringed seal (*Pusa hispida*) (de Wit et al. 2022). As a high trophic level species of cultural, nutritional, and economic value to northern communities, ringed seals are a key biomonitoring species for contaminants and environmental change across the Canadian Arctic (Houde et al. 2019, De La Vega et al. 2021).

Thus, the research aim for this thesis is to further investigate SCCP accumulation in the ringed seal food web, and to develop insight into the effects of climate-change driven feeding shifts on legacy and new POP and trace metal concentrations in ringed seals across the Canadian Arctic. The main research aim will be addressed with the following specific objectives:

(1) To measure the concentrations of SCCPs in Arctic marine fishes, invertebrates, and ringed seals, and then compare these concentrations to those of legacy POPs and evaluate SCCP biomagnification potential from fish-to-seals.

(2) To compare the feeding patterns of ringed seals from two Arctic and two sub-Arctic locations to test the extent to which contaminant concentrations between sites are related to feeding differences across the Arctic, providing insight into the potential effects of climate-induced feeding changes on contaminant concentrations in ringed seals.

Chapter 2: Literature Review

2.1 Persistent organic pollutants and Arctic contamination

With over 350,000 chemicals globally registered and 50,000-70,000 confidential or ambiguously described, it is difficult to understand the scope of pollution and its impact on environmental, animal, and human health (Wang et al. 2020). Persistent organic pollutants (POPs) are especially of concern for the environment as POPs are persistent, bioaccumulative, toxic and can be transported long distances by atmospheric, oceanic, and riverine transport (Lohmann et al. 2007). In the early 2000s, the UN Stockholm Convention on Persistent Organic Pollutants entered into force initially listing twelve "legacy" POPs that were recognized for causing adverse effects on humans and the ecosystem (Stockholm Convention 2001). This includes different types of pesticides, industrial chemicals, and chemical by-products such as polychlorinated biphenyls (PCBs) and organochlorine (OC) pesticides including dichlorodiphenyltrichloroethanes (DDTs), chlordanes (CHL) and hexachlorobenzene (HCB). Despite most of these POPs being listed under Annex A (Elimination of production and chemical use), they remain within the environment due to their chemical persistence. "Newer" POPs are chemicals that have only recently been studied or listed under the Stockholm Convention. While many of these chemicals have been in use for decades, their fate within the environment and biota is not yet fully understood (AMAP 2021a). This includes short-chain chlorinated paraffins (SCCPs), an additive used in adhesives, paints, leathers, and conveyor belts, which have only recently been added to the Stockholm Convention (Stockholm Convention 2017). SCCPs have previously received noticeably less attention in contaminant monitoring studies, in part due to difficulties with SCCP quantification and chemical complexity (van Mourik et al. 2016).

Through the atmospheric, oceanic, and riverine long-range transport of POPs, the Canadian Arctic receives contaminants from further south (Dietz et al. 2019). Certain communities and wildlife are among the most exposed populations on a global basis, despite being far from most POP sources (AMAP 1998, 2016). Once in the Arctic, POPs get deposited into the snow, ice, and water where they can enter into marine food webs and biomagnify and bioaccumulate through the food web to mammalian predators, including the ringed seal (*Pusa hispida*), the polar bear (*Ursus maritimus*) and northern community members (Lohmann et al. 2007, Braune et al. 2015). This is of particular concern as at high enough concentrations certain contaminants have known or suspected neurological, reproductive, immunological, endocrine, and carcinogenic effects on humans and wildlife (Dietz et al. 2019, AMAP 2021b). SCCP toxicity studies have documented effects on endocrine, reproductive, and immune systems in rats, mice and fish (Wyatt et al. 1993, Liu et al. 2016). However, no toxicity studies have been done on Arctic wildlife and very few studies have reported SCCP levels in Arctic marine mammals, seabirds or marine fish (Vorkamp et al. 2019). Therefore, to understand the potential impacts of SCCPs in the Canadian Arctic, SCCP levels in marine fish and marine mammals need to be further investigated.

Marine mammal predators have been documented with high concentrations of certain POPs and trace metals for several reasons. Due to their long lifespan, marine mammals are exposed to many contaminants throughout their lifetime, resulting in increased concentrations of certain POPs with age (Letcher et al. 2010). Most marine mammals also occupy relatively high trophic positions within the food web and have consequently been documented to show high concentrations of biomagnifying contaminants including legacy POPs such as PCBs, OC pesticides, newer POPs such as PFAS and trace metals such as mercury (Hg) (McKinney et al.

2012, Fair and Houde 2018, Remili et al. 2021). Lipophilic POP accumulation often occurs in the thick layers of fatty storage tissues (blubber) and protein binding contaminants (such as trace metals and perfluoroalkyl substances (PFAS)) in the blood and liver of marine mammals. Some marine mammals including ringed seals and polar bears can metabolize some POPs, while others such as cetaceans show limited biotransformation capacity toward POPs (Wolkers et al. 2007, Letcher et al. 2010). Regardless, all of these Arctic marine mammals commonly transfer contaminants through their placenta and milk to their offspring (Addison and Brodie 1977).

A recent assessment of temporal trends of POP found a general decrease in legacy POPs in Arctic marine and freshwater biota throughout the Arctic (Riget et al. 2019). The reduction in primary emissions of POPs through international regulations, such as the Stockholm Convention, is currently the main driver of temporal POP trends (de Wit et al. 2022). However, increasing trends have been found for some “newer” POPs in marine mammals, including polybrominated diphenyl ether (PBDE) flame retardants (Addison et al. 2020), per- and polyfluoroalkyl substances (PFAS), such as perfluoroalkyl carboxylates (PFCAs) (Muir et al. 2019), and SCCPs (Zeng et al. 2015). National long-term monitoring programs such as the Northern Contaminants Program (Crown-Indigenous Relations and Northern Affairs Canada) and international programs like the Arctic Monitoring and Assessment Programme (AMAP) are therefore essential in order to draw connections with regulations, production of current-use pesticides, legacy and newer POPs, and climate change (Houde et al. 2019, Borgå et al. 2022, McKinney et al. 2022). Understanding current POP trends in the Arctic environment and wildlife is crucial in order to provide contaminant information to northern communities about their wild and traditional foods and it is through the collaboration and involvement of Arctic Indigenous communities that these long-term monitoring programs are possible (Houde et al. 2019).

2.2 Influence of climate change on contaminants in the Arctic

The distribution and fate of contaminants within the Arctic may be altered by climate-related physical and ecological changes (McKinney et al. 2015, AMAP 2016, Rigét et al. 2020, de Wit et al. 2022). The effects of climate change in the Arctic are exceptionally profound, leading to a warming rate two to three times faster than the global average (Serreze and Barry 2011, IPCC 2021). This has resulted in the loss of vast amounts of sea ice, creating new climate characteristics and altered habitat (Stroeve et al. 2007, Post et al. 2013). Over the past decade, there has been an increase in studies documenting the effects of climate change on the fate of Arctic contaminants (de Wit et al. 2022). Diet changes linked to lower sea ice have been associated with higher levels of contaminants in native Arctic species including the polar bear, ringed seal and thick-billed murre (McKinney et al. 2015). Secondary sources of contaminants such as those from environmental stores found in soil, snow and ice are expected to be released as the climate warms leading to the remobilization and distribution of these POPs throughout the Arctic (Borgå et al. 2022, Chételat et al. 2022, McKinney et al. 2022). In addition to climate-related changes in the Arctic itself, environmental changes outside the Arctic can also affect the fate of contaminants within the Arctic. Depending on the chemical-physical properties of certain contaminants, it is predicted that the increasing global temperature will increase the volatility of certain POPs, impacting the transport of these contaminants to the Arctic (Ma et al. 2011). Increasing levels of certain POPs have been associated with their release and volatilization from melting sea ice (Hung et al. 2022). Other direct influences of climate change include changes in ocean currents, extreme weather events, changes in carbon cycling, sea-level rise and ocean acidification, which can affect contaminant processes such as long-range transportation and the biotic and abiotic transformation of contaminants (AMAP 2021a). For example, it has been documented that wildfires in Canada and Russia have been connected to elevated concentrations of polycyclic aromatic hydrocarbons (PAHs) and PCBs in the Arctic and increased water discharge from Eurasia as a result of extreme weather events has increased PAH concentrations in the Arctic Ocean (Hung et al. 2022).

Climate-related habitat changes can also shift species distributions, resource availability, predator-prey interactions, and subsequent food web structures (Wassmann et al. 2011, McKinney et al. 2013, Pecl et al. 2017). Since POP exposure in marine biota often occurs through diet and due to the biomagnification and bioaccumulation characteristics of POPs, these food web changes may impact contaminant levels in Arctic biota (Pedro et al. 2017, AMAP 2021a, Florko et al. 2021). Compared to the rest of the globe, the marine Arctic ecosystem is projected to have the highest rates of species turnover and invasion of non-Arctic species (Cheung et al. 2009). As a result of warming temperatures, patterns of northward range expansion of southern species, a phenomenon called “borealization”, has already been detailed in marine food webs from temperate and sub-Arctic regions (Fossheim et al. 2015). In particular, temperate capelin (*Mallotus villosus*) and sand lance (*Ammodytes*) have been observed entering the sub-Arctic and Arctic Canada, acting as a newly available prey for species such as the ringed seal and possibly replacing other keystone Arctic fish such as Arctic cod (*Boreogadus saida*) (Provencher et al. 2012, Fossheim et al. 2015, Falardeau et al. 2017). A recent study modelling how climate change will impact the base of Arctic marine predators projects a climate-driven decline of 50% of Arctic cod, and an increase of smaller temperate-associated fish by 2100, under a high greenhouse gas emissions scenario (Florko et al. 2021). A study comparing contaminant levels in native and invading species in the Canadian Arctic found that the invading capelin and sand lance (*Ammodytes spp.)* had slightly higher concentrations of most POPs compared to the native Arctic cod (*Boreogadus saida*), potentially acting as a biovector of contaminants for the Arctic marine food web (Pedro et al. 2017). Diet shifts of Arctic predators towards consumption of non-native species have been documented in many Arctic species such as the thick-billed murre (*Uria lomvia*) where the proportion of capelin in their diet has been increasing over a span of more than three decades (Gaston and Elliott 2014). Stomach contents of belugas (*Delphinapterus leucas*) have also revealed a diet primarily of sand lance with low levels of Arctic cod (Loseto et al. 2018). All of these recent studies combined have contributed to understanding the effects of climate change on contaminant levels marine biota and Arctic marine food webs. However, the main conclusion in most of these studies has been that there remain too many unknown variables among the Arctic ecosystem and climate change to fully understand and estimate the complex net effect of climate change on POP (and Hg) concentrations (Borgå et al. 2022, McKinney et al. 2022).

2.3 Ringed seals as a bioindicator for contaminants and climate change

While it is challenging to assess interactions between climate change and contaminants in the Arctic, one successful approach has been to monitor the changes in the diets of upper trophic level consumers, which can integrate environmental and food web changes (McKinney et al. 2013). One high trophic level species used as a bioindicator for contaminants and climate change throughout the Arctic is the ringed seal, the most abundant Arctic pinniped in its circumpolar Arctic range (Houde et al. 2017, Houde et al. 2019). The ringed seal is a sea ice-dependent species with its survival and reproductive success directly linked to sea ice characteristics (Smith 1987). Females require sufficient sea ice quality and snow depths to construct lairs in preparation for their single pup (Lydersen and Gjertz 1986). Females use this lair when they give birth, for lactation, and for the protection of their young from predators and the cold (Lydersen and Gjertz 1986). Their circumpolar range, distribution, abundance, and dependence on sea ice make ringed seals ideal bioindicators for climate-related environmental change.

Some effects of climate change on ringed seals have been documented, which include impacts on ringed seal health, contaminant loads, and feeding behaviour. Due to declines in sea ice over the past three decades, ringed seals sampled in Hudson Bay from 2003 to 2013 were documented to have lower body conditions, increased stress levels, and lower ovulation and pregnancy rates (Ferguson et al. 2017). Associations between climate parameters and POP levels have also been documented. A positive correlation between ringed seal POP concentrations and the Arctic Oscillation index has recently been detailed (Houde et al. 2019). In years with earlier sea ice break-up, specific legacy organochlorines have been reported at higher concentrations in ringed seals from the western Canadian Arctic, potentially related to changes in seal foraging opportunities (Gaden et al. 2012). The opposite has also been found, where seals from multiple Canadian Arctic sites were reported to have higher contaminant loads in years with greater sea ice, possibly related to the sea ice facilitating the delivery of POPs (Houde et al. 2019). In ringed seals from Greenland, positive associations have also been found between blubber ΣDDT and ΣPCB concentrations and temperature and salinity (Rigét et al. 2020). Temporal variation in THg accumulation in ringed seals in the Canadian Arctic has been explained by age, climate, and diet variables (Houde et al. 2020). Ringed seals in the western high Arctic have been traditionally documented consuming mostly Arctic cod and invertebrates (Smith 1987). However, in recent years (2015-2018), several sub-Arctic fish species including capelin and sand lance have been found by stomach content analysis (Insley et al. 2021).

Ringed seals are of high cultural, nutritional, and economic value to northern communities as they provide high-quality food and clothing. The Northern Contaminants Program has provided much information on spatial and temporal contaminant patterns for ringed seals (Muir et al. 2000, Houde et al. 2019, Houde et al. 2020). Between 1972 and 2016, concentrations of an extensive number of POPs have been documented in ringed seal blubber across the Canadian Arctic reporting a general slow decrease of most legacy POPs over time (Houde et al. 2019). The spatial trends of this study showed that concentrations of PCBs, DDT, HCH, CBz (chlorobenzene) and toxaphene were dominant in the ringed seals from all sites across the Arctic. Concentrations of DDT, CHL and HCH were found to be significantly lower in Arviat (Western Hudson Bay) compared to other locations such as Sachs Harbour (Beaufort Sea). Concentrations of PCBs, toxaphene and HCHs, were not significantly different between Resolute Bay (Arctic Archipelago), Arviat and Nain (Labrador). Site differences in contaminant levels can be related to many factors including the modes of transport of the contaminants into the Arctic. This includes the influence of different water bodies (Pacific vs Atlantic), atmospheric currents and the sites proximity to industrialization, airports, or large populations (Houde et al. 2019). While POPs are dominant in ringed seal blubber, the adverse effects of ringed seal POP exposure have been studied and are suggested to be relatively low (AMAP 2018). However, it remains important to assess climate-related contaminant changes in ringed seals to monitor contaminant dynamics under climate change, and also as polar bears, their cubs, and northerners consuming seal meat could face adverse effects from the contaminants in ringed seals (AMAP 2018, Houde et al. 2019). To date, no Canadian Arctic ringed seal studies have linked spatial contaminant trends with spatial feeding differences to gain a better understanding of the potential effects of climate-induced feeding changes on seal contaminant loads.

2.4 Fatty acids and stable isotopes as dietary tracers

Understanding an animal’s diet is essential for studying food-web dynamics, predator-prey interactions, and contaminant accumulation and can ultimately increase our understanding of the health of wildlife and ecosystems (Bowen and Iverson 2013). There are a variety of direct and indirect methods used to assess wildlife diets. Direct observation of an individual’s feeding activity is a non-invasive method to estimate diet (Pierce and Boyle 1991). This provides information about the last meal of the individual, however, for many marine species, this method is not ideal as there are few opportunities to directly observe feeding that occurs underwater and/or in remote, inaccessible environments (Pierce and Boyle 1991). Stomach content analysis and fecal analysis are two indirect methods that provide the dietary history of perhaps the last few meals eaten (Bowen and Iverson 2013). These two methods are relatively inexpensive and can provide large sample sizes of the consumed prey, however, diet information is limited to prey with hard parts that resist digestion, leading to a bias caused by the complete digestion of soft prey, and stomach contents can only be obtained from deceased individuals (Bowen and Iverson 2013). DNA-based diet analysis can also provide information about the last few meals consumed by the individual, and this approach can identify prey with high accuracy (Bowen and Iverson 2013). However, it can be difficult to isolate the DNA, requires species-specific primers and currently can only identify the presence of prey and not the quantity consumed (Nielsen et al. 2018). Two indirect methods that can provide dietary history over a larger period are fatty acid (FA) and stable isotope (SI) analysis. FA analysis can provide a feeding history anywhere between days and months depending on the species (Budge et al. 2006). However, the individual’s feeding location is less defined due to the long integration time of the diet. Stable isotopes can provide diet information spanning from days to even years depending on the tissue being sampled (Bowen and Iverson 2013). SI analysis can also provide information about the trophic position of the individual and whether they are consuming prey from benthic/in-shore or pelagic/offshore regions. However, SI analysis cannot provide exact prey species composition for complex diets consisting of many prey items and isotopic baselines have been documented to vary across the Arctic (De La Vega et al. 2019, De La Vega et al. 2021). While each of these diet analysis methods has its advantages and disadvantages, combining dietary estimation methods that complement each other, such as FA and SI analysis, has been successfully used to investigate ringed seal feeding ecology variation in the Canadian Arctic (Ross et al. 2022).

FAs are the main constituents of most lipids. They often consist of a straight carbon chain (4 to 24 length) with a terminal methyl group, a methylene group or groups (if mono or polyunsaturated), and an acid group (Budge et al. 2006). FA signatures represent a useful diet analysis method because many of the FAs consumed by monogastric predators, including the ringed seal, get deposited into the blubber tissue of the mammal with little or predictable modification (Budge et al. 2006). That is, the dietary FA signatures in the predator reflect the FA signatures in the prey that was consumed. While other tissues can contain FAs, it is the adipose tissue (blubber) in marine mammals that is the principal site for FA energy storage (Budge et al. 2006). There are two main ways FA analysis can provide information about diet. The first approach is analyzing the FA signatures of the predator. By comparing FA ratios between individuals of a population, this qualitative approach can provide information about the spatial or temporal variation of dietary patterns. This method has been used to make inferences about dietary differences for many Canadian Arctic species including bearded seals (*Erignathus barbatus*), ringed seals, beluga whales, narwhals (*Monodon monoceros*), polar bears, and many marine fishes (Thiemann et al. 2008a, b, Pedro et al. 2017). The second approach, quantitative

FA signature analysis (QFASA), is a mixed model approach to estimate the proportions of prey consumed in the predator's diet. This requires a substantial database with the FA signatures of potential prey items and a correction coefficient to account for the predator metabolism (Bowen and Iverson 2013). This approach has been successfully validated and used for various seabirds, pinnipeds and mink (Iverson 2009). One shortcoming with QFASA is that the appropriate species must be included in the prey database and because it is not possible to sample the whole ecosystem, species that are rare in the diet may not be included (Iverson et al. 2004). Prey species must also have FA patterns that are distinguishable from each other (Budge et al. 2002).

Bulk stable isotope values such as nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) have been used as indicators of diet, food web, and foraging behaviour in many marine Arctic species (McKinney et al. 2015, Pedro et al. 2020, McKinney et al. 2022, Ross et al. 2022). In the marine environment, the ratio of nitrogen stable isotopes can be used to estimate the trophic level of the consumer as $\delta^{15}N$ increases by approximately 3‰ for each trophic level (DeNiro and Epstein 1981, Post 2002). Whereas the ratio of carbon stable isotopes has little enrichment with increasing trophic level, $\delta^{13}C$ is instead used to understand the carbon source and foraging habits of the consumer (Post 2002). Relatively depleted $\delta^{13}C$ levels can suggest that the consumer is foraging for food in offshore or pelagic areas (a phytoplankton carbon pathway) and enriched $\delta^{13}C$ levels suggest the opposite, that the consumer is getting energy from inshore or benthic sources (an algae and detritus carbon pathway) (Post 2002). Carbon stable isotopes originating from sea ice algae can be up to 7‰ more enriched than from pelagic phytoplankton (De La Vega et al. 2019). There are known baseline differences of $\delta^{15}N$ and $\delta^{13}C$ values throughout the Arctic, an observation which has implications when comparing $\delta^{15}N$ and $\delta^{13}C$ values spatially or temporally (De La Vega et al. 2019, De La Vega et al. 2021). Environmental factors such as sea ice extent, freshwater inputs, and changes in primary productivity can influence δ^{13}C levels at the base of the food web (De La Vega et al. 2019). Per year, δ^{13}C declines by about 0.011‰ due to a phenomenon called the Suess effect which is caused by increasing anthropogenic carbon dioxide in the Arctic ocean (De La Vega et al. 2019). Bulk δ^{15}N values are also influenced by the base of the food web, making it difficult to disentangle the baseline influence from the trophic level of the consumer using δ^{15}N approaches (De La Vega et al. 2021). One solution to account for baseline differences in δ^{15}N is to use compound-specific stable isotope analysis (CSIA) of nitrogen amino acids ($\delta^{15}N_{AA}$). The source of $\delta^{15}N_{AA}$ undergoes very little fractionation with trophic transfer and can thus be used to trace the baseline of δ^{15}N (McMahon and McCarthy 2016). CSIA of fatty acids can also provide valuable information about isotopic variation among individual FA compounds that may not be apparent when using only bulk isotope approaches (Twining et al. 2020).

2.5 Space for time substitution

There are a variety of methods that have been used to investigate the effects of climate change on ecosystems. One method that has been particularly useful when temporal datasets are unavailable, or not possible to obtain (i.e, past or future events) is “space for time substitution” (SFT) (Pickett 1989). This is a method that uses spatial differences/phenomena in order to understand temporal processes. This can include studies that use cross-comparisons, and latitudinal or altitudinal gradients to compare ecosystems under different climatic environments in order to explain the long-term potential impacts of climate change (Meerhoff et al. 2012). Historically, this approach has been used by scientists for centuries, including Darwin to study the development of atolls, fringing and barrier reefs, and von Humboldt and Bonpland who used temperature and precipitation gradients to study vegetation distributions in South America in the 1800s (Meerhoff et al. 2012, Damgaard 2019). SFT has been widely used in current research as well, such as ecology, biodiversity, climate change, and species invasion studies (Thomaz et al. 2012). This is a particularly useful method to help predict whether a native species will be able to resist species invasion (Levine et al. 2004).

Comparing non-invaded sites to invaded sites is an SFT approach that can infer the effects of species invasions on the diversity of native species (Thomaz et al. 2012). Although the process of species invasions is complex, the potential geographical course of an invasion can be predicted based on the niche characteristics of the invading species (Peterson 2003). This approach has been used extensively in plants, for example, Hejda et al. (2009) compared the vegetation of vascular plants in invaded and uninvaded areas with similar conditions and found that species invasion negatively impacted native species richness, diversity, and evenness. Positive effects of species invasions have also been documented using the SFT approach. Barrientos and Allen (2008) found that invading macrophyte vegetation increases fish habitat and overall fish biomass compared to non-invading sites. While the SFT method is useful for predicting invasion patterns, using this approach in conjunction with data from pre- and post-invasion sites is necessary to provide accurate conclusions (Thomaz et al. 2012).

The sub-Arctic and Arctic environments can be useful systems to model climatic changes using SFT. Particularly because climate change is occurring at a faster rate within the Arctic compared to the global average, there is relatively little land-use change and Arctic biodiversity is relatively low, making it vulnerable to species invasions (Kutz et al. 2009). By comparing the population abundance of mussels in sub-Arctic and Arctic regions of west Greenland, Thyrring et al. (2017) found that increased climatic warming will enable an increase in mussel abundance in Greenland. In addition to using the SFT approach for species invasions, it can also be used to model contaminant concentration differences. Hallanger et al. (2011) used this approach to model contaminant bioaccumulation in a future borealized Arctic food web. This was done by comparing zooplankton species between Arctic and Atlantic fjord systems from Greenland. Findings showed that zooplankton in the Atlantic had higher POP concentrations and bioaccumulating factors compared to zooplankton in the Arctic (Hallanger et al. 2011). However, confounding factors such as differences in ice cover and snowmelt made it difficult to conclude possible climate-change effects.

While SFT can be a cost-effective and useful approach to studying long-term ecological processes, it can sometimes lead to erroneous conclusions (Damgaard 2019). Powell et al. (2011) found that the geographical spatial scale being compared can have an influence on the results. In addition, the main assumption in SFT is that drivers influencing spatial gradients also influence temporal changes (Blois et al. 2013). This can be problematic as factors that incorporate temporal variability are ignored, for example, species and site history, species interactions, factors interacting with climate change and the temporal variability of climate change (Damgaard 2019). It can therefore be difficult to disentangle these factors when comparing two spatial locations based solely on their climate differences. With this in mind, Blois et al. (2013) tested this assumption by comparing temporal turnover rates using predictions from the SFT method and the time-for-time method. They found that predictions using SFT were ~72% as accurate as predictions from the time-for-time approach (Blois et al. 2013). Damgaard (2019) also reported that it may be possible to combine spatial variation data with sparse time-series data to help further understand ecological processes. While SFT results should be interpreted carefully, it remains advantageous to use when temporal datasets are unavailable or sparse. This approach provides a starting point for highlighting the potential effects of climate change on ecosystems.

Connecting Text

All research objectives aimed at developing insight into the impacts of short-chain chlorinated paraffins on Arctic marine wildlife were addressed within Chapter 3. This manuscript “Measurable levels of short-chain chlorinated paraffins (SCCPs) in western Hudson Bay fishes, but limited biomagnification from fish to ringed seals”, is co-first authored by Nadia Facciola and Sara Pedro, with written consent from Sara Pedro to use the manuscript in this thesis. This manuscript was accepted and published in the journal of *Environmental Toxicology and Chemistry* on August 4th, 2021. Copyright approval was received from John Wiley and Sons to use the full manuscript in this thesis. Copyright license number 5303700273368.

Chapter 3: Measurable levels of short-chain chlorinated paraffins (SCCPs) in western Hudson Bay fishes, but limited biomagnification from fish to ringed seals

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3.1 Abstract

We investigated short-chain ($C_{10–13}$) chlorinated paraffins (SCCP) in an Arctic marine food web. In zooplankton, fishes, and ringed seals from western Hudson Bay, Canada, SCCP concentrations ranged from 38.3 to 687ng g^{-1} lipid weight. Monte Carlo–simulated trophic-adjusted biomagnification factors of individual SCCP congeners ranged from 0.07 to 0.55 for small pelagic fishes to seals. Despite relatively high concentrations in fishes, biomagnification of SCCPs within this food web appears limited.

Keywords: Short-chain chlorinated alkanes; Fish; Pinnipeds; Biomagnification; Arctic; GC Orbitrap MS

3.2 Introduction

Short-chain chlorinated paraffins (SCCPs) are chlorinated alkanes with a chain length of 10 to 13 carbons and chlorine content varying between 48% to 70%. Since the 1930s, SCCPs have been extensively used in industry (Fiedler 2010) with a global production of at least 165,000 tons/year (Glüge et al. 2016). Similar to legacy persistent organic pollutants (POPs) like polychlorinated biphenyls (PCBs), SCCPs show chemical stability and volatility leading to long-range transport, with some evidence of accumulation and toxicity in biota (Van Mourik et al. 2016). Consequently, they were listed under the Stockholm Convention on POPs in 2017, with some production exemptions (Stockholm Convention 2017). Given these properties, SCCP exposures and risks to biota should continue to be monitored.

SCCPs have received considerably less attention in Arctic contaminant monitoring studies compared to other POPs, partly because their quantification is still challenging due to lack of suitable standards, low analytical detection and chemical complexity (Van Mourik et al. 2016). Only a handful of studies on Arctic biota have reported SCCP levels in marine mammals and seabirds, and fewer still have monitored levels in marine fish (Vorkamp et al. 2019). SCCPs may biomagnify in marine food webs, similar to some of the most persistent POPs (Harju et al. 2013; Yuan et al. 2019). Although no SCCP toxicity studies have been done on Arctic biota, research on rats, mice and fish have shown effects on endocrine, reproductive, and immune functions (Wyatt et al. 1993; Cooley et al. 2001; Liu et al. 2016).

The objectives of the present study were to 1) quantify tissue SCCP concentrations in marine fish, invertebrates and ringed seals (*Pusa hispida*) from an Arctic food web, 2) compare SCCP levels with those of other legacy POPs in the same samples (Pedro et al. 2017), and 3) evaluate the SCCP biomagnification by calculating biomagnification factors (BMFs) from fishes to seals.

3.3 Materials and Methods

Five marine fish species and amphipods were collected as previously described (Pedro et al. 2017). In brief, capelin (*Mallotus villosus*), sand lance (*Ammodytes* spp.), cisco (*Coregonus artedi*), sculpin (*Myoxocephalus spp.*), Greenland cod (*Gadus ogac*), and amphipods (*Gammarus oceanicus*) were collected by community members near Arviat (low Arctic, 61° N, 94° W), Nunavut, Canada (Figure 3.1) during the summer of 2014. Collection methods included rod and reel, ice fishing, and grabbing from shore (Pedro et al. 2017). Collections consisted mostly of adult fish (Table 1), except for capelin, which were likely juveniles as they were below the 130-200 mm maturity size for this species (Hop and Gjøsaeter 2013; Ogloff et al. 2020). Fish and invertebrate samples were wrapped in solvent-rinsed foil and stored at -20 °C temporarily, and then kept at -80 °C in the lab. Fish species identifications were confirmed genetically at the Canadian Center for DNA Barcoding.

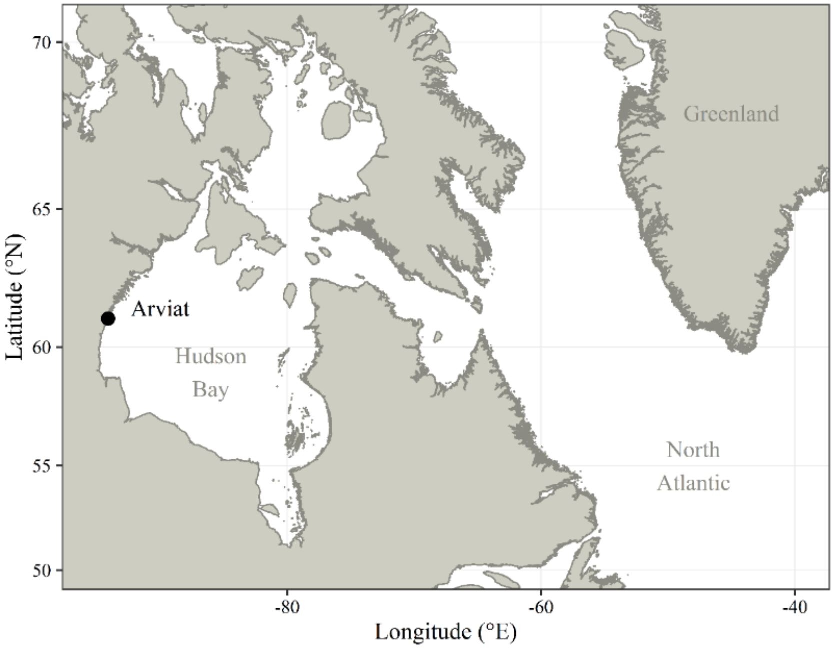

Figure 3.1 Sampling location of fish, and amphipods (2014) and ringed seal (2017) in Western Hudson Bay, Nunavut, Canada.

Ringed seal blubber samples were collected by community members during subsistence hunts around Arviat in the fall of 2017, including nine females (<1 to 21-yrs old), and one male (1-yr old). Collected blubber samples were placed in Whirl-Pak® bags and stored between -15°C and -25°C within hours of sampling and then kept at -20°C (Houde et al. 2019). Age was determined by Matson's Laboratory (Milltown, MT, U.S.A) by counting the annual growth layers of longitudinal sections of teeth (Houde et al. 2019). Seals ≤1 year-old were considered young of the year and >5 were considered adults; no animals between 2 and 7 years-old were collected. Due to the environmental persistence of SCCPs, evidence that SCCPs have been successfully analyzed in a soil core between 1950 to 2004 (Iozza et al. 2008), and the storage of the fish and invertebrate samples at -80°C, substantial SCCP degradation between the two collection dates (2014 and 2017), or between collection and analysis, is unlikely.

SCCP Analysis

Fish and invertebrate contaminant extractions were carried out at the Great Lakes Institute for Environmental Research, University of Windsor. These samples were originally extracted and analyzed for PCBs, organochlorine (OC) pesticides and brominated flame retardants (Pedro et al. 2017). Briefly, muscle tissues of individual fish and whole amphipods were homogenized prior to analysis. Eight capelin and all sand lance did not have sufficient mass for individual analysis, thus individuals of each species were pooled (capelin in pairs and sand lance in quadruplets) by combining equivalent mass aliquots of each homogenate. Aliquots of 2.5 g were homogenized with anhydrous sodium sulfate and eluted with 1:1 dichloromethane:hexane. Samples were subject to gel permeation chromatography to remove lipids, and lipid content was quantified gravimetrically. Samples were further cleaned-up and fractionated on a Florisil column into three fractions, which contained different organic contaminant groups (see Pedro et al. 2017).

Seal blubber samples were extracted for SCCPs at the Québec Laboratory for Environmental Testing (Environment and Climate Change Canada) in Montreal, Québec. Samples weighing 1 g were homogenized with diatomaceous earth (J.T. Baker, Phillipsburg, NJ, USA) and spiked with 100 µL of a 200 ng/mL internal standard solution (BDE-30, BDE-156, ^{13}C-BDE-209, and ^{13}C-*anti*-DP) (Simond et al. 2017; Simond et al. 2020). Samples were extracted with 1:1 dichloromethane:hexane on a pressurized liquid extraction system. Sample clean-up was performed on an acid-basic-neutral silica column, followed by a neutral alumina column. Lipid content was quantified gravimetrically.

Fish, invertebrate and seal samples extracts were analyzed for SCCPs at the National Laboratory for Environmental Testing (Environment and Climate Change Canada, Burlington, ON). For fish and invertebrates, SCCPs were not detected in fraction 1, so fractions 2 and 3 were combined for analysis. A range of SCCP congeners were examined: $C_{10}Cl_{5-10}$, $C_{11}Cl_{5-10}$, $C_{12}Cl_{5-10}$, and $C_{13}Cl_{5-10}$. Methods were adapted from Tomy et al. (1997). SCCPs were quantified using Q Exactive GC Orbitrap GC-MS/MS (Thermo Fisher Scientific, Mississauga, ON, Canada) with a TraceGOLD TG-5SilMS GC column (30 m× 0.25mm× 0.25 µm; Thermo Fisher Scientific) operated in negative chemical ionization mode at a mass resolution of 60,000. Specific m/z values corresponding to the molecular formulas of $[M - Cl]^-$ ions of all major C_{10}-C_{13} formula groups were monitored concurrently. Corrections were made for the fractional abundance of specific m/z values and number of Cl atoms. Quantification was performed by comparing the response of specific m/z values in the sample to that of an authentic standard, which was a C_{10}-C_{13} technical mixture containing 55.5% Cl (DRE-X23105500CY; LGC Standards).

For fish and invertebrates, reagent blanks were run at the start and every four to five samples. Some blanks showed concentrations (mean 0.04, range 0-0.87 ng mL^{-1}) higher than the detection limit of 0.27 ng mL^{-1}, so we blank subtracted the results on a batch-by-batch basis. Recovery of the internal standard spikes, PCB34 and BDE71 were 97 ± 10% and 99 ± 10%, as we reported previously (Pedro et al 2017). PCB and PBDE internal standards have previously been used as surrogates for SCCP recoveries (Houde et al. 2008; Ismail et al. 2009). Although no reference material exists for SCCPs, an in-house carp reference material (Table S3.2) was extracted alongside the fish and invertebrate samples; for legacy POPs, quantification showed concentrations within 9 ± 7% for ΣPCB and 15 ± 9% for ΣOC. For seals, $^{13}C_{10}$-1,5,5,6,6,10-hexachlorodecane surrogates were added to all samples during extraction, two blanks were run for the ten samples and standard reference material NIST 1946 was run twice for the ten samples (Table S3.3). All samples were blank corrected as the blanks were above the detection limit of

0.27 ng mL^{-1}. Spike recovery of $^{13}C_{10}$-1,5,5,6,6,10-hexachlorodecane ranged from 67.7 to 125%. Although the NIST 1946 was not analyzed for PCBs and OCs, for which certified concentrations are available, for the two NIST 1946 runs, the ΣSCCP concentrations were 355 ng g^{-1} and 279 ng g^{-1}, which is a percent difference of 24%. Results were corrected for weight and lipid content for all fish, invertebrate and seal samples and are presented in ng g^{-1} lipid weight (lw).

Stable isotope analyses

Extraction, quantification and quality control details for nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotopes analysis in fish and invertebrates were described previously (Pedro et al. 2017). Briefly, homogenized and lipid-extracted subsamples of 400-600 µg tissue were combusted in an elemental combustion system. After that, ratios of $\delta^{15}N$ and $\delta^{13}C$ were quantified with a coupled Delta V Advantage isotope ratio mass spectrometer (Thermo Scientific).

For seals, stable isotopes were analyzed in non-lipid extracted muscle by the University of Waterloo’s Environmental Isotope Lab. Results have been found to be comparable to lipid-extracted analyses with differences averaging 0.28 ‰ for $\delta^{13}C$ and 0.25 ‰ for $\delta^{15}N$ (see Supplementary Material for more details) (Houde et al. 2017). Stable isotope ratios were analyzed by continuous flow isotope ratio mass spectrometry (Finnigan MAT DeltaPlus; Thermo Finnigan).

Table 3.1 Biological, ecological, and contaminant measurements (mean (±SE) and range) in prey fish, invertebrates and ringed seals from 2014 and 2017 in Western Hudson Bay [a]

	Amphipods	Capelin	Sand lance	Sculpin	Cisco	Greenland cod	Ringed seal *Pusa hispida*	
	Gammarus oceanicus	*Mallotus villosus*	*Ammodytes* spp.	*Myoxocephalus* spp.	*Coregonus artedi*	*Gadus ogac*	*Adults*	*Young of the year*
n	1 pool	10	12	6	4	5	4	6
Length (mm)	NA	103 ± 3.32 (86-123)	96 ± 2.74 (84-109)	197 ± 16.6 (129-230)	352 ± 7.8 (331-378)	305 ± 24.0 (220-440)	1242.1 ± 69.5 (1160-1450)	985.0 ± 77.6 (790-1350)
Lipid %	0.75	1.8 ± 0.23 (1.3-3.2)	1.5 ± 0.06 (1.2-1.7)	0.8 ± 0.16 (0.2-1.3)	5.2 ± 1.2 (1.6-12.1)	0.6 ± 0.04 (0.5-0.8)	87 ± 1.5 (84-90)	89 ± 1.6 (83-93)
δ^{15}N ‰	10.0	14.7 ± 0.15 (13.9-15.3)	14.2 ± 0.22 (12.7-15.8)	15.7 ± 0.47 (14.4-14.5)	13.4 ± 0.35 (12.6-14.0)	18.0 ± 0.26 (17.1-19.7)	15.8 ± 0.35 (15.2-16.8)	14.5 ± 0.25 (13.9-15.6)
δ^{13}C ‰	-16.5	-21.1 ± 0.28 (-23.1- -20.0)	-21.6 ± 0.26 (-23.2- -19.6)	-16.6 ± 0.82 (-18.9--14.2)	-18.2 ± 1.08 (-16.6- -21.7)	-18.9 ± 0.16 (-19.5- -18.1)	-21.8 ± 0.55 (-23.4- -21.0)	-21.6 ± 0.41 (-23.3- -20.7)
Trophic position	2.46	3.67 ± 0.04 (3.48-3.85)	3.56 ± 0.05 (3.15-3.98)	3.98 ± 0.10 (3.62-4.41)	3.35 ± 0.08 (3.13-3.51)	4.53 ± 0.09 (4.34-4.70)	3.96 ± 0.09 (3.82-4.23)	3.64 ± 0.65 (3.47-3.91)
ΣSCCP	559	514 ± 49 (187-693)	332 ± 42 (220-528)	244 ± 29 (133-314)	345 ± 182 (86-885)	687 ± 117 (519-1,148)	38.3 ± 6.2 (28.1- 55.3)	59.9 ± 11.4 (28.9-105)
ΣPCB	510	138 ± 14.1 (83-218)	113 ± 10.3 (70-158)	234 ± 47.4 (117-437)	80.5 ± 15.7 (50.7-140)	226 ± 21.9 (118-301)	NA	NA
ΣOC[b]	662	241 ± 15.7 (158-317)	166 ± 13.1 (106-216)	345 ± 77.2 (131-695)	116 ± 18.4 (60.7-173)	225 ± 31.2 (117-368)	NA	NA

[a] Concentrations (nanograms per gram lipid wt) of short-chained chlorinated paraffins in prey fish, invertebrates and ringed seals. Ringed seal adults ranged from 8-21 years old; young of the year seals were ≤ 1 year old.

Σ_{40}polychlorinated biphenyl (ΣPCB), Σorganochlorine pesticides (ΣOC) concentrations for prey fish and invertebrate species only. PCB and OC data were taken from Pedro et al. (2017). Length measurements indicated refer to standard length. Results are denoted as arithmetic mean (min-max).

[b] Included in ΣOC are hexachlorocyclohexanes, chlorobenzenes, dichlorodiphenyltrichloroethanes, octachlorostyrene, Σchlordanes, mirex and dieldrin.

SCCP = short-chained chlorinated paraffins; NA = not available

Statistical analysis

Concentrations of lipid-corrected ΣSCCP in fish and invertebrates did not follow a normal distribution and were log-transformed to meet normality requirements of linear models. Amphipods were not included in statistical analyses due to the low sample size. We used one-way analysis of variance (ANOVA) to compare mean concentrations of ΣSCCP among species, followed by *post-hoc* Tukey's honestly significant difference tests. We also tested the effects of fish length, weight, $\delta^{15}N$ and $\delta^{13}C$ ratios on variation in SCCP concentrations among species, as these variables have been previously found to affect levels of other POPs (Pedro et al. 2017; Pedro et al. 2019). To test for among-species variation, we used mixed-effects models. Weight and length were significantly correlated ($R_s = 0.91$, $p < 0.001$), so we included length only in the models. A stepwise model selection was performed for SCCPs with $\delta^{15}N$, $\delta^{13}C$, fish length, and first-order interactions as fixed effects, and species as a random effect, within the R-package *nlme* (Pinheiro et al. 2015). The best relative fit models were evaluated based on lower Akaike information criterion for small sample sizes (*AICc*) within the R-package *AICcmodgav* (Mazerolle 2017), and then inspected for normality of residuals and homoscedasticity. All statistical analyses were performed using R software version 3.6.0 (R Core Team 2018). Statistical significance was considered at $p < 0.05$.

BMFs

To test for trophic transfer of ΣSCCPs, we calculated biomagnification factors (BMFs) as the ratio of the average lipid-normalized concentration between predator and prey. Capelin and sand lance feed mostly on planktonic crustaceans, including amphipods (Danielsen et al. 2016; McNicholl et al. 2016; Ogloff et al. 2020), while Greenland cod feed mainly on forage fish, including capelin (Nielsen and Andersen 2001). Adult ringed seal in western Hudson Bay feed primarily on sand lance during the open water season and on Arctic cod and capelin when sea ice is present (Chambellant et al. 2013). BMF means and standard deviation terms were produced using a Monte Carlo simulation (Crystal Ball Oracle Inc) (see Supplemental Material for more details). The trophic adjusted BMF equation is as follows (Borgå et al. 2004):

$$Adjusted\ BMF = \frac{\frac{SCCP_{predator}}{SCCP_{prey}}}{\mathrm{TPpredator} - \mathrm{TPprey}} \tag{1}$$

In Equation 1, $SCCP_{predator}$ and $SCCP_{prey}$ are specific SCCPs or ΣSCCP concentrations in ng g^{-1} lipid weight in the predator and prey species; and $TP_{predator}$ and TP_{prey} represent trophic position of predator and prey. A BMF value > 1 indicates biomagnification of the compound. Trophic position (TP) was calculated based on $\delta^{15}N$ values, assuming a trophic position of 2 for calanoid copepods and respective $\delta^{15}N$ values from previous studies in the region (averaged values for *Calanus hyperboreus* and *Calanus glacialis* of 8.3 ‰ (Pomerleau et al. 2016), and a constant trophic enrichment factor of 3.8 ‰ (Hobson and Welch 1992; Fisk et al. 2003):

$$TP = \frac{2 + (\delta^{15}N_{predator} - \delta^{15}N_{copepod})}{3.8} \tag{2}$$

3.4 Results and Discussion

SCCPs in Arctic marine amphipods, fish, and ringed seals

Detectable levels of SCCPs were found in all amphipod and fish samples. The concentration of ΣSCCPs in the pooled sample of *G. oceanicus* amphipods was 559 ng g^{-1} lw (Table 3.1). When comparing on a dry weight (dw) basis, these individuals showed much lower levels compared to amphipods (*Gammarus setosus*) sampled in Svalbard from 2011-2012 (518 ± 80 ng g^{-1} dw compared to 6 ng g^{-1} dw in our samples) (Li et al. 2017). Concentrations of ΣSCCP in fish here averaged 416 ± 38 (range from 244-514) ng g^{-1} lw (Table 3.1). Just a few other studies have reported ΣSCCPs in marine or landlocked fish from Arctic/sub-Arctic regions. Our results in fish were two orders of magnitude higher than those reported in muscle of anadromous Arctic char (*Salvelinus alpinus*) (sampled in 2004) (Vorkamp et al. 2019). As seen with mercury, lower SCCP concentrations in anadromous Arctic char could be explained by "growth dilution", where anadromous fish have fast growth rates, leading to lower contaminant concentrations per unit of body mass (Swanson and Kidd 2010). SCCP concentrations in our fish were also double those of landlocked (2010) Arctic char from Nunavut, and higher than those reported in Atlantic cod (*Gadus morhua*) liver in 2003-2004 from Lofoten, Norway and Iceland (mean 93 ng g^{-1} lw in cod, although SCCP levels seem to have increased in Lofoten in more recent years (Vorkamp et al. 2019)). However, our SCCP results were comparable to the means of 300 and 540 ng g^{-1} lw in Arctic char from Norwegian lakes sampled in 2001 (Reth et al. 2006; Dick et al. 2010; Muir et al. 2013) and also comparable to those reported in Icelandic Greenland shark (*Somniosus microcephalus*), an Arctic top predator, which ranged from 50-5200 ng g^{-1} lw in 2001-2003 (Strid et al. 2013). Conversely, SCCPs in fish here were two-three times lower than levels reported in cod from Svalbard (2012; ~ 1,303 ng g^{-1} lw, 41.7 ng g^{-1} ww with 3.2 lipid % or 208 ± 50 [SE ng g^{-1} dw]; Reth et al. 2006; Li et al. 2017).

Detectable concentrations of SCCPs were also measured in all ringed seals. The young of the year seals had higher concentrations of SCCPs than the adults; mean concentrations of ΣSCCPs for the adult ringed seals were 38.3 ± 6.24 (range from 28.9-55.3) ng g^{-1} lw and young of the year were 59.9 ± 11.4 (28.9-105) ng g^{-1} lw. This is expected as marine mammals can transfer their contaminant burdens to offspring through the placenta and milk (Schweigert and Strobo 1994; Brown et al. 2016). SCCP levels in seals were 4 to 18 times lower than what we found for the fish and invertebrates (Table 3.1), and lower than those found in other marine mammal predators in the Canadian Arctic. Levels in ringed seal blubber near Ellesmere Island in 1994 averaged 579 ng g^{-1} lw (or 520 ± 69 (SE) ng g^{-1} ww, range 370-770 ng g^{-1} ww with 90 % lipid content) and slightly lower in more recent years, 248 (males) and 79 (females) ng g^{-1} lw in 2002, and an average of 92 ng g^{-1} lw from 1998-2004 in other Nunavut regions (Tomy et al. 2000; Muir et al. 2013). In beluga from Pangnirtung, Nunavut, blubber ΣSCCP levels ranged from non-detected to 330 ng g^{-1} lw until 2007 (Muir et al. 2013), and in the Northwest Greenland these averaged 225 ng g^{-1} lw in 1989 (or 190 ± 60 (SD) ng g^{-1} ww, range 110-250 ng g^{-1} ww) (Tomy et al. 2000). Polar bears from Hudson Bay showed SCCP levels in blubber ranging from 30 to 598 ng g^{-1} lw spanning different age classes in 2013 and 2014 (Letcher et al. 2018).

Variation among studies may partly be related to temporal change in environmental levels and/or production sources of SCCPs. However, measurements of SCCP levels in Arctic biota are quite limited in number and time span. For example, in blue mussels and Atlantic cod from the Norwegian Arctic, SCCPs increased from 2012-2016 at least at one station (Green et al. 2017), while belugas from the Beaufort Sea and Cumberland Sound showed a non-significant decreasing trend from the 1980s to 2010 (Muir et al. 2013). In any case, comparisons among studies remain challenging due to the array of methods used to quantify SCCPs and on-going uncertainty surrounding the reliability and comparability of results among labs and over time (Van Mourik et al. 2015). Further, our relatively low sample size of 4 to 12 specimens of each species is another potential source of uncertainty regarding SCCP levels in these species.

Variation in ΣSCCP among fish species

Of all species, Greenland cod showed the highest ΣSCCP levels, significantly higher than sand lance ($p = 0.03$), cisco ($p = 0.02$) and sculpin ($p = 0.01$); capelin also showed significantly higher levels compared to sculpin ($p = 0.04$), which in turn had the lowest ΣSCCP levels (Figure 3.2). The best model explaining variation of ΣSCCP in fish was the null model, suggesting that trophic position, carbon source, and fish length did not influence ΣSCCP variation in these species. However, when looking only at $C_{12}Cl_{6-8}$ (the congeners at higher concentrations), the model with $\delta^{15}N$ (but not other variables) was important (delta $AICc = 0.96$, marginal $R^2 = 0.16$, beta estimate for $\delta^{15}N = 0.19$ (more details in the Supplementary Material)). Thus, trophic position may be more important for the heavier SCCP compounds. As seen in rainbow trout, the heavier (higher chlorinated and longer carbon-chain) chlorinated paraffins showed longer half-lives and greater bioaccumulation (Fisk et al. 1998; Fisk et al. 2000). Higher $\delta^{15}N$ values in Greenland cod could explain the higher concentrations of these particular SCCPs compared to most other species. While a previous study in lake trout from the Great Lakes also did not find any correlations between SCCP levels and fish length, weight, age, lipid content or trophic level (Saborido Basconcillo et al. 2015), recent studies found biomagnification of SCCPs in fish in Liaodong Bay, in a marine food web in the South China Sea and from oysters to crabs in the Pearl River Estuary in China (Huang et al. 2017; Zeng et al. 2017; Huang et al. 2019), as well as biomagnification potential in a food web of the Baltic Sea (de Wit et al. 2020).

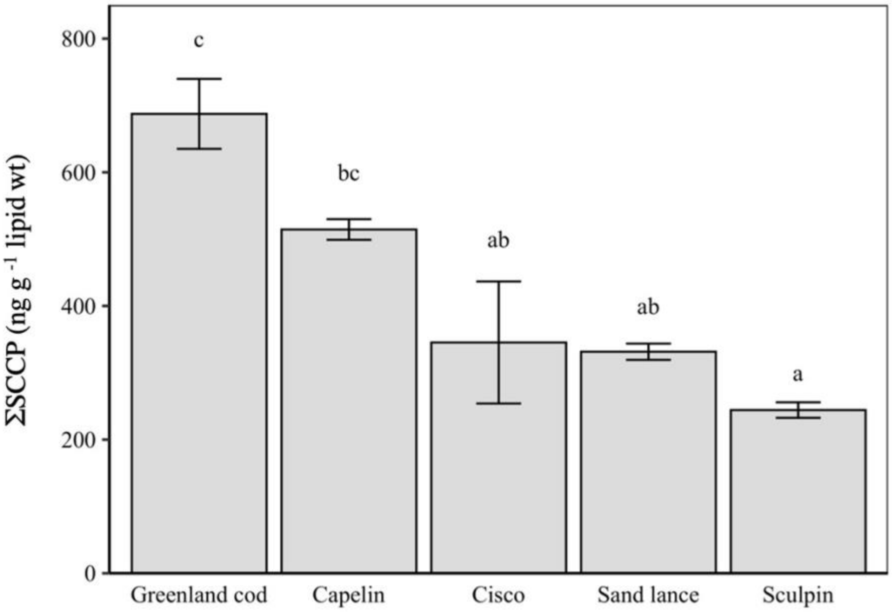

Figure 3.2 Mean concentrations of short-chained chlorinated paraffins (ΣSCCP) in fish from western Hudson Bay, Nunavut, sampled in 2014. Error bars represent standard errors. Significant differences in SCCP levels among species are indicated by different letters above each bar.

Congener patterns and composition within species

For fish and invertebrate species, only homologues with 6 to 9 chlorines were detected (6-8 chlorines were more prevalent), with C_{11} and C_{12} chains at higher concentrations, especially C_{12} (Figure 3.3a, Table S3.1). This is comparable to Greenland sharks in Iceland (Strid et al. 2013), Arctic char from the European Arctic (Reth et al. 2006), along with fish from the North and Baltic Seas (Reth et al. 2005), and freshwater bodies across Canada (Saborido Basconcillo et al. 2015), wherein the contribution of C_{11} and C_{12} homologues was higher compared to C_{10} and C_{11} (Reth et al. 2006; Strid et al. 2013). Among fish species, the most notable variation in congener patterns was observed in cisco, which had higher % of C_{11} (especially $C_{11}Cl_8$) and lower C_{12} congeners relative to other species (Figure S3.1). This could, in part, be related to the high lipid content of cisco, since longer chain homologues of SCCPs have a higher affinity for lipid rich tissues (Li et al. 2016), or their anadromous behavior could reflect congener composition of both marine and freshwater environments (Bernatchez and Dodson 1990).

In ringed seals, a broader range of homologues were detected (C_{10}-C_{13} with 5 to 10 chlorines), with C_{10} and C_{11} chains being dominant (75%) (Figure 3.3b). The more volatile homologue groups, C_{10} and C_{11} with 6-8 chlorines (Drouillard et al. 1998), have been prevalent in Arctic marine mammals and associated with a higher contribution from atmospheric deposition rather than local source contamination in Arctic environments (Tomy et al. 2000; Strid et al. 2013; Li et al. 2017; Letcher et al. 2018). Compounds of C_{13} were only detected in amphipods (only the congener with 7 chlorines) and seals. Differences between fish and marine mammals could indicate different assimilation and/or metabolism of SCCP homologues. In rainbow trout, lower chlorinated and shorter chain paraffins were more susceptible to biotransformation (Fisk et al. 2000). Further, the detection of C_{13} in ringed seals but not in fish could also be associated with the type of the tissue analysed, that is lipid-rich blubber could have higher affinity for the heavier congeners (Li et al. 2016).

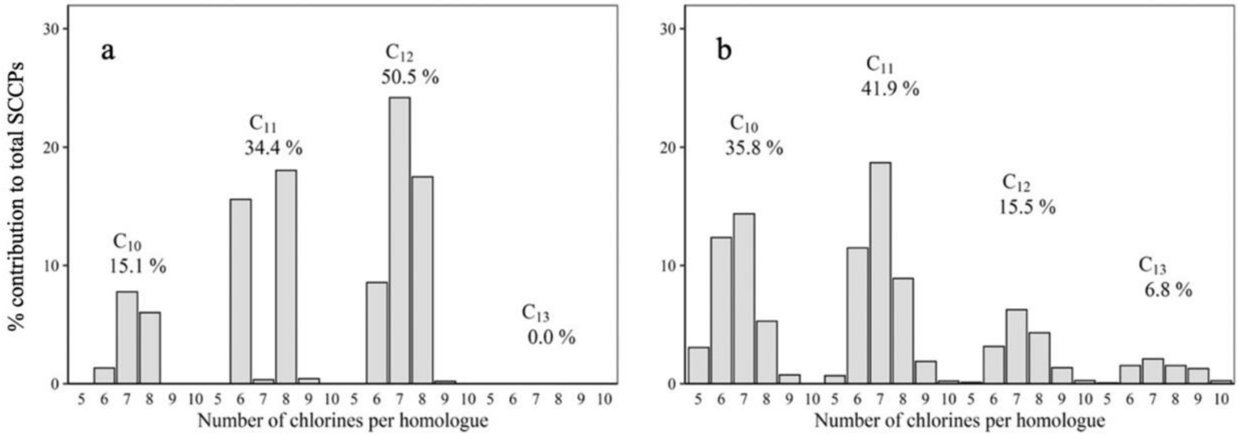

Figure 3.3 Average percent contribution of short-chained chlorinated paraffin homologs to total % in (**a**) fish and invertebrates sampled in 2014 and in (**b**) ringed seals sampled in 2017 from western Hudson Bay, Nunavut.

Table 3.2 Trophic position-adjusted biomagnification factors (±standard deviation) using Monte Carlo simulation for short-chain chlorinated paraffins for the western Hudson Bay marine food web.

Contaminant	Capelin/ amphipod	Sand lance/ amphipod	Greenland cod/ capelin	Adult Ringed seal/ prey fish[a]
ΣSCCP	0.85 ± 0.28	0.61 ± 0.26	1.72 ± 0.86	0.32 ± 0.17
$C_{10}Cl_6$[b]	ND	ND	1.56	2.23
$C_{10}Cl_7$	0.99 ± 0.24	0.90 ± 0.41	1.73 ± 0.93	0.55 ± 0.25
$C_{10}Cl_8$	0.79 ± 0.18	0.43 ± 0.21	1.08 ± 1.02	0.31 ±0.18
$C_{11}Cl_6$	1.11 ± 0.27	1.65 ± 0.67	3.16 ± 1.17	0.24 ± 0.13
$C_{11}Cl_8$	0.58 ± 0.25	0.35 ± 0.14	1.28 ± 1.02	0.20 ± 0.15
$C_{11}Cl_9$[b]	0.33	0.08	0.00	0.88
$C_{12}Cl_6$	0.97 ± 0.20	1.18 ± 0.57	2.05 ± 1.11	0.10 ± 0.06
$C_{12}Cl_7$	0.83 ± 0.32	0.46 ± 0.19	1.39 ± 0.79	0.09 ± 0.06
$C_{12}Cl_8$	0.69 ± 0.16	0.27 ± 0.11	1.57 ± 0.62	0.07 ± 0.05

[a] Prey fish includes capelin and sand lance

[b] Data could not be transformed to fit any distribution offered in Crystal Ball, thus the Monte Carlo simulation was not possible for these homologues and equation (1) was used instead

SCCP = short chain chlorinated paraffins; ND = not determined (the SCCP concentration was below the detection limit for at least one of the species in the ratio calculation).

Levels of SCCPs compared to legacy POPs in Arctic marine fish and amphipods

Levels of ΣSCCPs were double those of ΣPCBs and ΣOCs reported previously in these same samples of capelin, sand lance and cisco; and about three times higher in Greenland cod (Table 3.1) (Pedro et al. 2017). Similar results were also observed in Greenland shark, with SCCPs in the same range as individual PCBs (Strid et al. 2013). The opposite was observed in seals, were the SCCPs levels were 2-4 times lower than PCB and OC levels previously reported for seals in Arviat (2014-2016) (Houde et al. 2019). In previous studies on polar bears, belugas and ringed seals, ΣSCCP levels were 2-3 times lower than those of ΣPCBs (Tomy et al. 2000; Letcher et al. 2018). Thus, the high concentrations of ΣSCCPs in fish compared to those of other legacy POPs provide evidence of a substantial burden of SCCPs at least for some marine species in the Canadian Arctic. Furthermore, little is known about the burden of medium-chain chlorinated paraffins (MCCPs; C_{14-17}) and long-chain chlorinated paraffins (LCCPs; ≥ 18) in Arctic marine species, despite their presence in the Arctic and bioaccumulation potential (Reth et al. 2006; de Wit et al. 2020).

BMFs

Our BMF results were not always in agreement with the few studies that have analysed food-web transfer of SCCPs in Arctic ecosystems. The ΣSCCP BMFs were 0.85 ± 0.28 for amphipod-to-capelin, 0.61 ± 0.26 for amphipod-to-sand lance, 1.72 ± 0.86 for capelin-to-Greenland cod, and 0.32 ± 0.17 for prey fish-to-ringed seal (Table 3.2). In Svalbard, the average BMF for SCCPs calculated from gammarid amphipods to Atlantic cod was 0.46 (Li et al. 2017), comparable to what we found here, where amphipod-to-capelin and -sand lance BMFs were below one suggesting no biomagnification. In a food web in the Baltic Sea including mussels, fish, seals and seabirds, the BMFs ranged 1.5–5.0 for SCCPs, with higher BMFs between marine mammals and fish (BMF of 3.9 for herring-to-porpoise and 5.0 for herring-to-harbour seal) (de Wit et al. 2020). This is opposite to results found here, where the majority of the BMFs were below one for the prey fish-to-ringed seal, suggesting no biomagnification; however, BMFs were mostly above one for capelin-to-Greenland cod, suggesting biomagnification. The low fish-to-seal BMFs could be related, in part, to the difference in sample collection years between ringed seal (2017) and the prey fish (2014) and possible interannual variation of SCCPs in these species. The biomagnification between fish-to-fish, but not between fish-to-seal could in part be related to differences in metabolic capacities between poikilotherms and homeotherms, differences in feeding strategies, or the higher potential capacity for seals to biotransform chlorinated contaminants as seen for OCs in harbour seals (Boon et al. 1992).

Considering all BMFs, the lower chlorinated (Cl_6) SCCPs had the highest biomagnification factors. This is contrary to what was reported in Svalbard for gammarid-to-cod BMFs, where the higher chlorinated SCCPs were associated with higher biomagnification (Li et al, 2017). The influence of carbon atoms on the BMFs was variable with the exception of the fish-to-seal ratios, where the BMFs were highest for the C_{10} chain lengths and the lowest were found for C_{12}. This is consistent with biota in Antarctica and fish in Lake Michigan, for which SCCP BMFs decreased with increasing carbon chain length (Houde et al. 2008; Li et al. 2016). The ΣSCCP BMFs in the present study were up to two orders of magnitude lower than other legacy POPs from Arctic food webs (Hop et al. 2002; Hoekstra et al. 2003; McKinney et al. 2012; de Wit et al. 2020). Thus, SCCP biomagnification may be limited in this Arctic food web relative to biomagnification found for legacy POPs; however, more food web studies and improved analytical capabilities are needed to further evaluate SCCP biomagnification.

3.5 Conclusion

In this western Hudson Bay food web, SCCPs were detected in all amphipod, fish and seal species collected and concentrations were relatively high in fish, especially piscivorous species. Nonetheless, trophic position-adjusted BMFs suggest limited biomagnification potential of SCCPs in this pelagic marine food web. To better understand the biomagnification potential of SCCPs, a more complete representation of the food web is needed, including larger sample sizes and additional species including the apex polar bear predator. In fish, the SCCP levels were in the same range or higher than those of PCBs and OCs measured in the same specimens, while for seals the opposite was observed for individuals in the same region. The growing evidence of CP accumulation in biota in remote regions, often in the same range as legacy POPs as shown here for SCCPs, highlights the importance of continuing monitoring of Arctic ecosystems for SCCPs. Medium and long-chain chlorinated paraffins would also be of importance to investigate.

Supporting Information—The Supporting Information is available on the Wiley Online Library at https://doi.org/10.1002/ etc.5188.

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3.8 Supplemental Information

Additional details on laboratory analyses:

Lipid-extracted stable isotope analyses in seals

Muscle samples were freeze-dried and homogenized. For the subset of lipid extracted samples, the lipid was extracted by agitating the dried powdered muscle tissue in a 2:1 chloroform-methanol solution for 24 h. The tissue and solvent were then filtered, and the resulting residue-filter paper dried at 60 °C for 48 h to evaporate the remaining solvent.

Monte Carlo Simulation

A log normal distribution was used for each assumption of the simulation, except for amphipods (*n* = 1 pool) where a uniform distribution was used. The forecast of the simulation was defined by the trophic adjusted BMF equation and each BMF simulation was run 1000 times. Two homologues ($C_{10}Cl_6$, $C_{11}Cl_9$) were not able to be transformed to fit any of the distributions offered by Crystal Ball and so the simulation was not performed and the trophic position BMF equation was used.

Mixed effects models – AICc ranking results:

```
Global model call: lme.formula(fixed = log(sum_SCCPs) ~ C + N +
Len_mm, data = sccp4,
    random = ~1 | Species, method = "REML")
---
Model selection table
  (Int)       C     Len_mm      N df  logLik AICc delta weight
1 5.868                           3 -29.251 65.2  0.00  0.782
5 3.829                    0.1344  4 -29.523 68.3  3.07  0.169
2 6.726 0.04405                   4 -30.938 71.1  5.90  0.041
6 4.456 0.02637            0.1268  5 -31.430 74.8  9.57  0.007
3 5.948         -0.0003965        4 -34.680 78.6 13.38  0.001
7 3.910         -0.0007530 0.1389  5 -34.972 81.9 16.65  0.000
4 7.359 0.06383 -0.0012140        5 -36.070 84.1 18.85  0.000
8 5.230 0.05405 -0.0013860 0.1298  6 -36.550 87.9 22.67  0.000
Models ranked by AICc(x, REML = TRUE)
Random terms (all models):
```

```
‘1 | Species’
Global model call: lme.formula(fixed = log(sum_C12) ~ C + N +
Len_mm, data = sccp6,
    random = ~1 | Species, method = "REML")
---
Model selection table
  (Int)       C     Len_mm      N df  logLik AICc delta weight
1 5.095                           3 -28.256 63.2  0.00  0.589
5 2.270                    0.1859 4 -27.474 64.2  0.96  0.365
2 5.877 0.04020                   4 -29.968 69.2  5.95  0.030
6 2.926 0.03174            0.1832 5 -29.326 70.6  7.35  0.015
3 5.148         -0.0002591        4 -33.539 76.3 13.09  0.001
7 2.369         -0.0007439 0.1893 5 -32.778 77.5 14.25  0.000
4 6.243 0.05014 -0.0008353        5 -35.091 82.1 18.88  0.000
8 3.389 0.04601 -0.0012080 0.1875 6 -34.422 83.6 20.41  0.000
Models ranked by AICc(x, REML = TRUE)
Random terms (all models):
‘1 | Species’
```

Table S3.1 Short-chained chlorinated paraffin congener concentrations (ng g^{-1} lipid weight) in amphipods (2014), fish (2014), and ringed seals (2017) from western Hudson Bay, Nunavut. Results denoted as arithmetic mean (min-max). nd = not detected

	Amphipods	Capelin	Sand lance	Sculpin	Cisco	GreenlandCod	Ringed Seal
	Gammarus oceanicus	*Mallotus villosus*	*Ammodytes* spp.	*Myoxocephalus* spp.	*Coregonus artedi*	*Gadus ogac*	*Pusa hispida*
$C_{10}Cl_5$	nd	nd	nd	nd	nd	nd	1.8 (0.5-13.4)
$C_{10}Cl_6$	nd	2.8 (nd-4.5)	9.0 (5.7-15.0)	3.4 (nd-5.2)	5.8 (2.0-12.5)	5.9 (2.0-9.1)	5.9 (3.8-11.1)
$C_{10}Cl_7$	30.1	36.8 (18.2-47.7)	30.4 (17.7-50.2)	21.8 (11.5-34.3)	21.4 (7.7-39.6)	49.5 (32.7-83.4)	7.2 (4.4-16.4)
$C_{10}Cl_8$	35.9	34.8 (19.7-44.6)	17.1 (8.1-27.9)	23.4 (nd-39.7)	17.1 (5.3-26.4)	30.6 (nd-71.0)	2.7 (1.6-6.5)
$C_{10}Cl_9$	nd	nd	nd	nd	nd	nd	0.4 (0.2-0.8)
$C_{10}Cl_{10}$	nd	nd	nd	nd	nd	nd	nd
$C_{11}Cl_5$	nd	nd	nd	nd	nd	nd	0.3 (0.2-0.6)
$C_{11}Cl_6$	35.8	48.6 (23.6-55.3)	65.8 (43.5-103)	56.0 (39.3-72.4)	45.2 (18.2-92.9)	126 (93.2-181)	5.7 (2.7-11.8)
$C_{11}Cl_7$	nd	nd	4.0 (2.3-6.6)	0.7 (nd-3.9)	nd	nd	9.8 (4.8-25.4)
$C_{11}Cl_8$	129	90.0 (6.8-140)	50.5 (34.9-78.1)	34.9 (14.7-50.7)	147 (10.9-517)	82.4 (53.3-158)	4.7 (1.9-11.3)
$C_{11}Cl_9$	10.0	4.0 (2.1-7.1)	0.9 (0.4-1.5)	1.0 (nd-2.7)	0.2 (nd-0.4)	nd	1.0 (0.4-2.3)
$C_{11}Cl_{10}$	nd	nd	nd	nd	nd	nd	0.1 (nd-.0.4)
$C_{12}Cl_5$	nd	nd	nd	nd	nd	nd	0.1 (nd-0.2)
$C_{12}Cl_6$	28.9	34.0 (22.9-43.0)	37.8 (23.7-63.0)	22.0 (6.5-31.9)	27.0 (10.2-55.6)	57.7 (31.4-106)	1.6 (0.6-3.3)
$C_{12}Cl_7$	146	147 (11.4-204)	75.6 (53.4-118)	53.5 (24.1-106)	53.0 (19.3-99.2)	152 (110-261)	3.2 (1.5-6.0)
$C_{12}Cl_8$	137	114 (74.1-146)	39.7 (26.1-63.9)	23.4 (nd-79.9)	29.4 (12.7-44.4)	151 (120-240)	2.3 (0.7-4.8)
$C_{12}Cl_9$	2.5	3.0 (1.6-5.4)	nd	nd	nd	nd	0.7 (0.1-1.8)
$C_{12}Cl_{10}$	nd	nd	nd	nd	nd	nd	0.1 (nd-0.4)
$C_{13}Cl_5$	nd	nd	nd	nd	nd	nd	nd
$C_{13}Cl_6$	nd	nd	nd	nd	nd	nd	0.8 (0.3-1.3)
$C_{13}Cl_7$	4.3	nd	nd	nd	nd	nd	1.0 (0-2.6)
$C_{13}Cl_8$	nd	nd	nd	nd	nd	nd	0.8 (0.3-1.5)
$C_{13}Cl_9$	nd	nd	nd	nd	nd	nd	0.7 (0.2-1.9)
$C_{13}Cl_{10}$	nd	nd	nd	nd	nd	nd	0.1 (nd -0.6)

Table S3.2 Reference values for the in-house carp GLCARP11 Reference Material.

Chemical	Concentration ($ng\ g^{-1}$)	Standard deviation
PBDE-28	4.25	0.9
PBDE-49	1.83	0.37
PBDE-47	48.48	12
PBDE-100	6.17	1.4
PBDE-154	1.70	0.27
HCB	2.58	0.48
DDE	61.86	11.04
Oxychlordane	0.47	0.05
Dieldrin	1.69	0.26
PCB52	3.26	0.52
PCB101	11.79	1.43
PCB153	24.00	2.85
PCB138	19.90	2.39
PCB180	20.98	2.96

Table S3.3 SCCP values for NIST 1946 Standard Reference Material run with the ringed seal samples in ng/g wet weight.

	SRM1 NIST 1946	SRM2 NIST 1946
Sample weight	1.49	1.47
Spike recovery (13C10hexaCldecane)	87.9	143
C10Cl5	11.40	3.06
C10Cl6	68.69	30.58
C10Cl7	52.72	31.46
C10Cl8	14.41	7.32
C10Cl9	2.11	0.79
C10Cl10	0.04	0.13
C11Cl5	1.90	1.37
C11Cl6	49.06	37.28
C11Cl7	41.81	45.58
C11Cl8	12.62	15.03
C11Cl9	2.26	3.10
C11Cl10	0.19	1.01
C12Cl5	2.12	1.15
C12Cl6	20.71	17.94
C12Cl7	24.40	25.68
C12Cl8	9.16	10.92
C12Cl9	1.93	3.32
C12Cl10	0.25	1.32
C13Cl5	1.77	1.65
C13Cl6	12.59	15.21
C13Cl7	15.46	16.97
C13Cl8	5.73	5.13
C13Cl9	2.86	2.56
C13Cl10	0.64	0.46

Table S3.4 Short-chained chlorinated paraffin congener concentrations (ng g^{-1} lipid weight), δ^{15}N and δ^{13}C, length (mm), weight (g) and lipid % of a pool of amphipods (2014), individual prey fish (2014), and individual ringed seals (2017) from western Hudson Bay, Nunavut.

Species	**Capelin-1**	**Capelin-2**	**Capelin-3**	**Capelin-4**	**Capelin-5**	**Capelin-6**	**Capelin-8**	**Capelin-9**	**Capelin-10**	**Capelin-11**
δ_{13}C	-20.16	-21.11	-20.52	-19.99	-21.23	-20.94	-21.33	-21.02	-23.11	-21.56
δ_{15}N	14.88	14.82	14.98	15.14	15.34	14.82	14.42	14.23	14.08	13.93
Length (mm)	103.00	107.00	104.00	106.00	107.00	119.00	109.00	86.00	92.00	87.00
Weight (g)	7.81	9.53	9.66	9.50	7.70	13.82	9.84	4.00	4.33	4.57
Lipid%	1.38	1.38	2.39	1.25	1.25	2.5	1.61	1.61	1.61	1.61
ΣSCCPs	693.48	693.48	346.74	601.98	601.98	186.53	504.97	504.97	504.97	504.97
C10Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C10Cl6	2.52	2.52	nd	2.12	2.12	1.06	4.47	4.47	4.47	4.47
C10Cl7	47.68	47.68	23.74	36.36	36.36	18.22	39.44	39.44	39.44	39.44
C10Cl8	44.57	44.57	22.74	39.37	39.37	19.72	34.47	34.47	34.47	34.47
C10Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C10Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl6	53.91	53.91	29.99	51.80	51.80	23.60	55.28	55.28	55.28	55.28
C11Cl7	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl8	139.86	139.86	64.63	111.68	111.68	6.77	81.37	81.37	81.37	81.37
C11Cl9	7.13	7.13	2.05	4.13	4.13	3.59	3.04	3.04	3.04	3.04
C11Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl6	42.97	42.97	22.87	39.45	39.45	25.27	31.86	31.86	31.86	31.86
C12Cl7	203.62	203.62	103.98	186.14	186.14	11.41	143.48	143.48	143.48	143.48
C12Cl8	146.38	146.38	75.40	128.32	128.32	74.06	109.32	109.32	109.32	109.32
C12Cl9	5.37	5.37	1.59	2.74	2.74	2.88	2.22	2.22	2.22	2.22
C12Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl6	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl7	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl8	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd

nd: not detected

Table S3.4 Cont.

Species	Greenland Cod-1	Greenland Cod-2	Greenland Cod-3	Greenland Cod-4	Greenland Cod-5
δ13C	-18.64	-18.73	-19.21	-19.04	-19.49
δ15N	17.79	18.49	17.57	18.57	17.20
Length (mm)	311.00	278.00	290.00	40nd	265.00
Weight (g)	398.00	463.00	381.00	779.00	237.00
Lipid%	0.58	0.45	0.45	0.8	0.58
ΣSCCPs	518.98	1148.15	534.65	610.00	625.22
C10Cl5	nd	nd	nd	nd	nd
C10Cl6	2.00	9.13	4.33	8.49	5.52
C10Cl7	32.74	83.44	34.98	51.88	44.24
C10Cl8	37.79	71.02	nd	nd	44.06
C10Cl9	nd	nd	nd	nd	nd
C10Cl10	nd	nd	nd	nd	nd
C11Cl5	nd	nd	nd	nd	nd
C11Cl6	93.17	181.26	108.69	118.13	128.18
C11Cl7	nd	nd	nd	nd	nd
C11Cl8	54.68	158.39	53.25	75.88	69.66
C11Cl9	nd	nd	nd	nd	nd
C11Cl10	nd	nd	nd	nd	nd
C12Cl5	nd	nd	nd	nd	nd
C12Cl6	31.35	105.88	34.98	63.13	52.94
C12Cl7	110.24	261.00	117.93	142.25	128.70
C12Cl8	125.39	239.65	140.81	127.50	120.17
C12Cl9	nd	nd	nd	nd	nd
C12Cl10	nd	nd	nd	nd	nd
C13Cl5	nd	nd	nd	nd	nd
C13Cl6	nd	nd	nd	nd	nd
C13Cl7	nd	nd	nd	nd	nd
C13Cl8	nd	nd	nd	nd	nd
C13Cl9	nd	nd	nd	nd	nd
C13Cl10	nd	nd	nd	nd	nd

nd: not detected

Table S3.4 Cont.

Species	Sandlance-1	Sandlance-2	Sandlance-3	Sandlance-4	Sandlance-5	Sandlance-6	Sandlance-7	Sandlance-8	Sandlance-9	Sandlance-10	Sandlance-11	Sandlance-13
δ13C	-21.21	-23.15	-21.60	-21.20	-21.78	-22.06	-21.53	-21.97	-21.33	-21.76	-22.86	-19.61
δ15N	13.58	12.68	14.49	15.81	14.20	14.48	13.45	14.15	14.29	14.46	14.50	14.87
Length (mm)	109.00	109.00	93.00	84.00	108.00	102.00	84.00	91.00	86.00	92.00	100.00	101.00
Weight (g)	5.13	3.68	2.97	2.70	4.20	4.62	3.55	3.66	2.77	4.15	3.12	5.47
Lipid%	1.7	1.7	1.7	1.7	1.48	1.48	1.48	1.48	1.25	1.25	1.25	1.25
ΣSCCPs	220.48	220.48	220.48	220.48	245.66	245.66	245.66	245.66	528.37	528.37	528.37	528.37
C10Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C10Cl6	5.71	5.71	5.71	5.71	6.47	6.47	6.47	6.47	14.95	14.95	14.95	14.95
C10Cl7	17.69	17.69	17.69	17.69	23.41	23.41	23.41	23.41	50.17	50.17	50.17	50.17
C10Cl8	8.06	8.06	8.06	8.06	15.25	15.25	15.25	15.25	27.91	27.91	27.91	27.91
C10Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C10Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl6	43.49	43.49	43.49	43.49	51.06	51.06	51.06	51.06	102.82	102.82	102.82	102.82
C11Cl7	3.30	3.30	3.30	3.30	2.27	2.27	2.27	2.27	6.57	6.57	6.57	6.57
C11Cl8	34.86	34.86	34.86	34.86	38.61	38.61	38.61	38.61	78.14	78.14	78.14	78.14
C11Cl9	0.77	0.77	0.77	0.77	0.38	0.38	0.38	0.38	1.53	1.53	1.53	1.53
C11Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl6	26.80	26.80	26.80	26.80	23.65	23.65	23.65	23.65	62.95	62.95	62.95	62.95
C12Cl7	53.42	53.42	53.42	53.42	55.79	55.79	55.79	55.79	117.58	117.58	117.58	117.58
C12Cl8	26.14	26.14	26.14	26.14	28.96	28.96	28.96	28.96	63.88	63.88	63.88	63.88
C12Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl6	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl7	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl8	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd

nd: not detected

Table S3.4 Cont.

Species	Sculpin -1	Sculpin -3	Sculpin -4	Sculpin -5	Sculpin -6	Sculpin -7	Amphipod -1	Cisco-2	Cisco-3	Cisco-4	Cisco-5
δ13C	-18.13	-18.85	-18.91	-15.65	-15.49	-14.22	-16.50	-16.73	-18.09	-21.65	-17.69
δ15N	17.46	16.02	15.91	14.59	14.44	16.47	10.04	13.11	13.98	14.02	12.59
Length (mm)	230.00	129.00	168.00	220.00	230.00	185.00	nd	365.00	335.00	331.00	351.00
Weight (g)	227.52	25.23	53.52	151.73	191.88	160.06	nd	784.00	702.00	623.00	640.00
Lipid%	0.69	0.37	0.37	0.62	0.66	1.33	0.75	1.56	2.67	7.01	2.82
ΣSCCPs	314.49	293.52	293.52	132.67	232.01	199.05	558.70	884.62	243.70	86.16	166.74
C10Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C10Cl6	nd	5.23	5.23	2.76	3.73	3.73	nd	12.45	6.21	1.96	2.51
C10Cl7	12.62	34.29	34.29	11.45	19.97	17.89	30.12	39.56	21.28	7.69	17.09
C10Cl8	nd	39.67	39.67	20.81	25.65	14.48	35.94	26.37	14.16	5.25	22.52
C10Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C10Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C11Cl6	57.25	72.36	72.36	43.27	51.53	39.30	35.80	92.86	49.23	18.22	20.64
C11Cl7	nd	nd	nd	nd	nd	3.93	nd	nd	nd	nd	nd
C11Cl8	29.86	50.71	50.71	14.74	32.99	30.39	128.62	516.48	33.22	10.91	26.89
C11Cl9	nd	2.66	2.66	nd	nd	0.91	10.00	0.42	nd	nd	0.31
C11Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C12Cl6	6.52	23.54	23.54	17.26	31.88	29.28	28.93	55.63	27.56	10.15	14.71
C12Cl7	105.65	47.66	47.66	24.11	51.93	44.24	145.80	99.18	55.72	19.29	37.62
C12Cl8	79.86	16.42	16.42	-1.99	14.36	15.12	136.96	44.41	36.17	12.66	24.42
C12Cl9	nd	nd	nd	nd	nd	nd	2.52	nd	nd	nd	nd
C12Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl6	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl7	nd	nd	nd	nd	nd	nd	4.32	nd	nd	nd	nd
C13Cl8	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
C13Cl10	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd

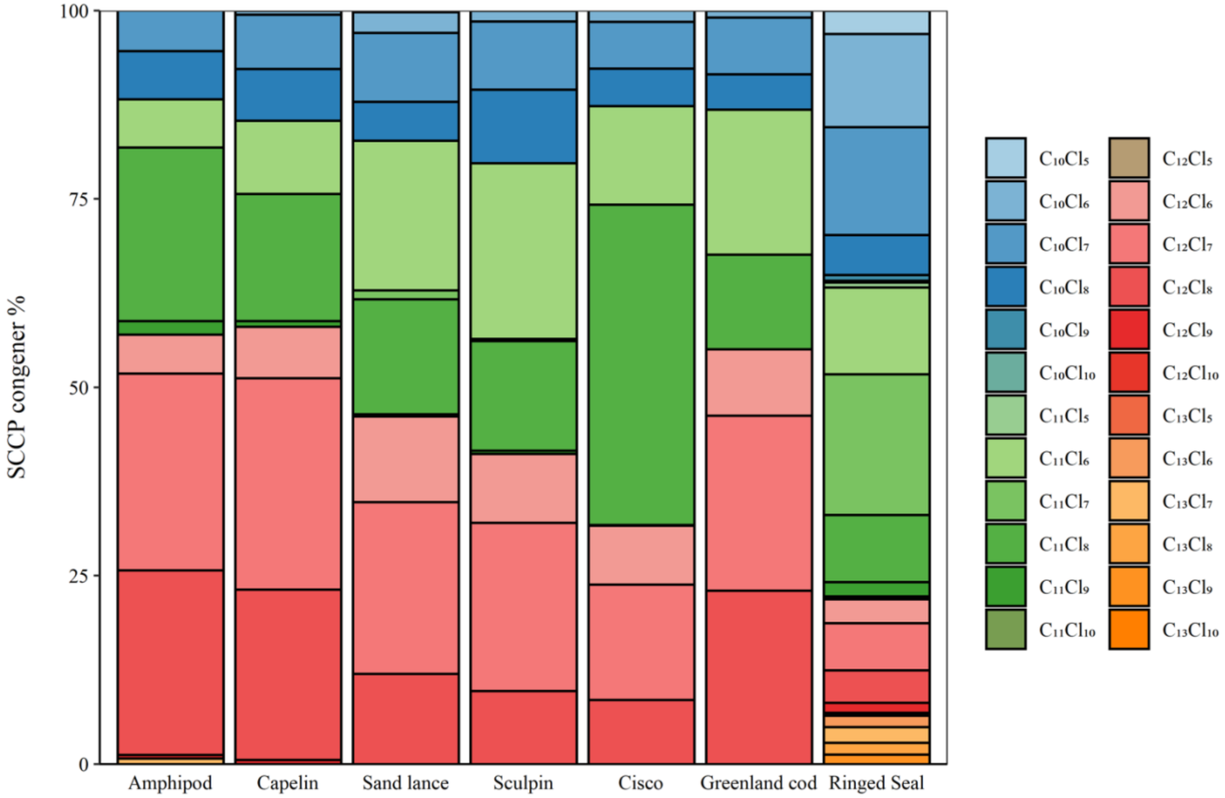
nd: not detected

Table S3.4 Cont.

Species	Seal-3	Seal-5	Seal-7	Seal-9	Seal-14	Seal-17	Seal-27	Seal-30	Seal-31	Seal-36
δ13C	-21.327	-21.392	-23.314	-20.664	-22.210	-23.391	-21.031	-20.815	-21.623	-21.074
δ15N	16.78	15.43	14.73	14.08	15.56	15.24	15.65	14.16	13.88	14.74
Lipid%	84.00	88.00	83.00	93.00	92.00	86.00	90.00	87.00	92.00	89.00
ΣSCCPs	29.76	28.14	74.61	46.99	66.62	40.00	55.27	105.00	28.87	37.55
C10Cl5	0.47	0.69	0.38	0.38	13.39	0.83	0.14	0.90	0.20	0.26
C10Cl6	3.80	4.76	6.49	4.67	9.12	6.93	3.97	11.07	4.34	4.15
C10Cl7	4.38	4.85	8.98	5.01	8.04	5.72	7.75	16.41	4.94	5.79
C10Cl8	1.95	1.55	3.59	1.82	2.13	2.13	3.33	6.45	1.57	2.22
C10Cl9	0.29	0.22	0.62	0.24	0.32	0.36	0.49	0.82	0.19	0.29
C10Cl10	nd	nd	nd1	nd	nd1	nd1	nd1	nd2	nd	nd1
C11Cl5	0.21	0.29	0.35	0.23	0.57	0.42	0.23	0.53	0.26	0.16
C11Cl6	2.67	3.31	7.27	4.86	6.94	5.09	6.03	11.81	4.15	5.30
C11Cl7	5.87	5.06	14.35	7.77	7.10	6.49	14.02	25.41	4.79	7.53
C11Cl8	3.60	2.25	8.11	4.05	3.01	3.13	6.42	11.31	1.93	2.93
C11Cl9	0.69	0.38	2.29	1.15	0.79	0.58	1.20	1.94	0.44	0.60
C11Cl10	nd1	nd	0.40	0.18	0.12	nd9	0.14	0.19	nd5	nd9
C12Cl5	nd	nd3	nd5	nd3	0.20	nd7	nd6	0.17	nd3	nd4
C12Cl6	0.60	0.72	2.28	1.25	2.28	1.31	1.84	3.31	1.20	1.47
C12Cl7	2.00	1.50	5.79	3.52	3.47	2.64	3.75	6.04	1.48	2.17
C12Cl8	1.32	0.73	4.81	3.82	2.99	1.15	2.19	3.48	0.96	1.23
C12Cl9	0.26	nd9	1.69	1.75	1.25	0.35	0.60	0.84	0.27	0.32
C12Cl10	nd1	nd	0.36	0.36	0.25	0.11	0.13	0.13	nd6	nd9
C13Cl5	nd	nd1	nd2	nd	0.14	nd1	nd2	0.11	nd4	nd9
C13Cl6	0.34	0.45	0.94	0.47	1.25	0.67	0.72	1.25	0.58	0.88
C13Cl7	0.70	0.67	2.62	1.76	nd	1.01	1.15	1.63	0.80	1.04
C13Cl8	0.34	0.39	1.31	1.36	1.49	0.52	0.64	0.74	0.37	0.55
C13Cl9	0.23	0.18	1.55	1.85	1.41	0.30	0.39	0.37	0.19	0.29
C13Cl10	nd2	nd2	0.35	0.45	0.33	nd7	nd6	nd7	nd3	nd4

nd: not detected

Figure S3.1 Homologue proportions (%) for short-chain chlorinated paraffins (SCCP) measured in prey fish and invertebrates (2014) and ringed seals (2017) from western Hudson Bay, Nunavut.



Connecting Text

The insight developed in Chapter 3 contributes to the growing evidence of SCCP accumulation in remote regions and Arctic marine life. Understanding the accumulation of this “new” contaminant in ringed seals is essential for the conservation of this biomonitoring species as well as the health of polar bears and community members consuming the ringed seal. Regardless of the type of contaminants, whether they are “new”, “legacy” or trace metals, further understanding of how climate change will impact the fate and accumulation of contaminants in Arctic marine life is continuously needed.

In Chapter 4 of this thesis, insight into the effects of climate-change-driven feeding shifts on legacy, new and trace metal contaminant concentrations in ringed seals across the Canadian Arctic is developed. This chapter goes beyond just reporting contaminant levels and compares feeding patterns of ringed seals across the Arctic, tests contaminant variation between sites and links climate-induced feeding changes with ringed seal contaminant loads. This manuscript, “Feeding and contaminant patterns of sub-Arctic and Arctic ringed seals: potential insight into climate change-contaminant interactions”, will be submitted to the journal *Environmental Pollution*.

Chapter 4: Feeding and contaminant patterns of sub-Arctic and Arctic ringed seals: potential insight into climate change-contaminant interactions

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Highlights:

- Fatty acids and stable isotopes used as diet tracers for ringed seals at four sites
- Diet tracers differ between ringed seals of the sub-Arctic and Arctic
- Stable isotopes best explain contaminant variation among ringed seals
- Diet differences linked to higher levels of PFAS in sub-Arctic seals
- Future climate-driven diet shifts in Arctic seals may increase levels of new POPs

4.1 Abstract

To provide insight into how climate-driven diet shifts may impact contaminant exposures of Arctic species, we compared feeding ecology and contaminant concentrations in ringed seals (*Pusa hispida*) from two Canadian sub-Arctic (Nain at 56.5°N, Arviat at 61.1°N) and two Arctic sites (Sachs Harbour at 72.0 °N, Resolute Bay at 74.7 °N). In the sub-Arctic, empirical evidence of changing prey fish communities has been documented, while less community change has been reported in the Arctic to date, suggesting current sub-Arctic conditions may be a harbinger of future Arctic conditions. Here, Indigenous partners collected tissues from subsistence-harvested ringed seals in 2018. Blubber fatty acids (FAs) and muscle stable isotopes ($\delta^{15}N$, $\delta^{13}C$) indicated dietary patterns, while measured contaminants included heavy metals (e.g., total mercury (THg)), legacy persistent organic pollutants (e.g., dichlorodiphenyltrichloroethanes (DDTs)), polybrominated diphenyl ethers (PBDEs), and per-/polyfluoroalkyl substances (PFASs). FA signatures are distinct between sub-Arctic and Resolute Bay seals, likely related to higher consumption of southern prey species including capelin (*Mallotus villosus*) in the sub-Arctic but on-going feeding on Arctic species in Resolute Bay. Sachs Harbour ringed seals show FA overlap with all locations, possibly consuming both southern and endemic Arctic species. Negative $\delta^{13}C$ estimates for PFAS models suggest that more pelagic, sub-Arctic type prey (e.g., capelin) increases PFAS concentrations, whereas the reverse occurs for, e.g., THg, ΣPBDE, and ΣDDT. Inconsistent directionality of $\delta^{15}N$ estimates in the models likely reflects baseline isotopic variation not trophic position differences. Adjusting for the influence of diet suggests

that if Arctic ringed seal diets become more like sub-Arctic seals due to climate change, diet-driven increases may occur for newer contaminants like PFASs, but not for more legacy contaminants. Nonetheless, temporal trends studies are still needed, as are investigations into the potential confounding influence of baseline isotope variation in spatial studies of contaminants in Arctic biota.

4.2 Introduction

Climatic changes to Arctic habitats have given rise to widespread changes in Arctic ecosystems (IPCC 2021), including northward invasions of southern species into Arctic marine food webs, from primary producers to top predators (Ferguson et al. 2010, Oziel et al. 2020). Forage fish, capelin (*Mallotus villosus*) and sand lance (*Ammodytes spp.*), have expanded their temperate-boreal ranges into sub-Arctic and to a lesser extent Arctic Canada, possibly replacing a keystone species, Arctic cod (*Boreogadus saida*) (Provencher et al. 2012, Falardeau et al. 2017). Changes in forage fish communities impact the prey available for Arctic piscivorous fish, seabirds, whales, and seals, including the ringed seal (*Pusa hispida*) (Chambellant 2010). Changes in ringed seal diet in response to warming temperatures have been previously documented in the Canadian Arctic. Increased niche widths in ringed seals from the 1960s to 2000s across the Beaufort Sea suggest diversification of ringed seal diet possibly caused by the range expansion of sub-Arctic prey (Boucher et al 2020). In lower latitudes, higher nitrogen stable isotopes ($\delta\,^{15}$N) linked to warm years in Western Hudson Bay, were suggested to be indicative of greater consumption of capelin (Young & Ferguson 2014). These climate-induced alterations within the Arctic food web have potential knock-on effects on contaminant exposures for Arctic biota such as the ringed seal.

Ringed seals are a high trophic level species of cultural, nutritional, and economic value to northern communities, making them a key biomonitoring species for contaminants across the Canadian Arctic (Houde et al. 2017, Houde et al. 2019). Extensive suites of legacy and emerging persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethanes (DDTs), chlordanes (CHLs), hexachlorocyclohexanes (HCHs), polybrominated diphenyl ether (PBDE) flame retardants, per-/polyfluoroalkyl substances (PFAS) and trace metals have been documented in ringed seal tissues across the Canadian Arctic and over the last 40 years (Gaden et al. 2012, Houde et al. 2019, Houde et al. 2020). Concentrations of legacy POPs and total mercury (THg) in Canadian Arctic ringed seals have generally decreased from 1972-2016 (POPs) and from 2003-2015 (THg in muscle) (Houde et al. 2019, Yurkowski et al. 2020). However, increasing trends have been documented in ringed seals for specific contaminants including PBDE flame retardants, and PFAS such as perfluoroalkyl carboxylates (PFCAs) and perfluoroalkyl sulfonates (PFSAs) (Muir et al. 2019, Addison et al. 2020).

The ringed seal may also be a valuable species for further investigating the impacts of climate change-modulated feeding shifts on contaminant dynamics in Arctic marine food webs (Borgå et al. 2022). Contaminant-climate relationships in Canadian Arctic ringed seals have been reported, with climate parameters including oscillation indexes and sea ice cover correlated with legacy POPs (Gaden et al. 2012, Houde et al. 2019). Temporal variation in THg in Canadian Arctic ringed seals have been accounted for by seal age, climate and diet variables (Houde et al. 2020). Diet has also been shown to influence cadmium (Cd) levels in Eastern Canadian Arctic ringed seals with high Cd levels found in seals feeding on lower trophic level prey (Brown et al. 2016). Consumption of northward range-shifting species may also influence contaminant concentrations in ringed seals, as higher concentrations of ΣPCB, dieldrin, ΣDDT and ΣCHL were found in invading capelin and sand lance than in native Arctic cod from Arviat, Resolute Bay, and Clyde River, NU (Pedro et al. 2017). While POP levels in ringed seals across the Arctic have been documented (Houde et al. 2019), to date, no studies have combined ringed seal spatial contaminants trends with spatial feeding behaviour trends to better understand contaminant-climate dynamics in the Canadian Arctic. Thus, it is unclear what future contaminant loads will look like in Canadian Arctic ringed seals experiencing climate-driven dietary shifts.

While it is challenging to predict climate change-contaminant interactions, one approach, which is often used in climate change community ecology research, in addition to manipulative experiments and long-term monitoring, is "space-for-time" substitution (Blois et al. 2013). Substituting spatial comparisons for temporal patterns may be a reasonable alternative when long-term temporal data sets are unavailable and/or to complement the interpretation of long-term datasets (Hallanger et al. 2011). Thus, insight into the impacts of future climate-driven feeding changes on contaminant exposures in biota may be gained by comparing current spatial differences in feeding patterns and their impacts on spatial variation in contaminant concentrations in ringed seals at sites of different latitudes. Supporting this approach, ringed seal feeding differences between the Arctic (> 66°N) and sub-Arctic (50-66°N) have already been documented (Yurkowski et al. 2016). One approach to analyzing the ringed seal diet is through fatty acid (FA) analysis. Many FAs consumed by monogastric predators, such as ringed seals, are deposited into lipid storage tissues like blubber with little or predictable modification (Budge et al. 2006). Thus, FA signatures of the predator reflect those of their prey, providing insight into predator feeding patterns. Nitrogen and carbon ($\delta^{13}C$) stable isotopes (SI) can also provide information about diet in pinnipeds; whether seal diet is primarily from benthic/inshore prey or from pelagic/offshore prey can be inferred through $\delta^{13}C$ values, and $\delta^{15}N$ values increase with trophic position (Hobson et al. 1997).

The objective of the present study was to develop new insight into how climate change-driven shifts in marine prey affect the tissue levels of an extensive suite of POPs (PCBs, OCs (organochlorine pesticides), PBDEs, PFAS, Mirex, Dieldrin) and trace metals (Hg, Cd) in ringed seals. To do so, we (1) compared feeding patterns of ringed seals at two Canadian low latitude sub-Arctic sites (Nain at 56.5°N, Arviat at 61.1N) and two high latitude Canadian Arctic sites (Sachs Harbour at 72.0 °N, Resolute Bay at 74.7 °N) using FAs as dietary tracers; (2) tested the extent to which contaminant concentrations varied between sub-Arctic and Arctic sites in relation to spatial differences in ringed seal feeding patterns; (3) used adjusted models to suggest future contaminant concentration changes of Arctic ringed seals if their diets become similar to seals in the sub-Arctic.

4.3 Materials and Methods

Sample details and study sites

Thirty-eight ringed seal samples were collected in 2018 by Inuit hunters during routine subsistence harvests in the Canadian sub-Arctic communities of Nain, Nunatsiavut (n=10) and Arviat, Nunavut (n=9) from August to November and in the Arctic communities of Resolute Bay, NU (n = 9) and Sachs Harbour, Northwest Territories (n=10) from June to August (Figure 1). Local hunters and trappers' groups (for Resolute Bay, Sachs Harbour, Arviat), the Nunatsiavut government (for Nain), and hunters led the collection of samples using sampling kits and instructions supplied by government partners. Date of collection, sex (determined by the hunter and confirmed by DNA markers), age (canine tooth growth layer groups), and blubber thickness (depth at sternum) as a body condition metric were recorded (Table S4.1) (see Houde et al 2019 for more details on the sex and age determination method). Collected blubber, liver, and muscle samples were stored frozen between -15 to -35 °C.

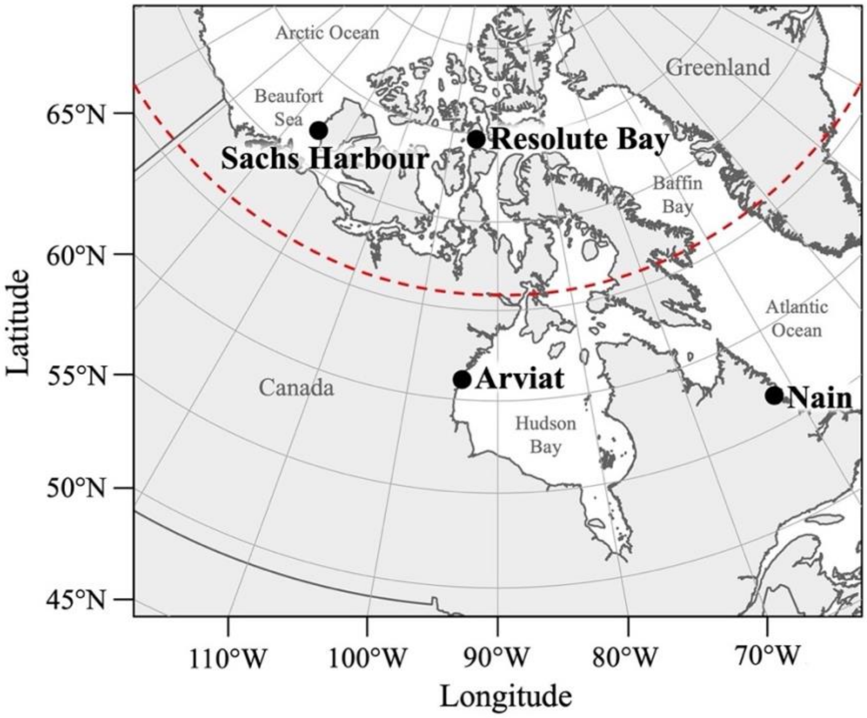

Figure 4.1 Sampling locations of ringed seals in the Canadian sub-Arctic (Arviat and Nain) and Arctic (Sachs Harbour and Resolute Bay) collected in 2018. The red dashed line at 66°30′ N identifies the start of the Arctic circle.

Dietary tracer analysis

A full-depth subsample of the core blubber was used to avoid any oxidized tissue. This approximately 0.5 g subsample was accurately weighed and then analyzed for FAs according to established protocols (Budge et al 2006) (see Supplemental Information for details). The following notation was used to represent each FA, A:Bn-X, with carbon chain length (A), the number of double bonds (B) and the position of the first double bond starting from the methyl group (X).

For $\delta^{15}N$ and $\delta^{13}C$, samples were freeze-dried, homogenized (without lipid extraction) and 500µg of each sample was weighed into tin capsules and then analyzed using a continuous flow isotope ratio mass spectrometer (Finnigan MAT Delta^{Plus}; Thermo Finnigan) at the University of Waterloo Environmental Isotope Lab (Houde et al. 2017).

Contaminant Analyses

Muscle THg concentrations were analyzed by the National Laboratory for Environmental Testing (ECCC, Burlington, ON, Canada) using a Direct Mercury Analyzer (DMA; Milestone Inc., Shelton, CT, USA) and the U.S. Environmental Protection Agency Method 7473 (US EPA 1998). PFASs including C_6 to C_{14} perfluorocarboxylates (PFCAs) and C_4 to C_{12} perfluoroalkane sulfonates were determined in the seal liver as described in Lescord et al. (2015). Blubber was analyzed for suites of 49 PCB congeners, 37 individual OCs and 25 individual or coeluting PBDEs by the Quebec Laboratory for Environmental Testing (ECCC, Montreal, QC) (Full list of analytes provided in Table S4.2). Samples were analyzed for each contaminant group by gas chromatography with high-resolution mass spectrometry (GC-HRMS) (Waters AutoSpec Premier). See supplemental information for more details on Cd, PFAS, PCB, OC and PBDE contaminant analysis.

Quality Assurance and Control

Details of quality assurance and control for all FA, contaminant and stable isotope analyses are found in the Supplemental Material (including Tables S4.3 and S4.4). The mean percent difference of all duplicate runs for the FA and contaminant analysis ranged from 0.5-11.7%. Standard reference materials were run for FAs, Cd, THg and PFAS. For Cd and PFAS blanks were below the detection limit and were not corrected. For PCBs, OCs and PBDEs results were blank- and recovery – corrected and all concentrations were reported on a lipid weight basis.

Data Analysis

Of the 70 identified FAs monitored, 33 were considered to be largely of dietary origin (raw data found in Supplemental Material Table S4.5) (Iverson et al. 2004), and thus were retained for subsequent analysis of feeding patterns. Hierarchical cluster analysis (HCA) and principal component analysis (PCA) were used to visualize the variation in FAs across locations. The FA proportions were first log-ratio transformed using 18:0 as a reference FA (Budge et al. 2006). To ensure the reliability and reproducibility of the PCA, more samples than variables (FAs) are required and a sample-to-variable ratio of 5:1 is recommended (Budge et al. 2006). Thus, the eight highest proportion dietary FAs were selected: 22:6n3, 20:5n3, 20:1n9, 22:1n11, 18:2n6, 18:4n3, 20:1n7 and 16:3n6. The FA 20:1n11 was excluded as it has been described as an unreliable dietary indicator (Thiemann et al. 2021), and the next highest proportioned FA was used instead. The PCA was performed using the *FactorMineR* package in R (Le et al. 2008). A permutational multivariate analysis of variance (PERMANOVA) with the Euclidian distance method was used to test for differences in the eight selected fatty acids between the four locations using the ‘adonis’ function in the *vegan* package (Oksanen et al. 2007). An analysis of the homogeneity of group dispersions was performed to ensure the assumptions of the PERMANOVA were met. This was followed by eight *post-hoc* one-way analyses of variances (ANOVA) and Tukey pairwise comparisons adjusted for multiple comparisons. Three ANOVAs were also performed to evaluate differences in $\delta^{15}N$, $\delta^{13}C$, and body condition between the four locations, followed by a *post-hoc* Tukey test to perform pairwise comparisons between locations. All ANOVAs were checked with diagnostic plots. Although $\delta^{15}N$ and $\delta^{13}C$ are useful tools for studying diet and food webs, spatial trends of these isotopes may be influenced by baseline differences in the food web (De La Vega et al. 2019, De La Vega et al. 2021). Baseline data for $\delta^{15}N$ and $\delta^{13}C$ at each study location is currently unavailable, thus, these values were not baseline corrected. A second PCA was similarly performed that included previously published FA signatures from potential ringed seal prey (Pedro et al. 2020) to visualize similarity between prey and ringed seal dietary FAs. For the HCA, the clustering analysis was performed using the *dendextend* package in R (Galili 2015) using logit-transformed FA proportions.

To test for an effect of feeding patterns on variation in contaminant concentrations, we examined the following seventeen contaminants/groups: ΣPCBs, Σ_{10}PCBs (congeners CB 28, 31, 52, 99, 101, 105, 118, 138, 153, 180)), PCB153, ΣDDT, ΣCHL, ΣHCH, hexachlorobenzene (HCB), mirex, dieldrin, ΣPBDE, ΣPFSA (perfluorosulfonic acids), ΣPFCA, PFOS (perfluorooctane sulfonate), PFOA (perfluorooctanoic acid), THg, and Cd (Tables S4.6 and S4.7). Contaminant concentrations were first tested for normality using the Shapiro-Wilk test and were log(x+1) or sqrt(x) transformed if the transformation improved their distribution. A linear regression model (LM) was used to test the effects of six independent variables: FA PC scores 1 and 2, $\delta^{15}N$, $\delta^{13}C$, body condition, location (Arviat, Nain, Resolute Bay, Sachs Harbour) and the interaction between FA PC scores and location on ringed seal contaminant concentrations. The FA PC 1 and FA PC2 variables are the PC scores from the ringed seal FA PCA from the first objective. PC 1 and PC 2 were selected as they explain the most variation in the PCA. Regression diagnostic plots were run for each model to ensure the LM assumptions were met. Every possible combination of independent variables was run for each contaminant using the

MuMIn package in R (Bartoń 2020). The top models with an Akaike's information criterion corrected for small sample size (AICc) <2 was then selected and averaged to produce the top average model for each contaminant. Although age and sex are important variables explaining contaminant concentrations in ringed seals (Muir et al. 2000), the low sample size of male seals and juvenile seals (seals between two and five years old) for each location prevented these variables from being included. Due to missing contaminant data, only 36 seals were included in the THg model, n=28 for the Cd, n= 36 for the PCB models, n=37 for the PBDE and OC models, and n=28 for the PFAS models.

Finally, to determine the extent to which contaminant concentrations vary between sub-Arctic and Arctic sites due to differences in feeding patterns, we compared the least square means of the location variable (at each location) before and after adjusting for the influence of diet (using FA PC scores 1 and 2, $\delta^{15}N$, $\delta^{13}C$, and/or body condition variables) (See similar approach in McKinney et al. 2011). Using this approach, the post-adjustment least-square mean values of the location variable represent the contaminant levels of the seals from each location if their diets were all the same (i.e. no influence of diet). We then compared the post-adjusted least square means from each location to the pre-adjusted least square means to infer how the diet is influencing the contaminant concentrations of the ringed seals at each location. In performing the adjustment, only contaminants with models from objective two that had one or more significant stable isotope or significant fatty acid variables were selected. The pre-and post-adjusted least-square means of location were calculated using the *emmeans* package (Lenth 2022). One outlier from Sachs Harbour was removed from all data analysis based on Rosner's outlier test. All statistical analyses were performed using R (Version 4.0.2).

4.4 Results

Geographical variation in ringed seal dietary patterns

Ringed seal FA signatures significantly differed between sites ($R^2 = 0.50$, $F_{3,33} = 11.11$, $p < 0.001$) (Table 4.1) and generally clustered by location (Figure S4.1). The FA signatures were separated in the PCA between the sub-Arctic and Arctic locations along the first principal component (PC1), on which most of the Arctic seals loaded positively, while most sub-Arctic seals loaded negatively (Figure 4.2A). This separation along PC1 explained 57.3% variation, with FAs 16:3n6, 20:5n3, 20:1n9, 20:1n11 and 20:1n7 contributing most to this variation and loading positively (Figure 4.2B). Arviat had significantly lower proportions of 16:3n6 ($p < 0.001$) compared to both Arctic locations, as well as significantly lower proportions of 20:1n9 ($p < 0.001$), 20:1n11 ($p < 0.001$) and 20:1n7 ($p = 0.001$) than Resolute Bay (Table 1). Nain had significantly lower proportions of 16:3n6 ($p < 0.001$) and 20:1n7 ($p < 0.001$) compared to Resolute Bay, but no significant differences in those five FAs compared to Sachs Harbour (Table 4.1). Arviat had significantly lower proportions of 20:5n3 compared to Nain ($p = 0.003$) and Resolute Bay ($p = 0.027$). Ringed seals at each location also showed some separation along PC2 in the PCA, with the Arctic seals from Resolute Bay loading mostly negatively and the sub-Arctic seals from both Arviat and Nain loading more positively (Figure 4.2A). Fatty acids 18:2n6, 18:4n3 and 22:6n3 contributed most to variation on PC2, loading positively and representing 29.8% of the PCA variation (Figure 4.2B). Both sub-Arctic locations had significantly higher proportions of 18:2n6 ($p < 0.001$) and 18:4n3 (Arviat: $p = 0.002$; Nain: $p < 0.001$) compared to the Arctic locations (Table 1). While for Arviat, 22:6n3 was significantly higher than at the Arctic locations ($p < 0.001$), for Nain 22:6n3 was lower ($p = 0.02$) compared to Sachs Harbour.

In the PCA including both ringed seals and previously published FA data from their potential prey (see Figure S2 for data and references), sand lance overlapped with ringed seals from Arviat, and capelin showed large overlap with ringed seals at both sub-Arctic sites, while none of the Arctic fish or Arctic amphipod species overlap with the sub-Arctic seals. Sand lance and capelin also loaded positively along PC2 along with sub-Arctic seals. However, there was no overlap of sand lance with Resolute Bay and Sachs Harbour ringed seals, and much smaller overlap of capelin with the Arctic seals. Arctic species including Arctic cod and amphipods loaded negatively along PC2 with slight overlap with the Arctic seals.

Table 4.1 Blubber depth, carbon (δ^{13}C) and nitrogen (δ^{15}N) stable isotopes values, and proportions of eight major fatty acids (FAs) (mean and range) measured in ringed seals from two Canadian sub-Arctic and two Arctic locations in 2018.

	Arviat	Nain	Resolute Bay	Sachs Harbour
Location	Sub-Arctic	Sub-Arctic	Arctic	Arctic
n	9	10	9	9
Blubber depth (cm)	5.8^{a} (4.0 to 8.0)	5.5^{a} (4.4 to 7.6)	4.5^{a} (2.5 to 6.0)	2.5^{b} (1.5 to 3.8)
δ^{13}C (‰)	-22.7^{a} (-25.2 to -20.2)	-18.9^{b} (-19.4 to -18.3)	-20.0^{b} (-21.8 to -19.3)	-21.6^{a} (-22.4 to -21.2)
δ^{15}N (‰)	15.6^{a} (15.0 to 16.1)	13.6^{b} (12.8 to 15.2)	16.1^{a} (15.1 to 16.8)	17.1^{c} (16.0 to 17.8)
FAs (%)				
16:3n6	0.40^{a} (0.29 to 0.76)	0.65^{b} (0.53 to 0.77)	0.84^{c} (0.73 to 1.03)	0.60^{b} (0.43 to 0.72)
18:2n6	2.21^{a} (1.89 to 2.48)	1.38^{b} (1.06 to 1.76)	0.84^{c} (0.67 to 1.01)	1.03^{c} (0.79 to 1.66)
18:4n3	1.52^{a} (0.85 to 2.26)	1.65^{a} (0.97 to 2.23)	0.72^{b} (0.46 to 0.92)	0.92^{b} (0.74 to 1.18)
20:1n11	0.79^{a} (0.39 to 1.18)	1.51^{bc} (0.84 to 2.46)	2.14^{b} (1.60 to 2.64)	1.22^{c} (0.45 to 2.24)
20:1n9	4.46^{a} (3.00 to 6.03)	7.10^{bc} (3.67 to 10.68)	8.76^{b} (5.91 to 10.95)	5.61^{ac} (3.55 to 9.50)
20:1n7	0.51^{a} (0.28 to 0.72)	0.47^{a} (0.30 to 0.66)	0.87^{b} (0.52 to 1.10)	0.52^{a} (0.26 to 0.93)
20:5n3	8.46^{a} (6.60 to 13.88)	10.30^{b} (9.31 to 11.33)	9.76^{b} (7.50 to 11.30)	8.82^{ab} (7.07 to 12.57)
22:6n3	13.51^{a} (11.04 to 15.76)	9.40^{b} (7.81 to 12.00)	8.15^{b} (7.50 to 9.73)	11.26^{c} (10.42 to 13.23)

Significant differences between locations are represented by different letters (^{a,b,c}) above the measurement.

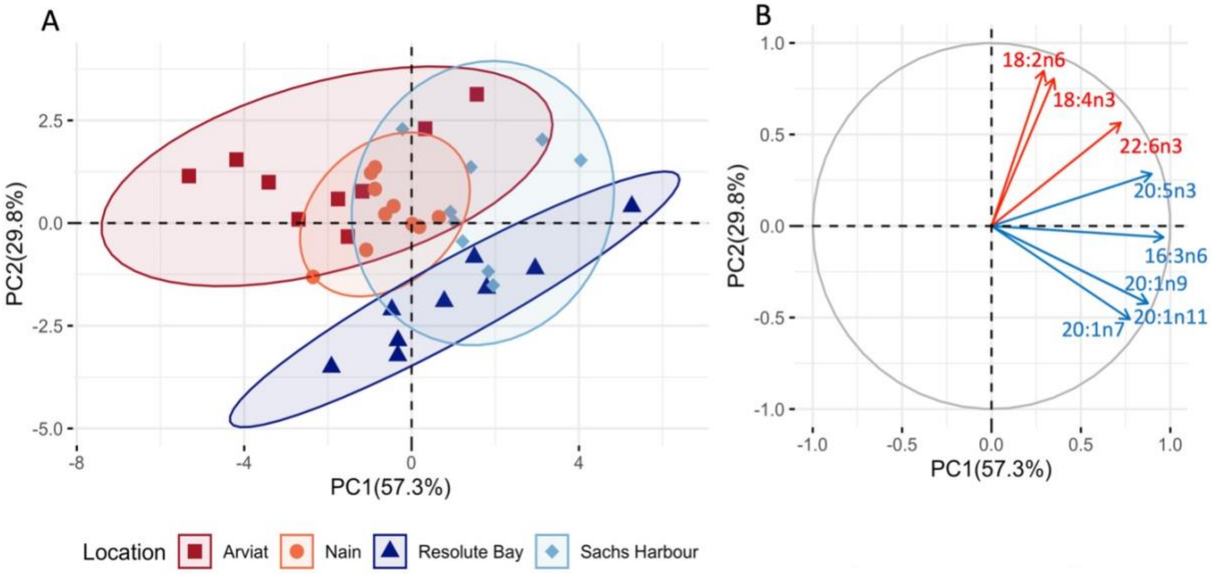

Figure 4.2 A: Principal component analysis of the eight highest proportion dietary fatty acids (22:6n3, 20:5n3, 20:1n9, 22:1n11, 18:2n6, 18:4n3, 20:1n7, 16:3n6) in ringed seals sampled in 2018 from Arviat (red) and Nain (orange), and Resolute Bay (blue) and Sachs Harbour (pale blue), in the Canadian sub-Arctic and Arctic, respectively. The first dimension of the PCA accounted for 57.3% of the total variation, and the second dimension accounted for 29.8%. Ellipses represent 90% confidence intervals. B: Variable correlation plot of the eight fatty acids used in the PCA. The vectors in blue represent the FAs that contributed above the expected average (12.5%) to PC1, while the red vectors represent the FAs that contributed above the expected average (12.5%) to PC2. The average contribution of the variable is calculated by 1/length(variables).

Both $\delta^{13}C$ ($p < 0.001$) and $\delta^{15}N$ ($p < 0.001$) significantly differed between locations (Table 4.1 and figure S4.3). The $\delta^{13}C$ values were significantly higher in Resolute Bay than Arviat and significantly higher in Nain compared to Sachs Harbour. For $\delta^{15}N$, values were significantly higher in Sachs Harbour compared to Arviat and Nain and significantly higher in Resolute Bay than Nain. Body condition showed significant differences between locations ($p < 0.001$), with Sachs Harbour seals showing significantly lower values than elsewhere.

Geographic variation in contaminant concentrations related to dietary tracers

The effects of FA-PC1, FA-PC2, $\delta^{15}N$, $\delta^{13}C$, body condition, and location on contaminant concentrations were tested using generalized linear models (Tables 4.2 and 4.3). For muscle THg and liver Cd concentrations, the best-averaged models explained over 50% of contaminant variation (Table 4.2). Top models for THg explained 67% of the variance and the strongest predictor variable was $\delta^{15}N$, followed by FA-PC2 (confidence intervals not overlapping zero). The predictor FA-PC1 was also included in the top THg models but was not considered important as the confidence interval overlapped zero. For Cd, the top models explained 51% of the variance, with location being the strongest predictor. The Cd models also included $\delta^{13}C$ but with confidence intervals overlapping zero, suggesting limited importance. For PFAS, the top models included body condition, $\delta^{15}N$, and $\delta^{13}C$, except in the case of PFOA (Table 4.2). Together, these variables explained over 60% of PFAS variation with stable isotopes and body condition significantly explaining concentrations of ΣPFSA and PFOS, and stable isotopes significant for ΣPFCA concentrations. For all PFAS excluding PFOA, $\delta^{15}N$ had a positive estimate, and $\delta^{13}C$ had a negative estimate. The top models explained 79% of PFOA variation with significant negative estimates for $\delta^{15}N$, and location estimates significantly contributing to the variation. FA-PC1 and $\delta^{13}C$ also appeared in the top averaged PFOA model, although the estimates were small compared to $\delta^{15}N$ and location, and confidence intervals overlapped zero.

Body condition was the only significant variable explaining PCB153, ΣPCB and Σ_{10}PCB concentrations, and the overall variance explained (R^2) in the PCB models was low (Table 4.3). The relationship between body condition and PCB concentrations was negative. Although $\delta^{15}N$ appeared in the PCB153 model, the confidence intervals overlapped zero, $\delta^{15}N$ was not considered an important variable. For ΣPBDE, only 26% of variation was explained by the top models, and concentrations were significantly explained only by $\delta^{13}C$ values. Although FA-PC2, $\delta^{15}N$, and location did appear in the best-averaged model for ΣPBDE concentrations, their confidence intervals overlapped zero. For OC concentrations, location, $\delta^{13}C$, and δ ^{15}N consistently appeared in the models, with FA-PC1 and FA-PC2 scores also appearing in all models except for dieldrin and mirex. Generally, OC models were well explained with 67% or higher explained variation for ΣCHL, HCB, dieldrin, and ΣHCH. Concentrations of ΣDDT, ΣHCH, Mirex models were positively associated with $\delta^{13}C$ values, while ΣHCH and HCB concentrations were negatively associated with $\delta^{15}N$ values. The model for ΣHCH also showed a significant positive effect for FA-PC2, while FA-PC1 appeared to be negatively related to ΣDDT, ΣCHL, ΣHCH and HCB, but FA-PC1 was not significant in these models.

Table 4.2 Best averaged model (all models with an AIC_c <2 were averaged) of generalized linear models for contaminants found in the muscle (THg) and liver (Cd, Pb, ΣPFSA, ΣPFCA, PFOS, PFOA) of ringed seals sampled in 2018 from Arviat, Nain, Resolute Bay, and Sachs Harbour in sub-Arctic and Arctic Canada.

Model	Parameter	Estimate	Confidence Interval 2.50%	Confidence Interval 97.50%	R^2
THg ~ $\delta^{15}N$ + FA-PC1 + FA-PC2	Intercept	**-3.21**	-4.02	-2.40	0.67
	$\delta^{15}N$	**0.16**	0.11	0.22	
	FA-PC1	0.02	-0.02	0.05	
	FA-PC2	**-0.06**	-0.10	-0.01	
Cd ~ Location + $\delta^{13}C$	Intercept	-2.11	-10.04	5.82	0.51
	Location Nain	-0.75	-2.47	0.96	
	Location Resolute Bay	0.55	-0.76	1.85	
	Location Sachs Harbour	**-1.11**	-2.06	-0.16	
	$\delta^{13}C$	-0.22	-0.57	0.13	
ΣPFSA ~ Body Condition + $\delta^{15}N$ + $\delta^{13}C$	Intercept	**-2.62**	-4.09	-1.15	0.60
	Body Condition	**0.07**	0.01	0.13	
	$\delta^{15}N$	**0.11**	0.02	0.21	
	$\delta^{13}C$	**-0.08**	-0.13	-0.04	
ΣPFCA ~ Body Condition + $\delta^{15}N$ + $\delta^{13}C$	Intercept	**-3.84**	-5.27	-2.42	0.74
	Body Condition	0.03	-0.02	0.08	
	$\delta^{15}N$	**0.18**	0.09	0.26	
	$\delta^{13}C$	**-0.10**	-0.15	-0.06	
PFOS ~ Body Condition + $\delta^{15}N$ + $\delta^{13}C$	Intercept	**-2.70**	-4.13	-1.27	0.60
	Body Condition	**0.07**	0.02	0.12	
	$\delta^{15}N$	**0.12**	0.03	0.20	
	$\delta^{13}C$	**-0.08**	-0.12	-0.04	
PFOA ~ $\delta^{15}N$ + FA-PC1 + $\delta^{13}C$ + Location	Intercept	1.79	-0.45	4.03	0.79
	$\delta^{15}N$	**-0.14**	-0.24	-0.03	
	FA-PC1	-0.03	-0.07	0.00	
	$\delta^{13}C$	-0.05	-0.11	0.01	
	Location Nain	**-0.63**	-1.00	-0.25	
	Location Resolute Bay	-0.21	-0.49	0.07	
	Location Sachs Harbour	-0.01	-0.29	0.27	

Significant estimates in bold. THg (total mercury), Cd (cadmium), ΣPFSA (perfluoroalkyl sulfonic acids), ΣPFCA (perfluoroalkyl carboxylic acids), PFOS (perfluorooctane sulfonate), PFOA (perfluorooctanoic acid).

Table 4.3 Best averaged model (all models with an $AIC_c < 2$ were averaged) of generalized linear models for blubber PCB, PBDE, and OC pesticide concentrations in ringed seals sampled in 2018 from Arviat, Nain, Resolute Bay, Sachs Harbour in sub-Arctic and Arctic Canada

Model	Parameter	Estimate	Confidence Interval		R^2
			2.50%	97.50%	
PCB153 ~ Body Condition + $\delta^{15}N$	Intercept	**148.41**	21.86	274.95	0.11
	Body Condition	**-6.13**	-12.23	-0.02	
	$\delta^{15}N$	-3.89	-10.93	3.15	
Σ_{10}PCB ~ Body Condition	Intercept	**250.54**	187.95	313.13	0.20
	Body Condition	**-18.93**	-31.74	-6.12	
ΣPCBs ~ Body Condition	Intercept	**424.33**	319.92	528.74	0.21
	Body Condition	**-32.92**	-54.28	-11.55	
ΣPBDE ~ $\delta^{13}C$ + FA-PC2 + Location + $\delta^{15}N$	Intercept	1.83	-1.85	5.51	0.26
	$\delta^{13}C$	**0.12**	0.01	0.22	
	FA-PC2	0.02	-0.08	0.12	
	Location Nain	-0.10	-0.71	0.52	
	Location Resolute Bay	-0.46	-0.93	0.02	
	Location Sachs Harbour	-0.35	-0.75	0.05	
	$\delta^{15}N$	0.10	-0.09	0.28	
ΣDDT ~ $\delta^{15}N$ + Location + $\delta^{13}C$ + FA-PC1	Intercept	**5.38**	2.20	8.57	0.45
	$\delta^{15}N$	0.03	-0.11	0.17	
	Location Nain	**-0.58**	-1.14	-0.02	
	Location Resolute Bay	-0.16	-0.60	0.27	
	Location Sachs Harbour	0.01	-0.41	0.43	
	$\delta^{13}C$	**0.17**	0.08	0.27	
	FA-PC1	-0.02	-0.07	0.03	
ΣCHL ~ $\delta^{13}C$ + FA-PC1 + Location + $\delta^{15}N$	Intercept	**5.61**	3.23	8.00	0.67
	$\delta^{13}C$	**0.11**	0.04	0.18	
	FA-PC1	-0.03	-0.07	0.01	
	Location Nain	**-0.71**	-1.10	-0.32	
	Location Resolute Bay	0.20	-0.10	0.50	
	Location Sachs Harbour	**0.44**	0.12	0.75	
	$\delta^{15}N$	-0.08	-0.19	0.04	
ΣHCH ~ FA-PC1 + FA-PC2 + $\delta^{15}N$ + $\delta^{13}C$ + Location	Intercept	**5.57**	3.54	7.61	0.84
	FA-PC1	-0.04	-0.08	0.00	
	FA-PC2	**0.07**	0.01	0.14	
	$\delta^{15}N$	**-0.16**	-0.26	-0.05	
	$\delta^{13}C$	**0.09**	0.03	0.15	
	Location Nain	-0.35	-0.70	0.00	
	Location Resolute Bay	**0.83**	0.51	1.15	
	Location Sachs Harbour	**1.05**	0.79	1.31	
HCB ~ FA-PC1 + Location + $\delta^{15}N$	Intercept	**2.24**	1.31	3.17	0.76
	FA-PC1	-0.02	-0.04	0.01	
	Location Nain	-0.16	-0.32	0.00	
	Location Resolute Bay	**0.41**	0.28	0.54	
	Location Sachs Harbour	**0.49**	0.33	0.66	
	$\delta^{15}N$	**-0.08**	-0.14	-0.02	
Mirex ~ Body Condition + Location + $\delta^{13}C$	Intercept	**6.60**	1.93	11.26	0.47
	Body Condition	-0.03	-0.21	0.15	
	Location Nain	**-1.86**	-2.83	-0.90	
	Location Resolute Bay	-0.84	-1.69	0.00	
	Location Sachs Harbour	-0.62	-1.50	0.26	
	$\delta^{13}C$	**0.19**	0.00	0.39	
Dieldrin ~ $\delta^{15}N$ + Location + $\delta^{13}C$	Intercept	**3.48**	1.51	5.44	0.72
	$\delta^{15}N$	**-0.09**	-0.18	-0.01	

	Location Nain	-0.12	-0.46	0.22
	Location Resolute Bay	**0.53**	0.29	0.76
	Location Sachs Harbour	**0.51**	0.30	0.72
	$\delta^{13}C$	0.04	-0.02	0.10

Significant estimates in bold. ΣPCB (polychlorinated biphenyls), Σ_{10}PCB (ten major PCB congeners), ΣPBDE (polybrominated diphenyl ethers), ΣDDT (dichlorodiphenyltrichloroethanes), ΣCHL (chlordane), ΣHCH (hexachlorocyclohexanes), HCB (hexachlorobenzene).

Influence of dietary patterns on geographic variation in contaminant concentrations

To investigate the influence of feeding patterns on contaminant concentration variation among locations, we calculated ringed seal contaminant trends before and after adjusting for diet variables (Figures 4.3 and 4.4). Before adjustment, the Arctic ringed seals showed the highest THg concentrations, while the sub-Arctic ringed seals showed the lowest concentrations (Figure 4.3). After adjustment, THg concentrations for all sites became more similar ranging between 0.15 and 0.25 mg/kg wet weight (w.w.). The ΣPFSA and PFOS concentrations were highest in Arviat and lowest in Nain before adjustment. After adjustment, concentrations for the sub-Arctic sites became similar (~20 ng/g w.w.), while the Arctic site concentrations did not change much after adjustment, staying at ~10 ng/g w.w. The adjustment did not change PFOA and ΣPFCA concentrations substantially for any of the sites (Figure 4.3).

For legacy POPs (Figure 4.4), ΣDDT concentrations in sub-Arctic ringed seals sites were similar before adjustment. Concentrations of ΣDDT in Arviat ringed seals increased from 100 ng/g l.w. to 250 ng/g l.w. after adjustment, while Nain decreased from 100 ng/g l.w. to < 50 ng/g l.w. Similar results were seen for ΣCHL concentrations, where those in Arviat seals increased after adjustment and in Nain seals decreased. Both ΣDDT and ΣCHL concentrations in Arctic ringed seals did not vary considerably after adjustment. ΣPBDE showed similar patterns to ΣDDT and ΣCHL, although concentrations did not differ substantially after adjustment. Concentrations of ΣHCH, HCB, and dieldrin in the sub-Arctic ringed seals showed lower concentrations both before and after adjustment for diet compared to the Arctic seals. Nain concentrations decreased after adjustment for all three contaminants, while Sachs Harbour concentrations increased after adjustment for HCB and ΣHCH.

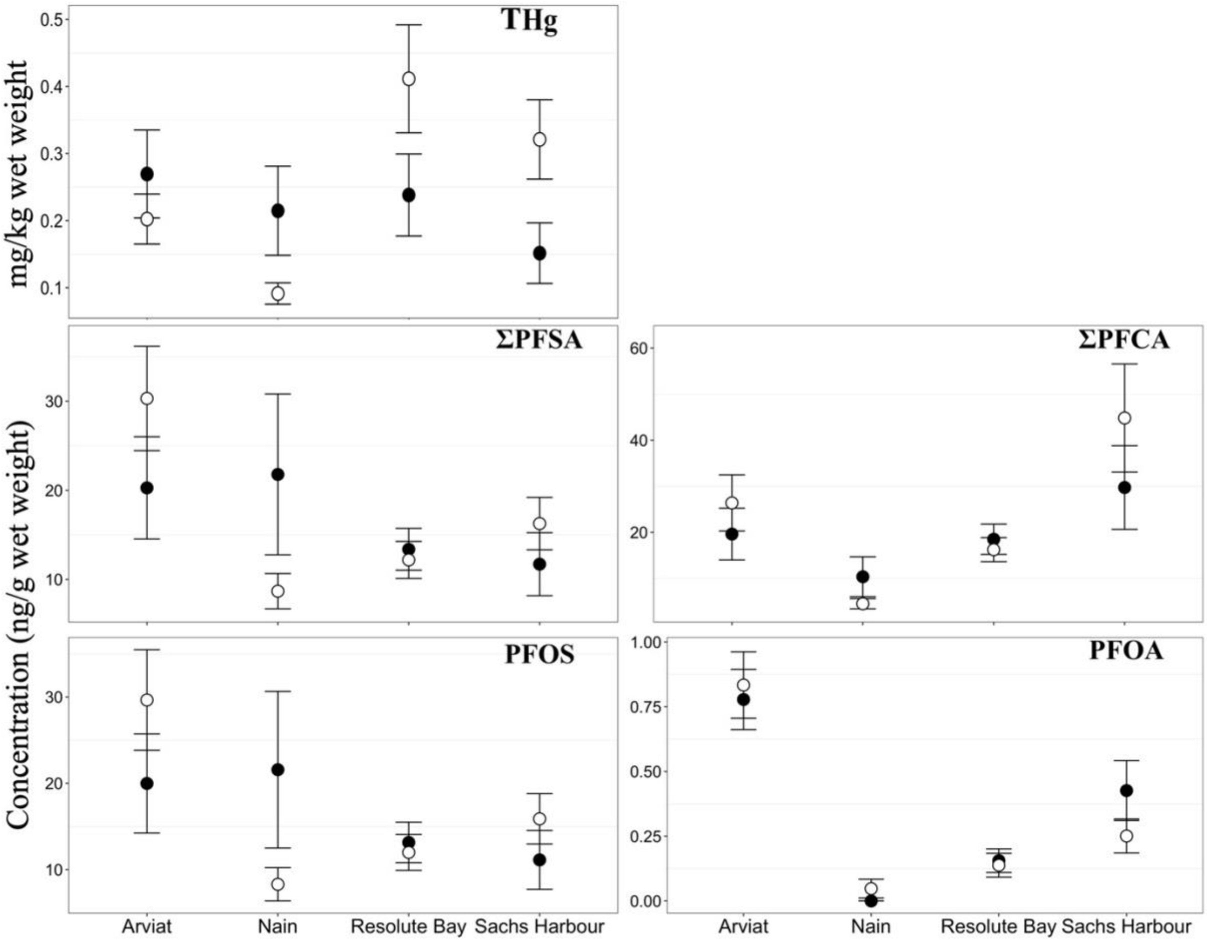

Figure 4.3 Least squares mean concentrations of muscle THg (total mercury) and liver ΣPFSA (perfluoroalkyl sulfonic acids), ΣPFCA (perfluoroalkyl carboxylates), PFOS (perfluorooctane sulfonate) and PFOA (perfluorooctanoic acid) (± standard error) in ringed seals collected in 2018 before (○) and after (●) adjusting for the following variables FA-PC1, FA-PC2, $\delta^{13}C$, $\delta\,^{15}N$, and body condition across sub-Arctic (Arviat and Nain) and Arctic (Resolute Bay and Sachs Harbour) locations.

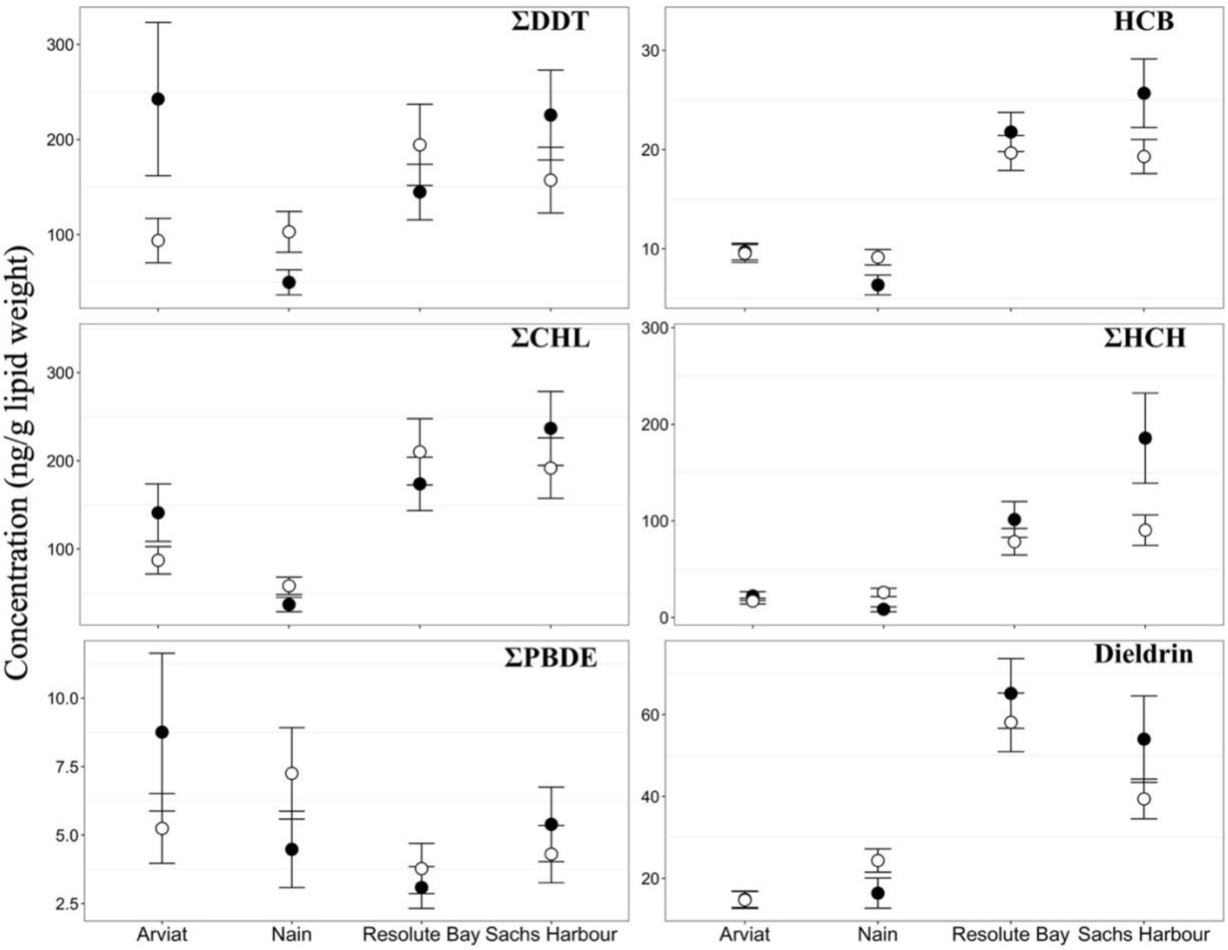

Figure 4.4 Least squares mean concentrations of blubber ΣDDT (dichlorodiphenyltrichloroethanes), HCB (hexachlorobenzene), ΣCHL (chlordane), ΣHCH (hexachlorocyclohexanes), ΣPBDE (polybrominated diphenyl ethers) and dieldrin (± standard error) in ringed seals collected in 2018, before (○) and after (●) adjusting for FA-PC1, FA-PC2, $\delta^{13}C$, $\delta^{15}N$, and body condition across sub-Arctic (Arviat and Nain) and Arctic (Resolute Bay and Sachs Harbour) locations.

4.5 Discussion

Geographical variation in ringed seal dietary patterns

The differences in ringed seal FA signatures between locations in this study were consistent with previous reports documenting spatial differences in feeding habits of Canadian Arctic ringed seals (Thiemann et al. 2007, Chambellant et al. 2013, Yurkowski et al. 2016). Specifically, the FA signatures in Arviat and Nain seals were nearly entirely overlapping, suggesting similar diets at the two sub-Arctic locations, and were separated from the Resolute Bay seals, suggesting feeding differences between the sub-Arctic animals and Arctic seals from that region. Collection date likely had little impact on these FA signatures as all ringed seals were collected within the same year and blubber FAs may represent multiple months of feeding information (Budge et al. 2006).

When combined with prey data from Pedro et al. (2020), Arctic ringed seal (Resolute Bay) FA signatures overlapped somewhat with Arctic cod, while sub-Arctic ringed seal FA signatures (particularly Arviat) partially overlapped with sand lance and particularly capelin. These findings suggest higher consumption of sand lance and capelin, and little consumption of Arctic cod, by Canadian sub-Arctic relative to Arctic ringed seals. Similarly, ringed seals sampled between 1991-2006 in Arviat were found to be sand lance specialists (Chambellant et al. 2013), and other reports indicate increasing capelin consumption in Hudson Bay ringed seals (Young & Ferguson 2014). Conversely, stomach contents and stable isotopes of Resolute Bay ringed seals in 2010 showed that Arctic cod was the main prey item (Matley et al. 2015).

The Sachs Harbour ringed seals showed wide variation in FA signatures, overlapping seals from the three other sites, possibly suggesting consumption of both native and sub-Arctic type prey. In this region, ringed seal diets primarily consisted of Arctic cod and invertebrates in the 1980s (Smith 1987); however, and in support of our findings, a shift to greater consumption of sub-Arctic forage fish has recently been documented. Insley et al. (2021) found several fish species, including Arctic cod, sand lance, and capelin in the stomach contents of ringed seals in 2015-2018 in the Amundsen Gulf region including Sachs Harbour. In 2014, stomach contents from belugas (*Delphinapterus leucas*) in the Eastern Beaufort Sea similarly showed primarily sand lance and little Arctic cod (Loseto et al. 2018). An alternative explanation for the wide FA variation in Sachs Harbour ringed seals could be a mixture of both young-of-the-year (YOY) and adult ringed seals in the dataset. Age class and sex influence FA signatures (Thiemann et al. 2007, Ross et al. 2022); however, it was not possible to include these variables in the FA analysis due to small sample sizes of different sex/age classes.

In addition to variation in FA signatures, the ringed seals showed some spatial variation in stable isotope values. Values of δ^{15}N were highest in Sachs Harbour, lower and similar in Arviat and Resolute Bay, and lowest in Nain seals. Ringed seals from Nain showed similarly lower δ^{15}N values than other Canadian Arctic and sub-Arctic sites in a 2002-2005 study (Butt et al. 2008). Variation in δ^{15}N seems unlikely to be largely related to differences in consumption of native versus invading sub-Arctic fish given similar values at Arviat and Resolute, and perhaps is more likely related to spatial variation in isotopic baselines, related to the input of different water bodies (Pacific vs Atlantic) (De La Vega et al. 2021). For δ^{13}C, values were highest in Nain and lower in Sachs Harbour and (although not significantly so) in Resolute Bay, which is consistent with baseline δ^{13}C values declining with increasing latitude (De La Vega et al. 2019) (Takahashi et al. 2002). Arviat seals, however, showed the most depleted δ^{13}C values, which is not consistent with latitudinal baseline variation. Although Arviat δ^{13}C values are generally more depleted due to the large influence of freshwater inputs (De La Vega et al. 2019), Arviat mean δ^{13}C values were 3.6‰ lower than in 2002-2005, seemingly more than the temporal decline at the other three sites of 1.0 to 2.1‰ relative to 2002-2005 (Butt et al. 2008). While the Suess Effect (increasing anthropogenic CO_2 leading to lower atmospheric δ^{13}C over time) may be contributing to declines at all sites, there are likely additional factors at Arviat that explain the greater δ^{13}C drop there, including possibly increasing freshwater inputs or increased consumption of phytoplankton-based carbon versus benthic and sympagic carbon (Yurkowski et al. 2020). We suggest that Arviat seals $\delta^{13}C$ values may be declining more so than elsewhere, at least in part, due to increasing consumption of prey from the phytoplankton carbon pathway, such as capelin and sand lance, two pelagic fish which show more depleted $\delta^{13}C$ values than, e.g., Arctic cod (Pedro et al. 2020).

Diet influence of ringed seal contaminant levels

Diet variables, especially $\delta^{15}N$ and $\delta^{13}C$, explained variation in concentrations of multiple contaminants in the ringed seals. For muscle THg models, $\delta^{15}N$ values were significant and positive. Although this could be related to THg levels being associated with higher trophic position (Brown et al. 2016, Houde et al. 2020), it could also be connected to spatial variation in baseline $\delta^{15}N$ as already discussed. FA-PC2 had a negative estimate suggesting that feeding on prey with lower proportions of FAs loading on PC2, such as Arctic cod, increased THg levels. Consistent with this, Arctic cod have shown higher THg concentrations than capelin and sand lance (Pedro et al. 2017, Pedro et al. 2019). For liver Cd concentrations, location was the only significant variable. Cd concentrations were higher in adult versus sub-adult ringed seals in an earlier study (Brown et al. 2016), suggesting that the current Cd findings may be related to variation in seal ages among locations. For liver PFAS, $\delta^{15}N$ and $\delta^{13}C$ were the main significant diet-related variables. Values of $\delta^{13}C$ had negative estimates for ΣPFSA, ΣPFCA, PFOS and PFOA, suggesting that feeding on pelagic and/or less ice-associated prey, such as capelin and sand lance, increases the concentrations. Consistent with this, capelin also showed higher concentrations of current-use pesticides than multiple native Arctic fishes, possibly related to seasonal migration into temperate regions (Morris et al. 2016). For ΣPFSA, ΣPFCA and PFOS, $\delta^{15}N$ had a positive relationship, which may again be related to spatial variation in baseline $\delta^{15}N$

(De La Vega et al. 2021), as well as biomagnification of most perfluoroalkyl acids in global marine food webs (Miranda et al. 2022). Liver concentrations may also not be representative of whole-body concentrations, as varying concentrations of PFAS have been documented in seals depending on the type of tissue (kidney, liver, blubber, muscle, spleen) selected for analysis(Van de Vijver et al. 2005).

The leading averaged models for PCBs included only body condition. The negative estimate of body condition is likely related to PCB concentrations increasing in ringed seal blubber as the pool of lipid decreases with lower body condition (Muir et al. 2000). For the other lipophilic contaminants, $\delta^{13}C$ was often the main diet-related variable, however, the relationship was opposite to what was found for PFAS: $\delta^{13}C$ had a significant positive estimate for ΣPBDE, ΣDDT, ΣCHL, ΣHCH and Mirex, suggesting that ice-associated or benthic feeding/habitats increased ringed seal legacy POP concentrations. This is consistent with high levels of these same contaminants found in benthic sculpin (*Myoxocephalus spp.*) and northern shrimp (*Pandalus borealis*) compared to pelagic prey fish (Pedro et al. 2017). However, in that study capelin tended to have higher levels of OCs than Arctic cod. $\delta^{15}N$ was not a consistently significant variable, but when it was (ΣHCH, HCB and dieldrin), the $\delta^{15}N$ estimate was negative. On the surface, these findings would suggest that individuals feeding at higher trophic levels have lower concentrations of these contaminants, which is inconsistent with most previous studies reporting their biomagnification in Arctic food webs (Fisk et al. 2001). Although PFAS are protein bound contaminants and legacy POPs are lipophilic, the inconsistent directionality of the $\delta^{15}N$ and $\delta^{13}C$ relationships between the legacy POPs and the PFAS seems to support an alternative interpretation, i.e., that contaminant relationships to isotope variation, at least for $\delta^{15}N$, are possibly driven more by baseline variation in isotope values than by trophic position differences of the ringed seals (Schell et al. 1998, De La Vega et al. 2019, De La Vega et al. 2021). Accounting for baseline isotope variation in future contaminant studies should be prioritized, such as through the development of ancillary compound-specific isotope analysis (CSIA) datasets to disentangle baseline effects from seal trophic position (Elliott et al. 2021). CSIA approaches should be used to further test our interpretations of how feeding differences across the Canadian Arctic contribute to differences in contaminant accumulation in ringed seals.

Out of all the OC models, the only model with significant FA predictors (FA-PC2) was ΣHCH, suggesting higher proportions of 18:2n6, 18:4n3, and 22:6n3 were associated with higher ΣHCH concentrations in ringed seals. Since these FAs were found in higher proportions in capelin and sand lance than in Arctic cod (Pedro et al., 2020), this finding suggests that all else being equal, greater feeding on these sub-Arctic prey is associated with higher ΣHCH concentrations. Location also appeared in most models with relatively high estimates compared to the diet variables. This is likely related to site-specific contaminant levels, as described previously in Houde et al. (2019) and also in Wong et al. (2011) where they found higher ΣHCH levels between 2007-2008 in seawater from the Beaufort Sea compared to Hudson Bay. Thus, many factors can contribute to spatial trends in ringed seal contaminant concentrations in addition to dietary patterns.

Climate-change influence on contaminant accumulation

Climate change can influence POP concentrations in Arctic compartments, including biota (Borgå et al. 2022, Chételat et al. 2022, McKinney et al. 2022). Some of the many ways ringed seals could be affected by climate change is through climate-induced northward range shifts of sub-Arctic prey fish, changing ringed seal feeding ecology and introducing new sources of contaminants. Without temporal data sets of ringed seal feeding and contaminant data, our study is limited in its ability to determine how climate-induced diet changes influence contaminant concentrations in ringed seals. However, using space-for-time substitution, we can make tentative predictions of possible impacts on contaminant concentrations in Arctic ringed seals if they begin to consume higher proportions of sub-Arctic fish species, as is occurring currently in sub-Arctic locations. Due to the known effect of age class on contaminant concentrations, Arviat (sub-Arctic) and Resolute (Arctic) comparisons are discussed below, as both these locations included mainly adult seals, while Nain and Sachs Harbour had more varied age compositions.

Concentrations of THg were higher in Resolute than in Arviat seals, but after adjusting for diet (δ^{15}N and FA-PC2), concentrations became similar. This suggests that THg levels in Resolute Bay seals may decrease if their diets shift towards that of Arviat seals, which includes higher proportions of sub-Arctic prey. This aligns with previous published higher THg levels in Arctic cod than in capelin and sand lance (Pedro et al. 2017). For ΣPFSA and PFOS, concentrations were lower in Resolute Bay than in Arviat seals before adjustment. After adjustment for δ^{13}C, δ^{15}N and body condition, levels of ΣPFSA and PFOS decreased by ~10 ng/g w.w. in Arviat, while Resolute Bay had little change. Thus, higher Arviat concentrations may be partly due to feeding, suggesting that ΣPFSA and PFOS may increase in Resolute Bay seals if diets become more similar to Arviat seals. ΣPFCA and PFOA concentrations did not vary much between Arviat and Resolute seals pre- or post-adjustment, suggesting a limited influence of diet for these contaminants. For ΣDDT, before adjusting for diet (δ^{13}C), concentrations were lower in Arviat than in Resolute Bay seals. Post-adjustment, Arviat levels increased 2.5-fold, becoming higher than levels in Resolute Bay, which is more expected as Arviat is closer to North American sources and inflowing rivers drain into the Hudson Bay with known past uses of DDT (McCrea 1984). Based on the pre-adjustment results, a decrease in ΣDDT levels in Resolute Bay seals could be expected if they shift to a sub-Arctic diet. A similar post-adjustment ($\delta^{13}C$) pattern is seen for ΣPBDE, suggesting a decrease for Resolute seals with a future more sub-Arctic diet, nonetheless, both pre-and-post adjustment ΣPBDE levels remain higher in Arviat. While the influence of diet was found for ΣCHL, HCB, ΣHCH and dieldrin, there was only slight variation in concentrations after adjusting, suggesting location-specific differences in contamination outweighed contaminant variation due to diet differences.

4.6 Conclusions

As previously predicted based on comparisons of contaminant concentrations among Arctic and sub-Arctic prey fishes (Pedro et al. 2017), these space-for-time substitution findings suggest that the shifting distribution of sub-Arctic fish into the Arctic could indeed influence the accumulation of certain contaminants in Arctic ringed seals. The climate-driven shift away from native ice-associated or benthic Arctic prey towards a reliance on pelagic (less-ice-associated) sub-arctic pelagic prey will possibly increase newer POPs, such as PFASs, and decrease more established POPs, such as THg, PBDEs, and DDTs, in Arctic ringed seals. Although speculative, the projected increase in newer POPs could be driven by the more uneven spatial distribution of POPs with more recent/ongoing emissions, whereas since the legacy POPs are perhaps somewhat more evenly distributed throughout the Arctic, their levels may thus be less influenced by shifts in dietary sources. However, it is critical that future studies using chemical tracers such as stable isotopes to track spatial variation in biotic contaminant concentrations further consider how spatial variation in baseline tracer values (related to, e.g., latitude, freshwater inputs, or other biogeochemical factors) can influence interpretations of feeding and food web changes. Future long-term temporal studies, particularly that incorporate CSIA approaches, are necessary to fully understand the extent to which climate change may impact ringed seal diets and contaminant accumulation.

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4.10 Supplemental Information

Dietary tracer analysis

Briefly, lipid was extracted from blubber using the Folch method (Folch et al. 1957). Extracted FAs underwent *trans*-esterification to obtain fatty acid methyl esters (FAMEs) using the Hilditch reagent. FAMEs were then analyzed using an Agilent 8860 gas chromatograph with flame ionization detection (Santa Clara, CA, USA). From each chromatogram, 70 FAMEs were quantified as percent total FAME using Agilent OpenLab CDS Data Analysis software (V. 2.5).

Cd/PFAS/PCB/OC/PBDE Instrumental Analysis

Cadmium in the seal liver was analyzed by the National Laboratory for Environmental Testing (ECCC, Burlington, ON, Canada). Liver samples were digested with nitric acid and hydrogen peroxide (8:1) in a high pressure microwave oven (200 °C; 15 minutes). The digest was then analyzed directly by ICP-MS. Total Hg (THg) in muscle was analyzed at Environment and Climate Change Canada (Burlington, ON, Canada) using a Direct Mercury Analyzer (DMA; Milestone Inc., Shelton, CT, USA) using U.S. Environmental Protection Agency Method 7473 (US EPA 1998).

PFASs including C6 to C14 perfluorocarboxylates (PFCAs) and C4 to C12 perfluoroalkane sulfonates were determined in the seal liver (see Table S3). The methodology followed that described in Lescord et al. (2015). The extraction procedure involved addition of mass-labeled internal standards (^{13}C2 PFCAs and PFOS) and an extraction with acetonitrile followed by carbon cleanup. PFASs in sample extracts were quantified by liquid chromatography with negative electrospray tandem mass spectrometry (LC-MS/MS).

Blubber was analyzed for suites of 49 PCB congeners, 37 individual OCs and 25 individual or coeluting PBDEs by the Quebec Laboratory for Environmental Testing (ECCC, Montreal, QC) (Full list of analytes provided in Table S3). Briefly, samples were extracted using Soxhlet extraction with a 16-hour reflux in acetone:hexane (1:1) v/v, and then run on gel permeation chromatography (GPC) using Biobeads XS3. Lipid percent was determined gravimetrically. Extracts then underwent a two-stage cleanup: a multilayer column clean-up (layers of $AgNO_3$-silica-NaOH-silica-H_2SO_4-silica) followed by either an alumina column (PCBs), a deactivated Florisil® column (PBDEs), or a deactivated silica column with 3% H_2O (OCs). Samples were then analyzed for each contaminant group by gas chromatography with high-resolution mass spectrometry (GC-HRMS) (Waters AutoSpec Premier). The analysis was conducted in EI mode with an electron energy of 35 eV using isotope dilution. For PCBs and OCs, the on-column injection was used, and the chromatographic separation was achieved for both PCBs and OCs, on a DB-5 60m x 0.25mm x 0.25µm (J&W Scientific, Agilent Technologies) column fitted with a 0.53 mm guard column (Restek, Chromatographic Specialties). For PBDEs, the on-column injection was used. The chromatographic separation of 25 native PBDEs was achieved on a DB-5 15m x 0.25mm x 0.1µm (J&W Scientific, Agilent Technologies) column fitted with a 0.53 mm guard column (Restek, Chromatographic Specialties).

Quality Assurance and Control

For the FA analysis, each location was run as a separate batch, and three duplicates were run in total to assess precision. The mean percent difference for the duplicates was 2.3% (range 0.0-11.7%). Any FAs less than 0.1% of the total were not included in further analysis to reduce uncertainty related to analytical variability. The U.S. National Institute of Standards and Technology (NIST) standard reference material NIST1945 whale blubber homogenate was extracted and analyzed with each batch to assess accuracy and showed a mean percent difference of 21% compared to published values (Kucklick et al. 2010).

Stable isotope abundances were expressed in delta (δ) values as the deviation from standards in parts per thousand (‰) using the equation δ_{sample} ‰ = $[(R_{sample}/R_{standard}) — 1] \times 1000$ where R is the ratio of heavy to light isotope ($^{15}N/^{14}N$ or $^{13}C/^{12}C$) in the sample and standard. The nitrogen stable isotope standard was atmospheric nitrogen; Pee Dee Belemnite limestone formation was the standard for the carbon stable isotope.

For Cd, all 18 blanks were below detection limits (<0.01 ng/kg); thus, blanks were not corrected. Multiple standard reference materials were run for the metal analysis, including Biota Reference DOLT-4 from dogfish liver, DORM-3 and TORT-2 from dogfish muscle, all supplied by the National Research Council of Canada. Standard reference material percent recovery ranged from 96.3-122 %. Two duplicates were run, and the relative percent difference ranged from 0.5—16.6%. Mean spike recovery n=4 was 91.7% (range 88.9-94.9%). For THg, the detection limit was 0.001 mg/kg for DMA analysis.

Blank results for PFAS (N=8) were generally less than detection limits (<0.01 ng/g ww) except for PFHxA (averaging 0.02 ng/g). The reference material, NIST SRM-1947 homogenized lake trout, certified for PFOS, was used for the PFAS analyses. Results for PFOS (N=2) averaged 5.5 ng/g ww which was within 8% of the certified value of 5.9 ng/g (Table S4.3).

For PCBs, OCs and PBDEs, each batch (n=12) contained a method blank. Samples were spiked with $^{13}C_{12}$-PCB, $^{13}C_{x}$-OC or $^{13}C_{12}$-BDE recovery standards from Cambridge Isotope Laboratories (MA, USA) for PCBs/OCs and Wellington Laboratories (Guelph, ON, Canada) for PBDEs. Spike recoveries for PBDEs ranged from 44.8% for $^{13}C_{12}$-BDE209 to 71.2% for $^{13}C_{12}$-BDE28. For PCBs, spike recoveries ranged from 58.0% for $^{13}C_{12}$-PCB52 to 65.7% for $^{13}C_{12}$-PCB194 and for OCs, recoveries ranged from 70.4% for $^{13}C_{10}$-Mirex to 109.4% for 13C_{10}-*p,p'*-DDT. Mean blank concentrations were 1.03 ng/g (range 0.03-6.45) for OCs, 0.29 ng/g (range 0.00-0.81) for PBDEs, and 0.06 ng/g (range 0.009-0.32) for PCBs (more details found in table S4.4) . Lipid percent for PCBs, OCs and PBDEs ranged from 72% to 92%, and all concentrations were reported on a lipid weight basis. Results were also blank- and recovery-corrected.

Table S4.1 Biological information (sex, age, blubber thickness at the sternum (body condition)) and harvest date of ringed seal individuals from Arviat, Nain, Sachs Harbour and Resolute Bay.

Sample ID	***Location***	***Sex***	***Age (yr)***	***Body Condition (cm)***	***Capture Date***
AV2018-02	Arviat	F	1	6	October 28, 2018
AV2018-06	Arviat	F	9	6	October 20, 2018
AV2018-12	Arviat	M	0	4	October 17, 2018
AV2018-17	Arviat	F	2	6	October 28, 2018
AV2018-34	Arviat	F	12	5	October 17, 2018
AV2018-37	Arviat	F	8	6.35	October 15, 2018
AV2018-38	Arviat	F	17	5	October 17, 2018
AV2018-40	Arviat	F	23	6	October 29, 2018
AV2018-50	Arviat	F	21	8	no info
NAIN2018-01	Nain	F	0	4.45	August 27 2018
NAIN2018-02	Nain	M	0	5.08	September 6 2018
NAIN2018-03	Nain	M	4	5.08	September 6 2018
NAIN2018-07	Nain	F	2	6.35	October 15 2018
NAIN2018-09	Nain	F	1	6.35	October 31 2018
NAIN2018-10	Nain	F	3	5.08	November 2 2018
NAIN2018-11	Nain	F	1	5.08	November 2 2018
NAIN2018-13	Nain	F	0	7.62	November 13 2018
NAIN2018-15	Nain	M	0	5.08	November 21 2018
NAIN2018-17	Nain	F	3	5.08	no info
RB2018-02	Resolute Bay	F	6	4	June 13, 2018
RB2018-04	Resolute Bay	M	5	6	no info
RB2018-05	Resolute Bay	M	0	5	June 14, 2018
RB2018-06	Resolute Bay	F	34	4	June 26, 2018
RB2018-08	Resolute Bay	M	3	6	June 22, 2018
RB2018-11	Resolute Bay	F	7	4	June 22, 2018
RB2018-16	Resolute Bay	F	6	2.5	June 26, 2018
RB2018-18	Resolute Bay	F	20	3	June 2, 2018
RB2018-20	Resolute Bay	F	23	6	June 4, 2018
SH2018-03	Sachs Harbour	M	0	3	July 24 2018
SH2018-05	Sachs Harbour	M	0	2	July 29 2018
SH2018-09	Sachs Harbour	F	8	2	July 29 2018
SH2018-13	Sachs Harbour	F	8	2.5	July 30 2018
SH2018-14	Sachs Harbour	F	1	3.81	July 30 2018
SH2018-15	Sachs Harbour	M	7	2.54	July 30 2018
SH2018-16	Sachs Harbour	M	0	1.5	August 2 2018
SH2018-17	Sachs Harbour	F	2	2.5	August 5 2018
SH2018-18	Sachs Harbour	F	9	3	August 5 2018

Table S4.2 Complete list of polychlorinated biphenyls (PCBs) congeners, organochlorine pesticides (OCPs) compounds, polybrominated diphenyl ether (PBDE) flame retardants congeners, per-/polyfluoroalkyl substances (PFAS) compounds and trace metals analyzed in ringed seal blubber harvested from sub-Arctic and Arctic sites in 2018.

PCBs	***OCPs***	***PBDEs***	***PFAS***	***Metals***
PCB17	Aldrin	BDE17	PFBS	Aluminum
PCB18	alpha-BHC	BDE28	PFHxS	Antimony
PCB28	alpha-Chlordane	BDE47	PFHpS	Arsenic
PCB31	beta-BHC	BDE49	PFOS	Barium
PCB33	Chlorothalonil	BDE66	PFDS	Beryllium
Trichlorobiphenyls (homologue group)	cis-Nonachlor	BDE71	PFHpA	Bismuth
PCB44	delta-BHC	BDE77	PFOA	Boron
PCB49	Deltamethrin	BDE85	PFNA	Cadmium
PCB52	Dieldrin	BDE99	PFDA	Calcium
PCB70	Endosulfan I	BDE100	PFUnA	Cesium
PCB74	Endosulfan II	BDE119	PFDoA	Chromium
Tetrachlorobiphenyls (homologue group)	Endosulfan sulfate	BDE126	PFTriA	Cobalt
PCB82	Endrin	BDE138	PFTetra	Copper
PCB87	Endrin aldehyde	BDE153	PFOSA	Gallium
PCB95	Endrin ketone	BDE154	L_PFOS	Iron
PCB99	gamma-BHC (Lindane)	BDE156	L_PFNA	Lanthanum
PCB101	gamma-Chlordane	BDE183	L_PFDA	Lead
PCB105	Heptachlor	BDE184	L_PFUnA	Lithium
PCB110	Heptachlor Epoxide (Isomer A)	BDE191	L-PFCAS	Magnesium
PCB118	Heptachlor Epoxide (Isomer B)	BDE196	PFECHSb	Manganese
Pentachlorobiphenyls (homologue group)	Hexachlorobenzene	BDE197	CL PFOS	Mercury
PCB128	Hexachlorobutadiene	BDE206	PFONS	Molybdenum
PCB132	Hexachlorocyclopentadiene	BDE207	PFOUdS	Nickel
PCB138	Methoxychlor	BDE209	F-53B	Palladium
PCB149	Mirex	HBB153	NaDONA	Phosphorus
PCB151	Octachlorostyrene		PFPeA	Platinum
PCB153	2,4'-DDD		PFHxA	Potassium
PCB156	o.p'-DDE		PFOS-L	Rubidium
PCB158	2,4'-DDT			Selenium
PCB169	Oxychlordane			Silver
Hexachlorobiphenyls (homologue group)	Pentachloroanisole			Strontium
PCB170	Pentachlorobenzene			Tellurium
PCB171	Permethrin			Thallium
PCB177	p-p'-DDD			Tin
PCB180	p-p'-DDE			Uranium
PCB183	p-p'-DDT			Vanadium
PCB187	trans-Nonachlor			Zinc
PCB191				Zirconium
Heptachlorobiphenyls (homologue group)				
PCB194				
PCB195				

PCB199				
PCB205				
Octachlorobiphenyls (homologue group)				
PCB206				
PCB208				
Nonachlorobiphenyls (homologue group)				
PCB209				
Decachlorobiphenyl (Homoloque group)				

Table S4.3 Quality control information for per-/polyfluoroalkyl substances (PFAS) in ringed seal liver harvested in 2018 from sub-Arctic and Arctic sites.

PFAS	**Mass Transitions** m/z	**Blanks**	**Nist 1947 #1** ng/g	**Nist 1947 #2** ng/g	**Average** ng/g	**Concensus Values[1]** ng/g
PFHxA	313/269	0.211	<0.001	0.444	0.22	0.10
PFHpA	363/319	0.009	0.048	0.079	0.06	0.11
PFOA	413/369	0.013	0.072	0.077	0.07	0.19
PFNA	463/419	0.006	0.223	0.214	0.22	0.26
PFDA	513/469	0.003	0.233	0.243	0.24	0.27
PFUnA	553/519	0.002	0.256	0.330	0.29	0.32
PFDoA	613/569	0.002	0.107	0.108	0.11	0.15
PFTRiA	663/619	0.002	0.566	0.255	0.41	0.25
PFTA	713/669	0.011	<0.001	0.098	0.10	0.13
PFBS	299/99	0.001	<0.001	<0.001	<0.001	
PFHxS	399/99	<0.001	0.030	0.016	0.02	0.04
PFHpS	449/99	<0.001	0.037	0.018	0.03	
PFOS	499/99	0.022	5.228	5.645	5.44	5.9[2]
PFOS-L	499/99	0.006	5.171	5.391	5.28	
PFDS	599/99	0.002	0.039	0.065	0.05	0.09
PFOSA	498/78	<0.001	0.253	0.258	0.26	0.18

[1] Concensus (uncertified) values in Reiner et al. 2012

[2] Certified value for PFOS in NIST 1947

Table S4.4 Quality control spike recovery information for mass-labelled polychlorinated biphenyls (PCB) congeners, organochlorine pesticides (OCPs), and polybrominated diphenyl ether (PBDE) flame retardants for ringed seal blubber samples across four Canadian sub-Arctic and Arctic sites sampled in 2018.

Spiked Analytes	Mean Spike Recovery (%) and range
$^{13}C_{12}$-PCB28	58 (37 to 96)
$^{13}C_{12}$-PCB52	58 (37 to 96)
$^{13}C_{12}$-PCB111	61 (38 to 100)
$^{13}C_{12}$-PCB153	64 (41 to 100)
$^{13}C_{12}$-PCB178	61 (39 to 98)
$^{13}C_{12}$-PCB194	66 (44 to 100)
$^{13}C_{12}$-PCB208	64 (41 to 100)
$^{13}C_{6}$-delta-BHC	107 (41 to 180)
$^{13}C_{8}$-Octachlorostyrene	110 (43 to 200)
$^{13}C_{12}$-p-p'-DDE	103 (41 to 180)
$^{13}C_{12}$-p-p'-DDT	109 (44 to 250)
$^{13}C_{10}$-Mirex	70 (25 to 120)
$^{13}C_{6}$-Pentachlorobenzene	79 (32 to 130)
$^{13}C_{12}$-BDE28	71 (39 to 120)
$^{13}C_{12}$-BDE47	67 (37 to 120)
$^{13}C_{12}$-BDE100	69 (41 to 120)
$^{13}C_{12}$-BDE153	69 (41 to 120)
$^{13}C_{12}$-BDE183	65 (41 to 120)
$^{13}C_{12}$-BDE209	45 (22 to 94)

Table S4.5 Raw data (mass percent) of the 33 fatty acids considered to be mainly of dietary origin (Iverson et al 2004) for all seal individuals from each location (Arviat = AV, Nain = NAIN, Resolute Bay = RS, Sachs Harbour = SH) harvested in 2018.

	AV02	**AV06**	**AV12**	**AV17**	**AV34**	**AV37**	**AV38**	**AV40**	**AV50**
16:2n6	0.064	0.072	0.127	0.064	0.077	0.062	0.06	0.07	0.057
16:2n4	0.576	0.427	0.213	0.461	0.509	0.492	0.57	0.496	0.488

16:3n6	0.3	0.453	0.761	0.326	0.393	0.406	0.34	0.347	0.292
16:3n4	0.175	0.21	0.361	0.203	0.182	0.208	0.176	0.2	0.22
16:3n1	0.024	0.022	0.034	0.02	0.023	0.021	0.022	0.025	0.022
16:4n3	0.149	0.114	0.137	0.111	0.141	0.128	0.143	0.155	0.127
16:4n1	0.184	0.187	0.317	0.221	0.143	0.168	0.123	0.211	0.269
18:2n6	2.101	2.208	2.081	1.89	2.479	2.439	1.99	2.308	2.409
18:2n4	0.112	0.103	0.145	0.098	0.094	0.096	0.085	0.119	0.112
18:3n6	0.137	0.105	0.22	0.116	0.1	0.113	0.101	0.13	0.125
18:3n4	0.08	0.098	0.188	0.079	0.106	0.092	0.099	0.104	0.06
18:3n3	0.902	0.621	0.964	0.608	0.9	0.716	0.589	0.765	0.976
18:3n1	0.079	0.047	0.042	0.048	0.039	0.054	0.06	0.046	0.046
18:4n3	2.085	0.854	1.971	1.381	1.525	1.098	1.105	1.394	2.256
18:4n1	0.075	0.084	0.183	0.079	0.081	0.082	0.068	0.094	0.107
20:1n11	0.387	1.178	0.831	0.847	0.796	1.01	1.029	0.605	0.418
20:1n9	2.999	6.037	4.536	5.3	4.251	5.164	4.484	3.581	3.795
20:1n7	0.485	0.719	0.283	0.574	0.488	0.576	0.599	0.467	0.435
20:2n6	0.327	0.255	0.263	0.28	0.264	0.278	0.309	0.294	0.311
20:3n6	0.068	0.058	0.091	0.056	0.075	0.07	0.075	0.083	0.065
20:4n6	0.358	0.285	0.458	0.331	0.294	0.296	0.365	0.329	0.311
20:3n3	0.146	0.146	0.13	0.11	0.146	0.1	0.028	0.162	0.172
20:4n3	0.646	0.358	0.625	0.429	0.607	0.443	0.456	0.523	0.667
20:5n3	8.64	6.827	13.878	8.492	7.212	7.07	6.593	7.908	9.54
22:1n11	0.67	1.353	0.193	1.917	0.521	1.045	0.695	0.806	0.975
22:1n9	0.241	0.446	0.093	0.485	0.206	0.347	0.286	0.259	0.266
22:1n7	0.072	0.088	0.008	0.107	0.048	0.071	0.059	0.077	0.066
22:2n6	0.03	0.013	0.008	0.021	0.007	0.014	0.02	0.00	0.021
21:5n3	0.394	0.362	0.594	0.312	0.439	0.384	0.327	0.011	0.385
22:4n6	0.113	0.062	0.044	0.058	0.077	0.063	0.089	0.083	0.056
22:5n6	0.189	0.14	0.117	0.122	0.171	0.175	0.196	0.162	0.122
22:4n3	0.109	0.067	0.065	0.046	0.113	0.08	0.081	0.086	0.073
22:6n3	14.618	11.556	15.762	11.043	15.396	13.676	13.718	13.175	12.649

	NAIN 01	**NAIN 02**	**NAIN 03**	**NAIN 07**	**NAIN 09**	**NAIN 10**	**NAIN 11**	**NAIN 13**	**NAIN 15**	**NAIN 17**
16:2n6	0.096	0.1	0.091	0.073	0.078	0.079	0.085	0.095	0.082	0.092
16:2n4	0.105	0.087	0.156	0.091	0.168	0.147	0.12	0.063	0.072	0.18
16:3n6	0.6	0.605	0.724	0.649	0.705	0.622	0.527	0.701	0.598	0.771
16:3n4	0.283	0.323	0.537	0.32	0.526	0.45	0.392	0.505	0.422	0.476

16:3n1	0.029	0.033	0.028	0.031	0.026	0.031	0.027	0.032	0.027	0.035
16:4n3	0.056	0.074	0.07	0.073	0.069	0.08	0.101	0.089	0.084	0.108
16:4n1	0.473	0.588	1.208	0.641	1.063	0.908	0.616	1.023	0.664	0.806
18:2n6	1.665	1.762	1.135	1.607	1.173	1.106	1.126	1.721	1.194	1.335
18:2n4	0.156	0.163	0.133	0.144	0.132	0.141	0.151	0.168	0.152	0.161
18:3n6	0.239	0.265	0.16	0.213	0.154	0.155	0.159	0.245	0.163	0.163
18:3n4	0.14	0.186	0.153	0.123	0.15	0.136	0.107	0.171	0.123	0.165
18:3n3	0.218	0.33	0.473	0.4	0.468	0.558	0.596	0.437	0.533	0.64
18:3n1	0.044	0.055	0.075	0.057	0.073	0.087	0.076	0.048	0.054	0.105
18:4n3	0.966	1.347	1.706	1.299	1.515	1.787	2.237	1.569	2.203	1.862
18:4n1	0.265	0.373	0.399	0.204	0.373	0.365	0.256	0.314	0.287	0.351
20:1n11	1.346	0.959	1.527	0.844	1.65	1.655	1.689	1.063	2.455	1.92
20:1n9	4.708	3.661	8.847	3.707	7.823	9.692	7.658	4.534	9.737	10.678
20:1n7	0.381	0.316	0.509	0.297	0.466	0.658	0.493	0.42	0.552	0.585
20:2n6	0.234	0.194	0.183	0.223	0.185	0.195	0.22	0.211	0.239	0.233
20:3n6	0.107	0.115	0.075	0.089	0.063	0.069	0.057	0.078	0.074	0.084
20:4n6	0.43	0.478	0.315	0.431	0.331	0.385	0.268	0.417	0.208	0.443
20:3n3	0.067	0.067	0.091	0.072	0.09	0.08	0.09	0.104	0.102	0.109
20:4n3	0.31	0.445	0.442	0.389	0.42	0.492	0.547	0.419	0.537	0.579
20:5n3	9.883	10.347	9.785	10.301	9.801	9.309	11.189	10.832	10.235	11.326
22:1n11	0.973	1.493	4.916	0.958	4.802	6.12	4.009	1.901	5.041	5.86
22:1n9	0.231	0.01	0.687	0.196	0.626	0.919	0.583	0.303	0.649	0.836
22:1n7	0.01	0.038	0.108	0.007	0.092	0.154	0.097	0.045	0.11	0.128
22:2n6	0.012	0.007	0.019	0.003	0.017	0.018	0.02	0.008	0.025	0.017
21:5n3	0.475	0.499	0.555	0.464	0.513	0.504	0.456	0.495	0.463	0.602
22:4n6	0.059	0.063	0.06	0.085	0.044	0.071	0.042	0.051	0.038	0.075
22:5n6	0.069	0.06	0.064	0.069	0.065	0.071	0.07	0.062	0.058	0.082
22:4n3	0.064	0.075	0.099	0.074	0.076	0.101	0.091	0.069	0.099	0.105
22:6n3	8.24	7.819	9.403	10.065	9.913	9.794	10.792	7.809	8.193	11.977

	RB02	**RB04**	**RB05**	**RB06**	**RB08**	**RB11**	**RB16**	**RB18**	**RB20**
16:2n6	0.081	0.07	0.069	0.077	0.071	0.081	0.073	0.077	0.087
16:2n4	0.074	0.119	0.131	0.135	0.087	0.112	0.11	0.125	0.128
16:3n6	0.797	0.729	0.745	0.96	0.821	0.885	0.781	0.825	1.026
16:3n4	0.383	0.321	0.232	0.318	0.387	0.3	0.337	0.285	0.279
16:3n1	0.026	0.025	0.024	0.027	0.026	0.029	0.027	0.026	0.026

16:4n3	0.05	0.05	0.055	0.047	0.049	0.049	0.045	0.059	0.042
16:4n1	0.547	0.416	0.232	0.337	0.572	0.359	0.513	0.307	0.264
18:2n6	0.83	0.673	0.943	0.808	0.819	0.906	0.775	1.009	0.82
18:2n4	0.161	0.15	0.162	0.109	0.155	0.149	0.173	0.13	0.107
18:3n6	0.193	0.169	0.157	0.171	0.182	0.182	0.183	0.184	0.149
18:3n4	0.123	0.099	0.114	0.14	0.134	0.166	0.143	0.141	0.165
18:3n3	0.273	0.244	0.259	0.29	0.337	0.26	0.262	0.36	0.287
18:3n1	0.043	0.055	0.047	0.047	0.048	0.042	0.048	0.049	0.045
18:4n3	0.901	0.72	0.456	0.749	0.92	0.692	0.71	0.704	0.619
18:4n1	0.181	0.211	0.102	0.161	0.217	0.23	0.246	0.165	0.167
20:1n11	1.813	1.9	2.179	2.36	1.962	1.597	2.637	2.571	2.238
20:1n9	9.604	10.437	10.946	8.711	8.719	6.423	9.227	8.902	5.915
20:1n7	0.976	1.05	1.1	0.857	0.842	0.577	1.015	0.901	0.521
20:2n6	0.166	0.165	0.211	0.2	0.159	0.179	0.173	0.179	0.198
20:3n6	0.095	0.098	0.11	0.121	0.092	0.118	0.115	0.121	0.132
20:4n6	0.241	0.331	0.293	0.283	0.255	0.336	0.27	0.295	0.278
20:3n3	0.038	0.054	0.047	0.028	0.041	0.029	0.029	0.034	0.025
20:4n3	0.296	0.3	0.245	0.322	0.304	0.307	0.304	0.31	0.316
20:5n3	10.615	10.628	7.498	9.881	11.266	10.102	9.313	7.788	10.766
22:1n11	1.959	4.627	1.431	0.648	1.5	0.919	2.305	1.368	0.283
22:1n9	0.771	1.215	0.765	0.414	0.571	0.346	0.792	0.588	0.168
22:1n7	0.119	0.213	0.099	0.03	0.086	0.048	0.11	0.077	0.008
22:2n6	0.023	0.011	0.03	0.022	0.005	0.018	0.021	0.018	0.02
21:5n3	0.408	0.325	0.378	0.458	0.467	0.421	0.415	0.416	0.518
22:4n6	0.047	0.054	0.061	0.058	0.051	0.06	0.079	0.056	0.038
22:5n6	0.049	0.068	0.07	0.085	0.072	0.062	0.079	0.07	0.064
22:4n3	0.053	0.053	0.056	0.058	0.061	0.046	0.061	0.062	0.047
22:6n3	6.671	7.688	7.607	9.643	8.158	7.981	7.5	8.336	9.731

	SH03	**SH05**	**SH09**	**SH13**	**SH14**	**SH215**	**SH16**	**SH17**	**SH218**
16:2n6	0.107	0.103	0.08	0.071	0.071	0.084	0.078	0.082	0.073
16:2n4	0.213	0.237	0.194	0.213	0.193	0.18	0.305	0.194	0.218
16:3n6	0.423	0.527	0.709	0.722	0.554	0.716	0.429	0.644	0.646
16:3n4	0.23	0.325	0.279	0.282	0.233	0.268	0.226	0.31	0.273
16:3n1	0.026	0.034	0.028	0.034	0.032	0.034	0.035	0.037	0.024
16:4n3	0.08	0.116	0.083	0.077	0.106	0.084	0.134	0.082	0.075

16:4n1	0.089	0.22	0.273	0.308	0.257	0.269	0.185	0.341	0.231
18:2n6	0.626	1.303	1.029	0.946	1.082	0.905	1.661	0.886	0.793
18:2n4	0.101	0.15	0.131	0.111	0.12	0.11	0.102	0.137	0.103
18:3n6	0.13	0.214	0.153	0.185	0.148	0.144	0.226	0.144	0.129
18:3n4	0.149	0.225	0.166	0.143	0.128	0.14	0.182	0.166	0.132
18:3n3	0.167	0.507	0.464	0.523	0.624	0.569	0.54	0.434	0.365
18:3n1	0.054	0.053	0.044	0.053	0.05	0.051	0.057	0.057	0.059
18:4n3	0.479	0.999	0.819	0.903	1.182	1.068	1.073	0.979	0.735
18:4n1	0.101	0.182	0.201	0.159	0.15	0.184	0.137	0.214	0.117
20:1n11	1.526	0.767	1.171	1.453	1.356	0.84	0.448	1.168	2.238
20:1n9	6.044	4.299	4.716	6.657	6.721	4.054	3.551	4.977	9.5
20:1n7	0.598	0.428	0.406	0.637	0.532	0.308	0.257	0.578	0.963
20:2n6	0.242	0.2	0.157	0.188	0.216	0.159	0.339	0.158	0.221
20:3n6	0.103	0.121	0.1	0.121	0.092	0.086	0.163	0.102	0.099
20:4n6	0.37	0.73	0.35	0.391	0.386	0.372	0.84	0.484	0.28
20:3n3	0.047	0.072	0.055	0.081	0.086	0.061	0.101	0.071	0.059
20:4n3	0.247	0.492	0.402	0.428	0.46	0.492	0.495	0.453	0.322
20:5n3	5.319	7.767	10.326	9.513	9.26	12.574	8.124	9.382	7.073
22:1n11	0.391	0.145	0.296	1.12	0.764	0.253	0.062	0.729	2.3
22:1n9	0.238	0.121	0.137	0.403	0.257	0.107	0.079	0.219	0.725
22:1n7	0.033	0.016	0.027	0.059	0.034	0.014	0.008	0.04	0.126
22:2n6	0.023	0.002	0	0.026	0.007	0.003	0.024	0.004	0.014
21:5n3	0.217	0.355	0.378	0.37	0.349	0.457	0.34	0.403	0.351
22:4n6	0.051	0.108	0.059	0.086	0.075	0.051	0.13	0.106	0.068
22:5n6	0.082	0.102	0.088	0.105	0.11	0.092	0.145	0.133	0.12
22:4n3	0.06	0.066	0.057	0.078	0.082	0.057	0.066	0.074	0.073
22:6n3	9.262	10.422	10.794	10.553	12.183	12.783	13.23	10.792	11.276

Table S4.6 Mean contaminant[a] and range of concentrations in ringed seals from 2018 in the sub-Arctic sites (Arviat, Nain), and Arctic sites (Resolute Bay, Sachs Harbour).

Contaminant[b]	**Arviat**	**Nain**	**Resolute Bay**	**Sachs Harbour**
ΣPCB	232.22 (56.33-489.33)	226.45 (58.33-369.33)	300.45 (109.33-489.33)	331.55 (179.33-409.33)
$\Sigma 10PCB$	137.55 (32.13-260.82)	141.21 (33.26-238.59)	176.60 (58.60-289.21)	197.94 (107.74-244.61)

PCB153	55.17 (10.95-99.95)	58.56 (10.95-109.95)	58.17 (14.95-93.95)	66.61 (38.95-85.95)
ΣCHL	116.81 (37.50-327.14)	62.18 (29.13-100.09)	224.96 (116.25-363.35)	210.10 (89.48-401.05)
ΣHCH	19.79 (9.27-55.41)	27.76 (13.63-47.91)	82.74 (44.57-120.51)	110.63 (24.62-181.71)
HCB	11.05 (5.74-21.53)	9.39 (6.37-14.47)	20.06 (13.4-26.32)	20.48 (12.36-33.32)
ΣDDT	183.13 (26.76-551.96)	118.18 (33.00-21.96)	223.01 (89.66-398.96)	162.92 (78.56-234.36)
Mirex	6.96 (1.06-29.96)	0.95 (0.00 -1.76)	3.23 (1.46-5.26)	3.17 (1.36-6.45)
Dieldrin	19.67 (12.00-47.00)	27.8 (12.00-72.00)	61.56 (38.00-110.00)	40.70 (28.00-60.00)
ΣPBDE	7.13 (1.01-19.38)	8.62 (2.81-16.38)	2.78 (0.65-6.65)	4.45 (2.57-6.27)
PBDE47	5.16 (0.85-14.65)	5.71 (1.55-11.65)	2.78 (0.65-6.65)	3.49 (1.25-5.95)
ΣPFSA	32.50 (14.12-49.33)	9.42 (3.91-14.41)	13.53 (4.84-26.40)	18.71 (5.65-33.91)
ΣPFCA	35.55 (14.87-59.04)	6.05 (2.60-9.82)	18.36 (5.61-35.81)	35.81 (18.99-55.47)
PFOS	31.81 (13.79-48.29)	9.10 (3.53-14.21)	13.33 (4.75-26.01)	18.31 (5.42-33.13)
PFOA	0.87 (0.36-1.34)	0.09 (0.00-0.40)	0.16 (0.05-0.45)	0.27 (0.12-0.50)
Total Mercury	0.23 (0.14-0.68)	0.01 (0.04-0.16)	0.44 (0.16-0.58)	0.41 (0.11-0.82)
Cadmium	9.2 (3.11-15.90)	1.63 (1.15-2.08)	8.6 (3.24-14.4)	3.24 (0.161-7.82)

[a] Σpolychlorinated biphenyls (ΣPCBs), ten major congeners of PCBs (Σ10PCB), Σchlordanes (ΣCHL), Σhexachlorocyclohexanes (ΣHCH), hexachlorobenzene (HCB), Σdichlorodiphenyltrichloroethanes (ΣDDTs), mirex, dieldrin, polybrominated diphenyl ether (ΣPBDE) flame retardants, PBDE47, perfluorosulfonic acid (ΣPFSA), Σperfluoroalkyl carboxylates (ΣPFCA), perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA), mercury, cadmium

[b] ng/g lipid weight for PCBs, CHL, HCB, DDT, Mirex, Dieldran, PBDEs
mg/kg wet weight for Mercury (muscle), and Cadmium (liver).
ng/g wet weight for PFAS (liver)
Breakdown of each contaminant group is found in table S6.

Table S4.7 List of compounds analyzed in ringed seals in all four study locations (Arviat, Nain, Sachs Harbour, Resolute Bay) harvested in 2018.

Contaminant Group	**Compounds included**

ΣPCB	PCB 28,47,49, 52, 70, 74, 82, 87, 95, 99, 101, 105, 110, 118, 128, 132, 138, 149, 151, 153, 156, 158, 169, 170, 171, 177, 180, 183, 187, 191, 194, 195, 199, 205, 206, 208, 209
Σ10PCB	Major congeners: PCB28, 31, 52, 101, 105, 118,138, 153, 156, 180
ΣCHL	Alpha-Chlordane, *cis*-Nonachlor, gamma-Chlordane, Heptachlor Epoxide (Isomer B), Oxychlordane, *trans*-Nonachlor
ΣDDT	p-p`-DDD, p-p`-DDE, p-p`-DDT, 2,4`-DDD, o.p`-DDE, 2,4`-DDT
ΣHCH	Alpha-BHC, beta-BHC, gamma-BHC
ΣPBDE	BDE17, 28, 47,153, 209
ΣPFSA	PFBS, PFHxS, PFHpS, PFOS, PFDS
ΣPFCAs	PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFTriA, PFTetra

Σpolychlorinated biphenyls (ΣPCBs), ten major congeners of PCBs (Σ10PCB), Σchlordanes (ΣCHL) Σdichlorodiphenyltrichloroethanes (ΣDDTs), Σhexachlorocyclohexanes (ΣHCH), Σpolybrominated diphenyl ether (ΣPBDE) flame retardants, perfluorosulfonic acid (ΣPFSA), Σperfluoroalkyl carboxylates (ΣPFCA).

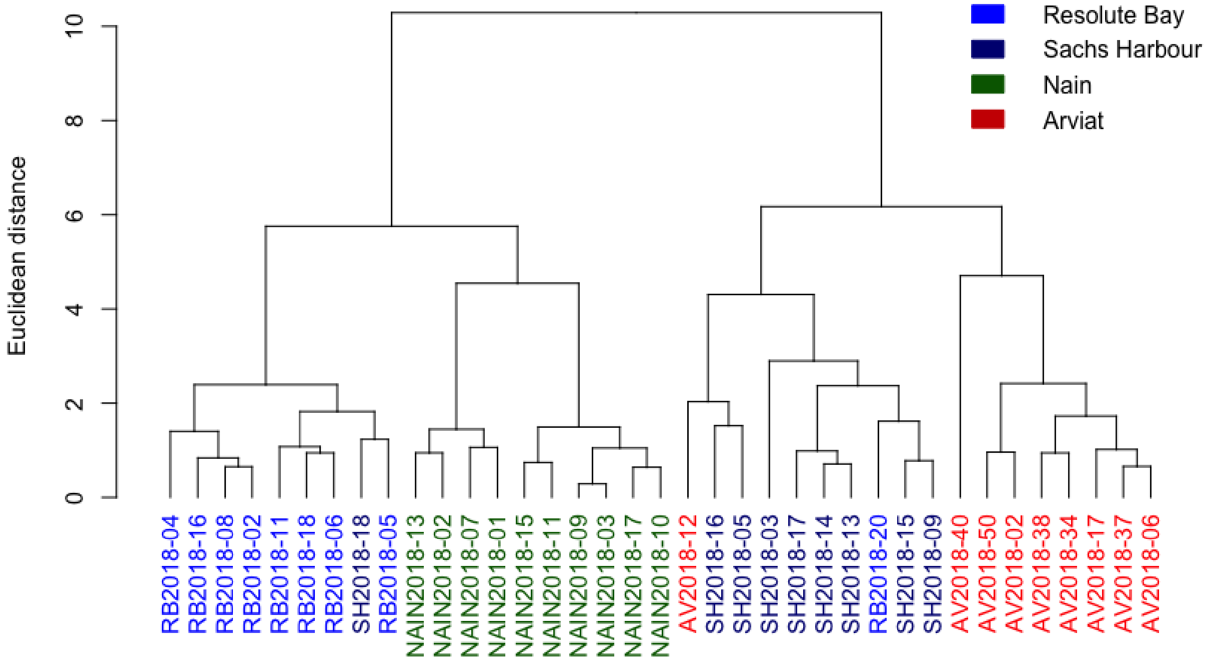

Figure S4.1 Hierarchal clustering analysis based on blubber fatty acid signatures as dietary tracers for ringed seals (Location2018-sample#) sampled from Canadian sub-Arctic (Arviat (AV), Nunavut and Nain (NAIN), Labrador) and Arctic (Resolute Bay (RB), and Sachs Harbour (SH), Nunavut) sites in 2018.

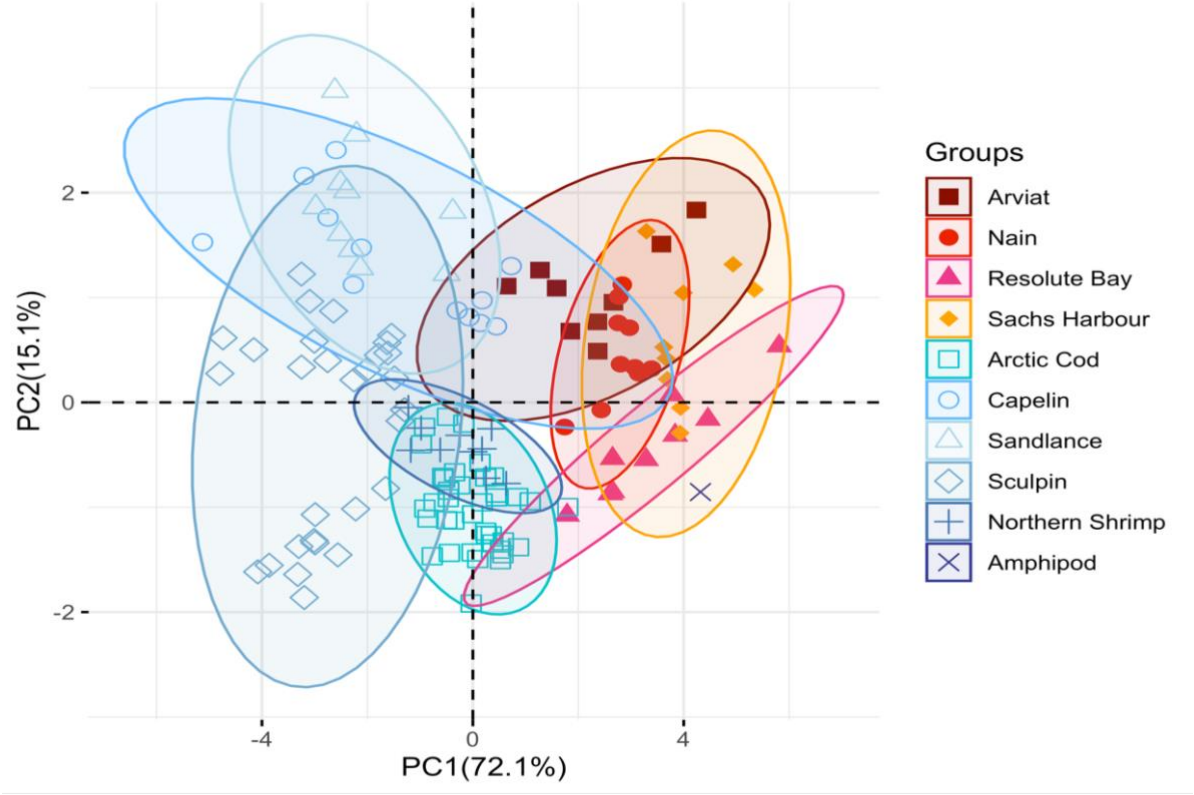

Figure S4.2 Principal component analysis of ringed seal sampled in 2018 from Arviat, Nain, Resolute Bay and Sachs Harbour and their potential prey (Arctic cod, capelin, sand lance, sculpin, northern shrimp, and amphipod (Themisto libellula)) sampled in the low, mid and high regions of the eastern Canadian Arctic between 2012 to 2014 from Pedro et al 2020.

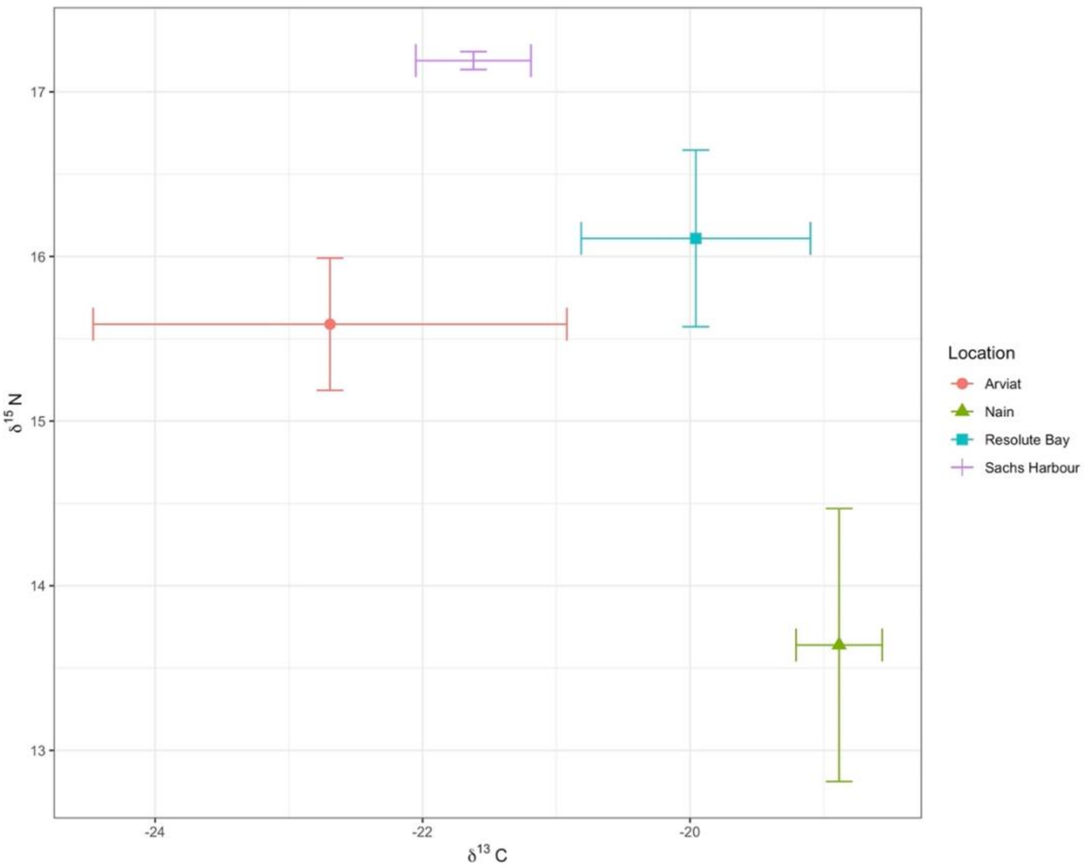

Figure S4.3 Isotopic biplot of bulk carbon and nitrogen stable isotopes from ringed seals from each location (red = Arviat, green = Nain, blue = Resolute Bay, purple = Sachs Harbour) sampled in 2018. Not corrected for isotopic baseline differences at each location.

Chapter 5: General Discussion

The overarching aim of this thesis was to enhance the understanding of new contaminants in Arctic marine wildlife and gain insight into climate change-contaminant interactions. Having a sufficient understanding of POPs within the Canadian Arctic is essential to provide adequate scientific advice to governments and international regulatory agencies, such that evidence-based actions are taken that ultimately protect the Arctic ecosystem. By reporting measurable levels of SCCPs within Arctic marine fishes and ringed seals, contrasting the concentrations of SCCPs with those of legacy POPs in the same biota, and calculating the biomagnification potential of SCCPs within the food web, Chapter 3 contributed to the growing evidence of the POPs-like behaviour of chlorinated paraffin including long-range transport and bioaccumulation within Arctic biota. The relatively high levels of this new contaminant group in marine fishes highlights the need for continuous monitoring of SCCPs in national and international long-term monitoring programs. Chapter 4 showed dietary pattern differences between ringed seals from the sub-Arctic and Arctic, which were likely associated with the higher consumption of southern invading species in the sub-Arctic. After adjusting for the influence of diet, this study found a potential for a diet-driven increase in some of the PFASs, which are newer contaminants, but a decrease in THg, DDT, and PBDE concentrations in ringed seals. This chapter calls attention to the potential implications of climate-induced feeding changes in ringed seals on their contaminant loads and emphasizes the need for follow-up temporal trends studies on contaminant-feeding changes. Combined, these two studies in this thesis contribute to the greater understanding of POPs distribution and the influence of trophic and food web ecology on POPs within the Arctic and provide insight into future directions for SCCP and climate-contaminant interactions studies.

While Chapter 3 improved the understanding of SCCPs in the Canadian Arctic, further research would enhance these findings. This study found detectable levels of SCCPs in the marine fishes and amphipods sampled in 2014 and ringed seals sampled in 2017 from Hudson Bay. SCCP levels in the amphipods were very low compared to congenerics from Svalbard from 2011 and 2012 (Li et al. 2017). This could partly be related to the challenges in the comparability of SCCP results between studies due to differences in quantifying methods (van Mourik et al. 2016). However, the small sample size of one pool in Chapter 3 limits the strength of conclusions that can be drawn for comparisons of amphipod SCCP levels among regions. Moving forward, having multiple pools of amphipods and amphipod species from both pelagic and benthic sources would be useful to understand the distribution of SCCP levels at the base of this Canadian sub-Arctic food web. In the marine fishes, SCCP levels were comparable to the levels found in a top marine predator, the Greenland shark (*Somniosus microcephalus*) caught around Iceland from 2001 to 2003 (Strid et al. 2013). The high concentrations of SCCPs in lower trophic level species, such as the marine fishes, suggest high concentrations of SCCPs in the Canadian Arctic environment. Compared to concentrations of legacy POPs in the same samples, SCCP levels were two to three orders of magnitude higher than PCBs and OC concentrations for capelin, sand lance, cisco and Greenland cod. This SCCP and PCB pattern was also documented in the Greenland shark (Strid et al. 2013). These results demonstrate the burden of SCCPs on these Arctic marine fishes and raise a concern about the potential for toxicological impacts of SCCPs on these species. Due to the wide global application and release of SCCPs, there has been an increase in SCCP toxicity studies in the past decade. These studies have documented lethal, carcinogenic, endocrine, metabolism and immune effects of SCCPs in wildlife and humans (Wang et al. 2019). None of these SCCP toxicity studies have been performed in Arctic marine fish. However, toxicity studies on zebrafish (*Danio rerio*) larvae have documented a 13-day 50% lethal concentration dose of 34.4 μg/L (Ren et al. 2018). Without marine water SCCP concentrations from Arviat or a lethal concentration dose in marine fish tissue, it is difficult to infer from current literature if the high concentrations of SCCPs found in these marine fishes pose health risks to the population. Caution should also be used in extrapolating toxicity threshold concentrations to other species due to differences in sensitivity. Given the high levels of SCCPs found in these marine fish compared to legacy POPs, toxicity studies in these Arctic fishes are needed to evaluate the risk of the SCCPs concentrations measured in this study.

Despite such high levels of SCCPs found in the marine fishes, SCCPs levels in the ringed seals were four to eighteen times lower than those in the fishes. The trophic-adjusted biomagnification factor between the adult ringed seals and their prey (capelin and sand lance) was 0.32 ± 0.17 for ΣSCCPs and ranged from 0.0-2.23 for congeners $C_{10}Cl_6$ to $C_{12}Cl_8$, suggesting limited biomagnification. The sampling time differences (ringed seals were sampled in 2017, while the fishes were sampled in 2014) could be a factor influencing the reported levels. Nonetheless, these results could also or alternatively be an indication of the ringed seal's ability to biotransform or eliminate these contaminants. High levels of chlorinated paraffins have also been reported in lower trophic organisms, such as zebra mussels, but not in higher trophic level species, such as beluga whales (Fisk et al. 2009). There is also some evidence of SCCP biotransformation in other homeotherms, including birds and mice, but further study is needed in this field (Fisk et al. 2009). Therefore, to better understand SCCP accumulation and the potential for SCCP biotransformation in marine mammals, a more complete Arctic food web including lower trophic level species and more top marine mammal predators is needed, as well as in vitro biotransformation studies, such as has been done for other POPs (McKinney et al. 2006, McKinney et al. 2011, Letcher et al. 2014). The in vitro biotransformation of SCCPs has been documented in the human liver, chick embryos, hens, and juvenile rainbow trout but not in marine mammals (Darnerud and Bergman 2022). In addition to further investigating the biotransformation of SCCPs in marine mammals and the toxicity of this contaminant group in marine fishes, understanding how climate change can impact contaminants within the Arctic is a research priority.

In Chapter 4, climate change-induced feeding differences in ringed seals were investigated. As contaminant exposure in ringed seals occurs largely through diet, this study contributed to insight into climate-contaminant interactions. The results of Chapter 4 indicate differences in ringed seal feeding habits between the two sub-Arctic locations (Arviat and Nain) and the Arctic location (Resolute Bay). By comparing fatty acid (FA) signatures of the ringed seals to prey data from Pedro et al. (2020), these results suggest higher consumption of southern invading species (sand lance and capelin) in the sub-Arctic. These findings contribute to the evidence of "borealization" in the sub-Arctic and are supported by previous studies documenting feeding differences in ringed seals between the Arctic and sub-Arctic (Yurkowski et al. 2016). To gain a better understanding of ringed seal diet differences across the Arctic, a quantitative fatty acid signature analysis (QFASA) approach with a substantial prey database would be useful. This approach has the potential to provide species-level resolution of the ringed seals' diets, that is, the proportions of each prey species being consumed. The QFASA approach has been used in various recent polar bear studies evaluating the proportions of ringed seals in the polar bears' diets (Bourque et al. 2020, Florko et al. 2020). However, ringed seal diets have not been estimated using the QFASA method. Instead, other diet methods have been used to estimate, such as stomach content analysis, stable isotopes, and qualitative comparisons of fatty acid composition (Florko et al. 2021, Insley et al. 2021, Ross et al. 2022). The lack of QFASA studies in ringed seals is likely due to the substantial requirements needed to perform the analysis, such as sampling all major potential prey items and having a species-specific correction coefficient (CC) to account for the predator's metabolism (Bowen and Iverson 2013). Although a ringed seal CC is not currently calculated, it may be possible to use a CC from another seal species such as the grey seal (*Halichoerus grypus*) or harp seal (*Phoca groenlandica*) (Iverson et al. 2004). This has previously been done before where a CC from mink (Mustela vision) was successfully used in QFASA to estimate the polar bear diet (McKinney et al. 2013). Understanding how the ringed seal diet is changing due to climate change is important for understanding contaminant dynamics within the ringed seal population, as well as the influences on polar bears and on northern communities that rely on traditional country foods that include seal meat and other tissues.

The second objective of chapter 4 was to investigate the influence of dietary patterns on ringed seal contaminant levels. Out of all the dietary variables, the two main variables that significantly explained contaminant variation for most contaminant groups were $\delta\ ^{15}N$ and $\delta^{13}C$. The FA variables only appeared in two out of the seventeen models. For each contaminant group, Chapter 4 discusses various ecological and/or feeding reasons for the outcomes of each model. While bulk stable isotopes are a powerful tool for studying food webs, one discussion point that appears frequently is the spatial differences in isotopic baselines across the Arctic. The input of different water bodies and spatial differences at the base of the food chain can influence the baseline values of $\delta\ ^{15}N$ and $\delta^{13}C$ (De La Vega et al. 2019, De La Vega et al. 2021). These differences are potentially contributing to some of the unusual stable isotope-contaminant relationships observed in Chapter 4. For example, a negative relationship between $\delta^{15}N$ values and ΣHCH, HCB and dieldrin concentrations suggests that ringed seals feeding at higher trophic levels have lower contaminant concentrations, which is unexpected given consistent reporting of the biomagnification potential of these POPs across many previous studies (Fisk et al. 2001). These Chapter 4 results, in conjunction with recent other studies (De La Vega et al. 2019, De La Vega et al. 2021), demonstrate a need for the collection and use of marine/riverine particulate organic carbon, $\delta^{15}N$ in seawater nitrate, and/or compound-specific analysis of nitrogen isotopes of individual amino acids, and not just bulk nitrogen isotopes in seals alone to assess trophic position . Compound-specific isotope analysis (CSIA), such as $\delta^{13}C$ values of fatty acids and $\delta^{15}N$ and $\delta^{13}C$ of amino acids, can be helpful to disentangle baseline isotope variation through space or time from differences related to feeding or food web variation. Such approaches may be particularly useful in temporal studies where baseline values of $\delta\ ^{15}N$ and $\delta^{13}C$ were not sampled and archived.

The third objective of Chapter 4 was to investigate the influence of climate change on contaminant accumulations using a space-for-time substitution (SFT) approach. When accounting for differences in dietary patterns across locations, the results suggested a potential diet-driven decrease in THg, DDT, and PBDE concentrations and a potential increase in newer POPs, such as some PFAS contaminants, if the diets of the Arctic ringed seals become more similar to the diets of the sub-Arctic ringed seals in the future. While there is some critique about using SFT (Damgaard 2019), these results predict some potential for impacts of climate change on ringed seal contaminant loads and represent a good starting point for future climate change-contaminant studies. By combining these predictions with available ringed seal temporal data from the same four study locations, more robust conclusions will be formed in the future.

General Conclusion

This thesis investigated SCCP accumulation in an amphipod, marine fish and ringed seal food web and developed insight into the effects of climate-change-driven feeding shifts on contaminant concentrations in ringed seals across the Canadian Arctic. In Chapter 3, SCCPs were detected in all sampled species with relatively high levels found in the marine fish. Despite these high levels, the biomagnification potential of SCCPs from fish to ringed seals appears to be limited. This Chapter demonstrates the importance of monitoring SCCPs in the Canadian Arctic and highlights the need for toxicity and biotransformation studies in Arctic marine wildlife. Using FA and SI as dietary tracers, Chapter 4 found feeding differences between sub-Arctic and Arctic (Resolute Bay) ringed seals. These differences are likely related to the higher consumption of invading southern species in the diets of sub-Arctic ringed seals. Diet differences were linked to higher levels of PFASs in the sub-Arctic seals and suggest that future climate-driven diet shifts in Arctic seals may increase levels of new POPs. This chapter provides a starting point for future detailed studies of contaminant-climate interactions in ringed seals and demonstrates the need to disentangle feeding effects from isotopic baselines when using bulk stable isotopes to interpret contaminant spatial and temporal patterns. Combined, these two chapters successfully addressed the thesis objectives and contributed to an improved understanding of contaminants in Arctic marine wildlife.

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