LEVELS OF PERFLUOROOCTANE SULFONATE IN THE BEAUFORT SEA SHELF FOOD WEB: MODELLING IMPACTS OF CLIMATE CHANGE AND IMPLICATIONS FOR HUMAN HEALTH

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ABSTRACT

Perfluorooctane sulfonate (PFOS) is a persistent organic pollutant of global concern due to its capacity to bioaccumulate, biomagnify, and cause adverse human health effects. It is ubiquitous in the Arctic environment, particularly biota, and PFOS exposure for Inuit has been associated with the consumption of marine country foods such as beluga and ringed seals. Climate change can increase the exposure to and bioaccumulation of pollutants in marine organisms, partly through the alteration of ecosystem trophodynamics. To date, there has been limited research on dietary exposure to contaminants via marine country foods for Inuvialuit in the Western Arctic relative to other regions of Inuit Nunangat, particularly in the context of future climate changes. Further, modelling of climate change induced changes to trophodynamics and subsequent impact to contaminants in marine ecosystems has not examined any perfluoronated compound, including PFOS. To address these gaps, ecological modelling software Ecopath with Ecosim was used to assess PFOS concentrations in the Beaufort Sea Shelf marine food web under two climate change scenarios. Projected sea surface temperature and sea ice cover data for the Beaufort Sea corresponding to IPCC AR6 Shared Socioeconomic Pathways (SSP) 1-2.6 and 5-8.5, representing respective low (2°C of warming) and high (4°C of warming) carbon emission scenarios, were used to inform the climate change scenarios. Empirical toxicokinetic and environmental concentration data for PFOS were used to simulate the model area as of the model start year 2010. Model bias analysis indicates that the model simulates PFOS concentrations fairly representative of the species with available data, with model bias values ranging from 0.01-1.7. The projected amplification of PFOS in key species was between 9-19% and 11-30% by 2100 under SSP1-2.6 and SSP5-8.5, respectively, relative to a no-climate change control scenario. These projected PFOS concentrations under each climate change scenario correspond to marginal increases in dietary exposure to PFOS for Inuvialuit from beluga, Arctic char, and whitefish consumption compared to no climate change. Hazard quotients (HQ) for the dietary exposure to PFOS from these country foods remain low; HQ <0.01, indicating that any increases in PFOS concentrations in key country foods driven by climate change are unlikely to pose a health risk to the population level for this region. As this modelling approach has not yet been used for perfluoroalkyls, this study serves as a first attempt at building a model for PFOS and provides novel insight into the impacts of climate change on this class of compounds. The findings from this study may also be useful to Inuvialuit communities for making informed health decisions, prioritization for country food biomonitoring programs, and developing climate change adaptation plans.

LIST OF ABBREVIATIONS

AMAP – Arctic Monitoring and Assessment Programme
ARI – Aurora Research Institute
AR6 – Sixth Assessment Report (of the IPCC)
ATSDR – Agency for Toxic Substances and Disease Registry
BSS – Beaufort Sea Shelf
BW – Body Weight
CMIP6 – Coupled Model Intercomparison Project 6
UK COT – United Kingdom Committee on Toxicity
DAR – Direct Uptake Rate
DEE – Dietary Exposure Estimates
DIV – Dietary Intake Value
DK EPA – Danish Environmental Protection Agency
AE – Assimilation Efficiency
EFSA – The European Food Safety Administration
ESM4 – Earth System Model 4.1
EU – European Union
EwE – Ecopath with Ecosim
FSANZ – Food Standards Australia New Zealand
GFDL – Geophysical Fluid Dynamics Laboratory
HBV – Health-Based Value
HC – Health Canada
HQ – Hazard Quotient
HTC – Hunters and Trappers Committee
IARC – International Agency for Research on Cancer
IHS – Inuit Health Survey
IPCC – UN Intergovernmental Panel on Climate Change
IPY – International Polar Year
IRC – Inuvialuit Regional Corporation
ISR – Inuvialuit Settlement Region
MB – Model Bias
MTT – Mean Tolerance Temperature
NOAA – National Oceanic and Atmospheric Administration
NOAEL – No Adverse Effect Level
PCB – Polychlorinated Biphenyl
PCE – Proportion of Contaminant Excreted
PFAS – Poly- and Perfluoroalkyl Substances
PFCA - Perfluorinated Carboxylic Acids
PFOA - Perfluorooctanoate Acid

PFOS – Perfluorooctanoate Sulfonate
POD$_{HEQ}$ – Human-Equivalent Points of Departure
POP – Persistent Organic Pollutant
RCP – Representative Concentration Pathway
SIC – Sea Ice Cover
SSP – Shared Socioeconomic Pathway
SST – Sea Surface Temperature
TDI – Tolerable Daily Intake
TL – Trophic Level
TMF – Trophic Magnification Factor
UN – United Nations
US EPA- United States Environmental Protection Agency

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CHAPTER 1. INTRODUCTION

1.1 Background

Poly- and Perfluoroalkyl Substances (PFAS) are a large group of chemicals classified as persistent organic pollutants (POPs) due to their resistance to breaking down in the environment (Blum et al., 2015; Landrigan et al., 2020; Sunderland et al., 2019; Zhang et al., 2019). They have the capacity to bioaccumulate in organisms and biomagnify in food webs, which can lead to high concentrations in species at higher trophic levels (Blum et al., 2015; Landrigan et al., 2020; Sunderland et al., 2019; Zhang et al., 2019). One type of PFAS that has been produced for decades is perfluorooctane sulfonate (PFOS). Exposure to PFOS can cause many adverse effects, including disrupted endocrine, immune, and liver function (Blum et al., 2015; Sunderland et al., 2019; Zeng et al., 2019). Despite its production being heavily regulated in most countries, it has been transported globally and is found in all ecosystems, including the Arctic. For example, PFOS has been detected in seawater, belugas, ringed seals, and other marine species in the Beaufort Sea (Benskin et al., 2012; Butt et al., 2008; Martin, Smithwick, et al., 2004; Powley et al., 2008; Smythe et al., 2018). PFOS in Inuit country foods has been previously analyzed and found to be highest in marine mammals at higher trophic levels: polar bear, beluga, and ringed seal (Ostertag, Tague, et al., 2009). Biomonitoring efforts in Nunavik have found that PFOS blood concentrations in Inuit are associated with the consumption of marine country foods (Caron-Beaudoin et al., 2020). Climate change can alter how POPs move through and are taken up by the environment, specifically in marine ecosystems (Alava et al., 2018; McKinney et al., 2015).

The Arctic is one of the most vulnerable environments to climate change as it has warmed at three times the rate of the global average (AMAP, 2021). Previous modelling studies have explored the impacts of climate change on other POPs in marine food webs. However, none to date have looked specifically at PFOS or other PFAS. There is also limited research directly linking these potentially altered contaminant pathways in marine food webs to dietary exposure in humans through the consumption of seafood.

This research aims to address these research gaps, using ecological modelling tools and toxicological data reported in the literature to predict the impact of climate change on the bioaccumulation of PFOS in the Beaufort Sea Shelf food web, and estimate the potential risk of dietary exposure to PFOS through the consumption of marine country foods. The marine ecological modelling software Ecopath with Ecosim (EwE) and module Ecotracer were used to trace PFOS through the Beaufort Sea Shelf food web under two different climate change emissions scenarios. The model output provides an estimate of possible PFOS concentrations in key species in the food web under the climate change scenarios. Dietary exposure estimates to PFOS were calculated using the modelled concentrations in species consumed as country foods and corresponding dietary intake information reported for the Inuvialuit Settlement Region in the International Polar Year Inuit Health Survey (Egeland, 2010; Laird et al., 2013). These exposure estimates were then used to calculate hazard quotients for each country food based on Health Canada's tolerable daily intake limit for PFOS, to determine where future exposures from each country food may fall relative to a health-based guidance value and health risks under each climate change scenario (Health Canada, 2018). These results may help organizations and communities in the Inuvialuit Settlement Region to make informed health decisions and climate change adaptation plans.

1.2 Literature Review

Climate Change and Contaminants

The impacts of climate change on the marine environment and ecosystems are far-reaching. Climate change impacts the biogeochemical processes of oceans, including sea surface temperature, pH, oxygen levels, currents, sea ice melt, and many others (Alava et al., 2017; Climate Change 2014 Synthesis Report, 2015). Climate change has also been found to affect ecosystems and food webs; changing migration patterns, fish size and distribution, trophic level structures, and predator-prey dynamics (Alava et al., 2017, 2018). These functional and structural changes to food webs have implications for the ecosystem services they provide, such as food sources through commercial and subsistence fishing and hunting (Alava et al., 2017). These changes also present pathways for the interaction of food web alterations due to climate change and the exposure of marine animals to environmental contaminants (Alava et al., 2017; Ma et al., 2016). Emerging research findings show the potential for climate change to impact bioaccumulation and biomagnification of contaminants such as methyl mercury and POPs in ecosystems by way of altered exposure, environmental transport, and persistence of contaminants (Alava et al., 2017; Ma et al., 2016).

The Arctic has warmed at three times the rate of the global average, causing its environment to be more vulnerable to climate change (AMAP, 2021). The environmental changes observed in the Arctic marine environment, such as increased sea surface temperature, ocean acidification, loss of sea ice, and many others, have the potential to alter the transport and fate of contaminants in the environment (AMAP, 2017; Ma et al., 2016). Inland environmental changes such as permafrost thaw and glacial melt also present avenues for increased transport of legacy contaminants to the oceans (Ma et al., 2016). The Beaufort slope and shelf have been reported as likely to experience elevated amounts of terrestrial sediment input due to extended ice-free periods resulting from climate change (Goñi et al., 2013). PFOS has been detected in glaciers and lakes in the Canadian Arctic along with all other environmental media (Lescord et al., 2015; MacInnis et al., 2017, 2019; Muir et al., 2019). Thus, major rivers like the Mackenzie present potential avenues for increased transport of PFOS to the marine environment.

Studies conducted across the Arctic have reported potential climate change induced changes to POP levels in food webs, such as increased POP levels due to increased temperatures (Macdonald et al., 2005), changes to POP and mercury exposure, and increased contamination in polar bears and ringed seals due to less sea ice (McKinney et al., 2009), and potential increases in POP levels in polar bears due to limited access to prey, prolonged fasting periods, and habitat loss from sea-ice reduction (Jenssen et al., 2015). Modelling work to assess the impact of climate change on the accumulation of mercury and PCBs in a northeastern Pacific marine ecosystem found alterations in PCB accumulation throughout the food web under climate change scenarios, and increased concentrations in apex predators under a high-emissions climate change scenario (Alava et al., 2018). These studies provide significant evidence for interactions between the biogeochemical and food web alterations from climate change and increased or altered exposure of Arctic marine animals to POPs.

Given the recorded impacts of climate change on other POPs in the Arctic and marine ecosystems, it is possible that climate change is also altering the movement of PFAS through marine ecosystems. Due to the chemical differences between more water-soluble PFAS and other more lipid-soluble POPs, such as PCBs, the impacts of climate change on the latter cannot be applied to PFAS in similar ecosystems. No modelling studies have investigated the impacts of climate change on any type of PFAS in marine ecosystems, which is a gap in our understanding of climate change-contaminant interactions (Smythe et al., 2018). Given this ubiquity of PFOS in the Arctic environment and the magnitude of climate changes occurring in the Arctic, it is possible that there are interactions between climate change-induced food web alterations and PFOS levels in species within the food web that warrant further research.

Perfluorooctane Sulfonate

Poly- and perfluoroalkyl substances (PFAS) are a group of synthetic, organic chemicals that have been produced for over 60 years for their use as surfactants in various materials such as wetting agents, lubricants, corrosion inhibitors, stain-resistant treatment, and for fire foam extinguishers (Beach et al., 2006; De Silva et al., 2021; Giesy & Kannan, 2002). They are extremely persistent in the environment and have been detected in environments across the globe, including the Arctic and other remote locations (Beach et al., 2006; Giesy & Kannan, 2002; Yamashita et al., 2005). One of the most widely distributed and commonly detected PFAS in the environment is perfluorooctanoate sulfonate (PFOS) (Beach et al., 2006; Giesy & Kannan, 2001). Its production in North America was voluntarily phased out from 2000-2002 by its main producer, 3M, and it was added to the Stockholm Convention Annex B in 2009, restricting its production to only a few exceptions for all signatory states (Idowu et al., 2013; *PFOS, Its Salts and PFOSF: Overview*, 2019; Zhang et al., 2019). Despite this significant reduction in its use globally, PFOS is still among the PFAS that are detected in the highest concentrations in oceans and wildlife (Beach et al., 2006; Giesy & Kannan, 2001; Zhang et al., 2019).

Like the broader group of perfluorinated compounds, PFOS is extremely persistent in the environment as the strong carbon-fluorine bonds lead to low potential for volatilization and enable resistance to hydrolysis, photolysis, microbial degradation, and metabolism by animals (Beach et al., 2006; Giesy & Kannan, 2002). With a Henry's law constant of 2.0×10^{-6}, PFOS is unlikely to volatilize (Beach et al., 2006). However, other compounds known as precursors are more volatile, and can be oxidized into PFOS in the atmosphere (Beach et al., 2006; De Silva et al., 2021; Lin et al., 2020; Muir et al., 2019). These precursors play an important role in PFOS transport and partitioning in the environment and biota as they are ionic and thus more able to transform than the anionic PFOS (Giesy & Kannan, 2002; Glaser et al., 2021; Muir et al., 2019). Some precursors exhibit different partitioning than others, and toxicokinetics are unique to each, which complicates understanding of PFOS partitioning overall between environmental compartments and tissues in various species (Glaser et al., 2021). PFOS has a high capacity to accumulate in organisms, with measured bioconcentration factors up to 200 depending on the species and length of exposure (Beach et al., 2006; Giesy & Kannan, 2001; Muir et al., 2019), and has been found to bioaccumulate in many aquatic and marine species (Casal et al., 2017; Kannan et al., 2001; Martin et al., 2003; Savoca & Pace, 2021). Unlike other POPs that are mostly lipophilic, PFOS is water soluble and therefore unlikely to partition in lipids based on a $\log K_{ow}$ of -1.08, and accumulates primarily in blood and liver tissues in animals relative to other POPs which accumulate primarily in lipid tissues (Beach et al., 2006). It has, however, also been detected at relatively high levels in beluga blubber, providing evidence for a capacity to partition to lipids as well (Kelly et al., 2009; Ng & Hungerbühler, 2014; Ostertag, Tague, et al., 2009). As such, it is suggested that both lipid partitioning and protein binding are important mechanisms for PFOS bioaccumulation (Armitage et al., 2013, 2017; Sun et al., 2022).

PFOS in the Arctic Environment

PFOS has been detected in every environmental compartment of the Arctic despite not being produced in the Arctic (AMAP, 2017; Muir et al., 2019). Of all the perfluorinated compounds found in the Arctic, PFOS has been consistently found in the highest amounts within biota, despite perfluorooctanoic acid (PFOA) being found in higher levels in the abiotic environment, and lower in biota (Muir et al., 2019). Even with this significant regulation and subsequent decreases in production and in use over the past two decades, PFOS is a contaminant of emerging concern identified by the Arctic Monitoring and Assessment Programme (AMAP), along with other PFAS, due to their environmental presence, persistence, and potential adverse human health effects (AMAP, 2021).

The ocean is believed to be the terminal sink for PFAS via surface water run-offs from inland contaminated sites and wastewater (Garnett, Halsall, Vader, et al., 2021; Prevedouros et al., 2006; Zhang et al., 2019). The two mechanisms for PFAS, including PFOS, transport to the Arctic are by 1) ocean currents, and 2) long-range atmospheric transport of precursors with wet and dry deposition directly to the ocean and to snow and glaciers, which release PFAS to the ocean upon melting (Garnett, Halsall, Vader, et al., 2021; Muir et al., 2019; Yeung et al., 2017; Zhang et al., 2017). Oceanic transport is believed to be primarily from the North Atlantic, with prior modelling work estimating that 30% of the PFOS in the North Atlantic Ocean is transported to the Arctic (Zhang et al., 2017). There is evidence of atmospheric deposition in the Canadian Arctic, with PFOS detected in sea ice, lakes, and ice caps in the Canadian Arctic (Cai et al., 2012; Lescord et al., 2015; Veillette et al., 2012; Young et al., 2007). It is also likely that PFOS deposited atmospherically onto sea ice is a significant source to the Arctic Ocean. Studies have found high concentrations of PFOS in the top layer of sea ice, and concentrations in under-ice seawater that are three times higher than those in deeper water (Garnett, Halsall, Thomas, et al., 2021, 2021). This suggests that sea ice and snow melt contribute significantly to surface water concentrations of PFOS in the Arctic Ocean (Garnett, Halsall, Thomas, et al., 2021; Garnett, Halsall, Vader, et al., 2021). Temperature is believed to play an important role in the process of atmospheric transport, with precursors being deposited via precipitation in the winter months and then released with snow and ice melt in spring rather than volatilizing back to the atmosphere (Muir et al., 2019). There is also an association with flux to snowpack in warmer temperatures, which may be associated with greater scavenging from the air with wet snow and/or that the wet snow has a larger surface area for photochemical transformation of precursors, which then are deposited and are released in snow and ice melt in spring (Muir et al., 2019). Given the remoteness of these environmental compartments, PFOS is unlikely to have come from local sources through the use of consumer products, indicating long-range transport via oceans and atmosphere as the most likely source (Muir et al., 2019).

An additional potential source of PFOS in the Beaufort Sea area, specifically, is the Mackenzie River. The Mackenzie River delivers a significant amount of surface and groundwater, inorganic sediment, and terrestrial organic carbon from the Mackenzie basin to the Beaufort Sea (Hoover et al., 2021; Macdonald et al., 1995, 1998). The sediments in the Beaufort slope and Mackenzie basin are reported to be primarily from terrestrial sources, and PFOS levels in the Chukchi Sea sediments are purported to be transported via the Beaufort slope and Mackenzie basin, supporting this possibility (Goñi et al., 2013; Lin et al., 2020). Based on this current literature, oceanic current transport from the North Atlantic, atmospheric deposition of precursors, and possible input from the Mackenzie River are the likely primary sources for PFOS in the Beaufort Sea Shelf region.

Due to this long-range transport, PFOS in the Arctic is not declining proportionally despite the significant global emissions reduction. Atmospheric PFOS levels in the Arctic have been increasing slightly since 2010, with doubling times of 2.8 years observed at Alert, a monitoring station at the northern tip of Nunavut (Muir et al., 2019). Within the Beaufort Sea, PFOS has been measured in seawater, beluga, ringed and bearded seal, and various species of fish (Benskin et al., 2012; Powley et al., 2008; Smythe et al., 2018; Yeung et al., 2017). Temporal patterns of C7-14 perfluorinated carboxylic acids (PFCA), a grouping that includes PFOS, in beluga monitoring data saw decreases from the mid-1990s to early 2000s, followed by increases from around 2005-2015 (Muir et al., 2019). In ringed seals, PFCA levels have been steadily increasing since the 1970s (Muir et al., 2019). Given the emissions reduction from regulation, these temporal trends in PFOS and precursors suggest that transport from other regions, primarily via ocean currents and long-range atmospheric transport, are the main source of PFOS in the Arctic and are driven by other factors aside from just emissions (Armitage et al., 2017; Muir et al., 2019; Yeung et al., 2017). Further, the temporal trends are not consistent across the entire Arctic environment or across species, suggesting the influence of local climate or ecological drivers in the transport and uptake of PFOS in Arctic environmental compartments.

Human Health Impacts of PFOS

Numerous adverse health effects across many organ systems are associated with exposure to PFOS. These include impaired liver function and hypertrophy, changes in thyroid hormone levels, impaired kidney function, immune suppression, lipid and insulin dysregulation, neurodevelopmental delays, fertility challenges, delayed pregnancies, and delayed onset puberty (Fenton et al., 2021; Zeng et al., 2019). The mechanisms of PFOS toxicity are not entirely understood, but are believed to be caused by oxidative damage to cells (Sunderland et al., 2019;

Zeng et al., 2019). It is thought that PFOS interrupts fatty acid oxidation, resulting in the increased production of reactive oxidative species (Zeng et al., 2019). The presence of higher amounts of oxidative species in cells causes oxidative damage to DNA, which disrupts cellular function and replication and can cause cell death (Zeng et al., 2019). The altered cellular function and/or cell death in the affected organs and tissues across many systems causes the widespread observed health effects from exposure (Zeng et al., 2019). PFOS toxicity endpoints have been evaluated in greater detail using animal models, specifically hepatocellular hypertrophy, thyroid hormone disruption, immune suppression, developmental toxicity, tumor induction, and obesity (Fenton et al., 2021; Health Canada, 2018). Health Canada has established two health-based values (HBV) for non-cancer critical health effects of PFOS exposure on the liver and thyroid. Human-equivalent points of departure (POD_{HEQ}) were calculated using the greatest dose or level at which there is no observed adverse effect (NOAEL) for the critical effects of hepatocellular hypertrophy and thyroid hormone changes in rats and monkeys, respectively. The respective POD_{HEQ} and appropriate species-conversion factors were used to calculate tolerable daily intake (TDI) values considered to be protective against the human health effects of PFOS exposure on the liver and thyroid. Immune function, neurodevelopmental, and lipid levels were also evaluated. However, a lack of consistent endpoints did not allow for clear dose-response relationships to be determined for those health effects. Of all the effects evaluated, those observed in the liver occurred at the lowest dose. Thus, the TDI for liver effects is considered to also be protective against PFOS effects on the thyroid, immune system, neurodevelopment, and lipid levels (Health Canada, 2018).

The International Agency for Research on Cancer (IARC) completed carcinogenicity evaluations of PFOA and PFOS in December 2023, classifying PFOS as Group 2B – possibly carcinogenic to humans (Zahm et al., 2024). The Group 2B classification of PFOS was based primarily on mechanistic evidence, which was considered strong in test systems, including epigenetic alterations, immune suppression, and other indicators of carcinogenicity in exposed humans (Zahm et al., 2024). The evidence for cancer in experimental animals (liver) was considered limited, and in humans (testis, breast, thyroid) was considered inadequate, meaning in both cases that the link between PFOS exposure and cancer incidence was not strong enough to be considered causative based on the research available at this time (Zahm et al., 2024). Therefore, while there are observed associations between PFOS exposure and the risk of specific and overall cancers, there is currently no consistent epidemiological link (Health Canada, 2018; Sunderland et al., 2019; Zahm et al., 2024; Zeng et al., 2019). Given that the outcome of the IARC classification, and the evidence that prompted it, does not rule out carcinogenicity, some agencies are now considering HBVs for PFOA and PFOS for cancer risk (Wee & Aris, 2023). The US EPA is currently developing a cancer slope factor for PFOA and PFOS (Wee & Aris, 2023). Health Canada previously established an HBV for cancer risk, using a benchmark dose approach for hepatocellular tumors (adenomas and carcinomas) in rats to determine a POD_{HEQ} and subsequent TDI for PFOS (Health Canada, 2018). However, this was done before IARC evaluated PFOS for carcinogenicity and established a TDI as the most appropriate HBV, assuming that PFOS was non-mutagenic (Health Canada, 2018). As such, Health Canada's TDI for cancer risk is not based on the most recent evidence available, which indicates the mutagenic properties of PFOS and a non-negligible association with cancer risk.

Exposure to PFOS occurs primarily through consumer products and the environment. Contact with consumer products such as clothing, food packing, upholstery, carpets, cleaners, and other household products, present inhalation, dermal, and direct ingestion exposure pathways (Sunderland et al., 2019). Environmental pathways include the ingestion of contaminated water and seafood (Sunderland et al., 2019). Contaminated drinking water is believed to be a significant source of PFAS for populations living near contaminated sites such as airports and firefighting bases, however, is not a significant exposure for the general population in Canada (Health Canada, 2018; Sunderland et al., 2019). Many countries, including Canada, have drinking water guidelines for PFOS and perfluorooctanoic acid (PFOA), the two most common PFAS detected in the environment. The current guidelines have established the maximum acceptable concentration of PFOS in drinking water to be 0.6ug/L (Health Canada, 2018). Contaminated seafood is a second direct ingestion route that serves as a significant exposure pathway (Health Canada, 2018; Sunderland et al., 2019). The European Food Safety Authority estimated that seafood could account for up to 86% of exposure to PFAS in adults (EFSA Panel on Contaminants in the Food Chain et al., 2018). The significance of this exposure route varies across populations, but is expected to be significant for those whose diet is composed of significant amounts of seafood (Caron-Beaudoin et al., 2020; Sunderland et al., 2019). This is particularly relevant for many Indigenous Peoples, including Inuit across the Arctic, who consumed higher amounts of fish or seafood than non-Indigenous or non-Inuit populations.

Inuit Country Foods

Inuit living across the Canadian North have an intimate relationship with the land and rely on traditional or country foods that are locally harvested as sources of essential nutrients (Kenny, Fillion, et al., 2018; Wesche & Chan, 2010). Country foods are not only a source of essential nutrients, they hold significant cultural and spiritual value for Inuit and are an important contributor to overall health and wellness (Wesche & Chan, 2010). Country foods are also, however, a source of contaminants for Inuit across the Canadian Arctic, with climate change impacts also affecting the transport and exposure pathways of various contaminants in country foods (Kenny, 2019; Wesche & Chan, 2010). Previous work measured PFOS concentrations in Inuit country foods in Nunavut and found that PFOS levels were highest in animals at higher trophic levels, such as polar bears, beluga, and ringed seals (Ostertag, Tague, et al., 2009). This study also found that the primary sources of PFOS exposure from marine country foods came from beluga muktuk and Arctic char (Ostertag, Tague, et al., 2009). Further, country foods contributed to a significantly greater proportion of exposure to perfluorinated compounds than market foods (Ostertag, Tague, et al., 2009). It was noted as well that PFOA was rarely detected in country food samples, which aligns with previous reports of PFOA levels in biota being lower than PFOS and other PFAS (Muir et al., 2019; Ostertag, Tague, et al., 2009). Even so, the health risk assessed from these dietary exposure estimates was of minimal concern given the exposure estimates were lower than the provisional TDI set by the German Drinking Water Commission, in the absence of a TDI from Health Canada at the time of the study (Ostertag, Tague, et al., 2009).

In other regions of Inuit Nunangat, biomonitoring efforts are ongoing to better characterize PFOS exposures in Nunavik compared to the general Canadian population. It is estimated that between 98-100% of Canadians have detectable blood serum concentrations of PFOS (Health Canada, 2021). PFOS is found in serum, plasma, kidneys, and the liver, and has a half-life of roughly 4.8 years (Health Canada, 2021; Olsen et al., 2007). In a recent survey of pregnant Inuit women in Nunavik, PFOS serum concentrations were 1.8 times higher than non-Inuit women of the same age measured in the Canadian Health Measures Survey for the same time period (Caron-Beaudoin et al., 2020). Of the various PFAS measured, PFOS was detected at the highest concentrations in serum samples (Caron-Beaudoin et al., 2020). The study also compared PFOS serum concentrations with marine country food consumption using omega-3, -6, and polyunsaturated fatty acids as biomarkers. There was a strong, significant positive correlation between these fatty acid biomarkers and PFOS serum concentrations, indicating an association between consumption of country foods and PFOS serum concentrations, providing further evidence for seafood, and in this case of marine country foods, as sources of PFOS exposure (Caron-Beaudoin et al., 2020). These findings were consistent with an earlier study that also found an association between PFOS exposure and fish and marine mammal consumption for Nunavik Inuit (Dallaire et al., 2009).

While these studies provide insight into the link between marine country food consumption and dietary exposure to PFOS in Nunavut and Nunavik, these studies in the eastern Arctic cannot be applied to the western Arctic as there are regional variations in contaminant concentrations in country foods as well as country food consumption patterns. Much less research has been done on PFOS in country foods in the Inuvialuit Settlement Region (ISR). To begin addressing this knowledge gap and a desire from Inuvialuit communities to better understand contaminants in country foods, a multidisciplinary project called Country Foods for Good Health was launched in the ISR in 2020 to generate regional contaminant data in Inuvialuit country foods and to understand best practices desired by communities for how to communicate information about contaminants in country foods (Gyapay et al., 2022). The current study aims to address the knowledge gaps and community needs as well, offering a novel perspective into the possible future climate change-food web-contaminant interactions specific to PFOS and to the western Arctic, as well as the implications of this for potential future health risks from PFOS in country foods.

1.3 Research Goals

This study had two main overarching goals. The first was to apply ecological modelling methods to provide insight into how climate change may impact the accumulation of PFOS in an Arctic marine food web under two climate change emissions scenarios. The application of ecological modelling to a type of PFAS helps to characterize how climate change impacts trophodynamics and may change the movement of this class of compounds within an ecosystem in the future, an area of research that is not well understood.

The second overarching goal was to use these modelling results to conduct a preliminary risk assessment for dietary exposure to PFOS from the consumption of marine country foods under the two climate change emissions scenarios. There are limited studies in the ISR addressing the relationship between climate change and PFOS levels in marine country foods and fewer that provide insight into potential future exposure under climate change scenarios. This research provides information about the potential impacts of climate change on contaminants in country foods, with the aim of supporting communities in the ISR to make informed health decisions and climate change adaptation plans.

1.4 Research Questions, Objectives and Hypotheses

Two research questions have been developed to guide the research. A full chapter is dedicated to answering each of these research questions.

Research Question 1

How will the bioaccumulation of PFOS in the Beaufort Sea Shelf food web change from baseline under two climate change scenarios, low-emissions and high-emissions, over the period of 2010-2100?

Research Objective 1

Estimate potential future levels of PFOS in Beaufort Sea country foods under two climate change scenarios.

Hypothesis 1

PFOS bioaccumulation will be amplified and will increase concentrations in higher trophic level species in the Beaufort Sea Shelf food web under the higher emissions scenario due to increased sea surface temperature and ice melt. Under the low-emissions scenario, PFOS bioaccumulation will also be amplified, and concentrations in higher trophic level species will increase due to increased sea surface temperature and sea ice melt, but at a lesser magnitude.

Research Question 2

How will the level of PFOS intake from the consumption of marine country foods change from baseline under two climate change scenarios, low-emissions and high-emissions, over the period of 2010-2100, and will this pose a risk to Inuit health?

Objective 2

Estimate dietary exposure and calculate hazard quotients under two climate change scenarios

Hypothesis 2

Increased bioaccumulation of PFOS under the high emissions scenario in higher trophic levels species will pose elevated health risks for Inuit because oral intake of PFOS will be increased at an unchanged level of consumption. Changes to PFOS bioaccumulation under the low emissions scenario will not markedly increase dietary exposure estimates or risk compared to baseline.

1.5 Conceptual Framework

Figure 1.1 is a conceptual framework, adapted from Alava et al., (2018), illustrating the key modelling processes and their links with dietary exposure data. The modelling software Ecopath with Ecosim is the primary modelling tool used to investigate the relationship between climate change and PFOS in the Beaufort Sea Shelf marine ecosystem and predict the concentrations of PFOS in functional groups under the climate change scenarios. The model components are represented by the rectangular boxes, with the base Ecopath model in the middle and empirical data inputs to the model Ecosim and Ecotracer on the left and right, respectively. In Ecosim, climate change data from the National Oceanic and Atmospheric Administration's (NOAA) Geophysical Fluid Dynamics Laboratory (GFDL) Earth System Model 4.1 (ESM4) for sea ice cover (SIC) and sea surface temperature (SST) under the International Panel on Climate Change 6th Annual Report (IPCC AR6) Shared Socioeconomic Pathways (SSP) 1-2.6 and 5-8.5 were used as the two climate scenarios. SSP1-2.6 describes the 'low emissions' climate change scenario, where global policy action results in significant greenhouse gas emission reduction keeping global warming levels below 2°C by the end of the century (Chen et al., 2021). SSP5-8.5 describes what is colloquially called a worst-case scenario, where limited global action is taken to curb greenhouse gas emissions resulting in global warming levels above 4°C by the end of the century (Chen et al., 2021). These two scenarios are often used in climate change modelling to represent the bookends of 'ideal' climate change mitigation policies (SSP1-2.6) and no climate change mitigation policies (SSP5-8.5) (Chen et al., 2021; Lee et al., 2021). In Ecotracer, empirical contaminant concentrations in the seawater and functional groups, base inflow rate to the system, and toxicokinetic values for functional groups were included. The output from model runs, bolded rectangular box, are predicted PFOS concentrations for functional groups in the food web under the two climate change scenarios. These predicted concentrations along with dietary intake data and tolerable daily intake levels, each in oval shapes, were used to calculate dietary exposure estimates and hazard quotients to provide a preliminary risk assessment for PFOS under the climate scenarios. The three key outputs from the study are represented by bolded shapes: predicted PFOS concentrations, dietary exposure estimates, and hazard quotients.

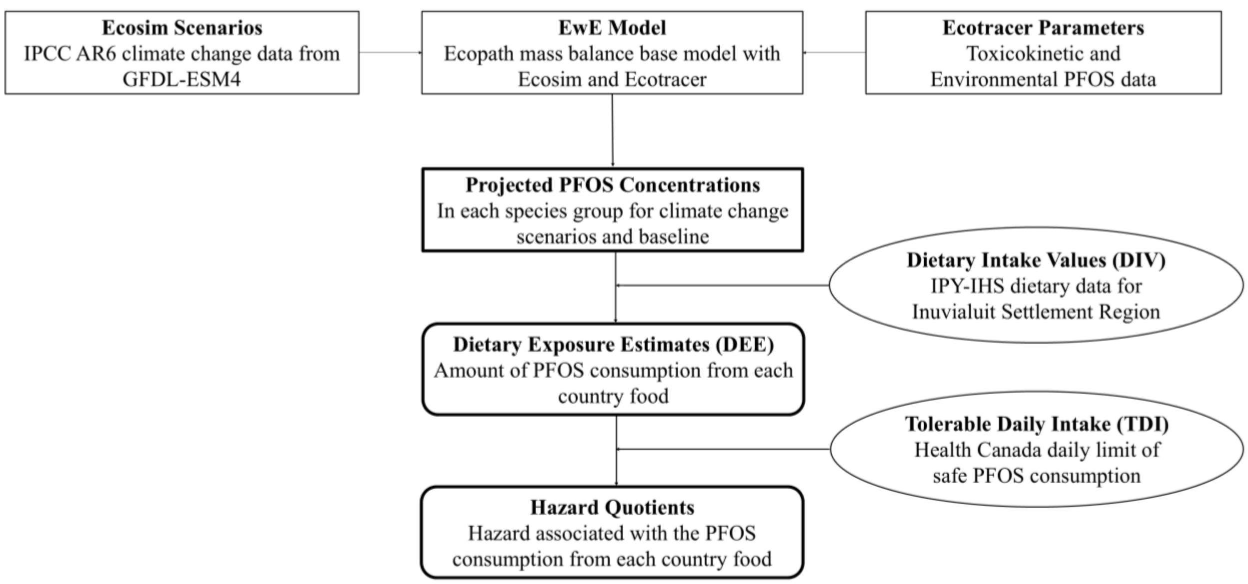

Figure 1.1. *Conceptual framework to visualize the modelling approach and dietary exposure risk assessment for PFOS.* Adapted from Alava et al., (2018).

1.6 Ethics and Research License

This research consisted solely of secondary data analysis. As such, no primary data was collected from any person or the land. Therefore, no ethics approval was required as per the Research Ethics Board of the University of Ottawa. All data used was from published literature, which includes the contaminant levels and toxicokinetic properties used to create the model and the dietary intake data collected as part of the IPY-IHS for the dietary exposure estimates and hazard quotient calculations. Nevertheless, best practices were followed when doing research in the Inuvialuit Settlement Region.

Following these best practices, a research license application was submitted and granted by the Aurora Research Institute (ARI) in August 2022 (Figure A-1). A progress update was provided to the ARI in June of 2023 for publication in their annual research update for projects in the Northwest Territories. Additionally, a letter of support was also obtained from the Inuvialuit Regional Corporation (IRC) for the project and submitted with the research license application (Figure A-2).

Even though this project was not part of a larger research initiative, it aligned with similar projects, specifically the Country Foods for Good Health project based out of the University of Waterloo and the Influences of Environmental Change on Levels and Trends of Methylmercury in the Beaufort Beluga Food Web project based out of the University of British Columbia. As part of this alignment, a one-page research summary was prepared (Figure A-3) to be shared with organizations within the ISR. The research summary has not yet been shared at the time of submitting this thesis, as it will be shared as part of a larger PFAS package of research results from the Country Foods for Good Health project. Sharing the research summary with the Country Foods for Good Health project aims to help contextualize this research project and avoid engagement fatigue for communities, given the limited capacity of communities relative to the number of ongoing research projects seeking input and involvement. The summary will be shared with each of the six Hunters and Trappers Committees (HTC) in the ISR, the Inuvialuit Game Council, the IRC, and the Northwest Territories Department of Health and Social Services. A one-pager was chosen as the best method to share these results due to the high cost of travelling to each of the six ISR communities, the limited availability of community organizations to meet and discuss results, as well as the nature of the results. As discussed further in later chapters, the potential future health risks evaluated through this study were found to be low. If the findings had suggested the potential for higher health risks in the future, meeting with communities and organizations would have been chosen as the best way to disseminate results to ensure the health risks were communicated with care and accuracy, and to avoid misinterpretations and causing any fear around country food consumption. This has been noted as a problem in the past, where risks were not appropriately communicated by researchers, and subsequently, communities received the message that country foods were unsafe to consume. Given the results of this study describe low health risks in the future from PFOS dietary exposure and also used modelling to predict possible future health risks aligned with potential climate change scenarios, the findings are also not reflective of the current risk to health and therefore should not be used to make any individual decisions, but rather can inform health and climate change planning at the regional level.

Following this introduction chapter, the thesis will be made up of two chapters detailing the modelling and health risk assessment, respectively, followed by a conclusion chapter.

CHAPTER 2. MODELLING THE EFFECTS OF CLIMATE CHANGE ON TROPHODYNAMICS AND PFOS IN THE BEAUFORT SEA SHELF ECOSYSTEM

2.1 Introduction

Environmental contaminants produced for various uses and functions have been of increasing global concern in past decades. An ever-growing group of chemicals called per- and polyfluorinated substances (PFAS) are among these, having been produced since the 1940s and made up of close to 5000 different compounds (Blum et al., 2015; De Silva et al., 2021; Giesy & Kannan, 2002). PFAS is of growing concern due to their extreme persistence in the environment, owing to the strong carbon-fluorine bonds, resulting in the inclusion of two of the most common types of these compounds, PFOA and PFOS, on the Stockholm Convention on Persistent Organic Pollutants (Blum et al., 2015; Giesy & Kannan, 2001, 2002; Idowu et al., 2013). As a result of their environmental persistence, PFAS can be transported around the globe and have been found in even the most remote locations, such as the Arctic (Muir et al., 2019; Veillette et al., 2012; Young et al., 2007). Exposure to these compounds are also a potential human health risk, associated with a number of adverse health effects on the liver, thyroid, and immune system, among others (Sima & Jaffé, 2021; Sunderland et al., 2019). Given that the number of compounds in this class of chemicals exceeds 4000, many of which are being newly produced as supposed preferable alternatives to some of the older compounds, most of what is known of these compounds is from research done on PFOA and PFOS.

Both PFOA and PFOS are ubiquitous in the environment, including the Canadian Arctic (Muir et al., 2019). PFAS, as a class of chemicals, have been identified as chemicals of emerging concern by the Arctic Monitoring and Assessment Programme due to their aforementioned persistence in the environment and likely adverse impacts on human health (AMAP, 2017). While they are both found in every environmental compartment in the Canadian Arctic, they diverge in their environmental partitioning between compartments. PFOA tends to be detected in higher levels relative to PFOS in abiotic environments such as seawater, lake water, and snow, while PFOS tend to be found in higher concentrations relative to PFOA in biota such as fish and marine mammals (Muir et al., 2019). This is noteworthy as many Arctic Indigenous communities rely heavily on marine country foods, especially Inuit communities for whom beluga, seal, and various fish species are integral parts of their diet for rich nutritional and cultural importance (Kenny, 2019; Ostertag, Tague, et al., 2009). Seafood is one of the primary sources of exposure to PFOS in the general population, and marine country foods have also been identified as significant sources of PFOS exposure for Inuit communities in Nunavik, northern Quebec (Caron-Beaudoin et al., 2020; Dallaire et al., 2009; Sunderland et al., 2019).

Alongside the concern of environmental contaminants is the concern for climate change, and especially climate change-contaminant interactions. The biogeochemical aspects of climate change have been well documented to alter the sources, transportation, and fate of many contaminants within the environment, such as methylmercury and PCBs (Landrigan et al., 2020; Ma et al., 2016). However, emerging research is showing the importance of food web dynamics as the pathway for climate change impacts on contaminants in marine environments (Alava et al., 2017; Booth & Zeller, 2005; Schartup et al., 2019). As ocean temperatures rise, some marine species have been moving northward to remain in habitats that favour their preferred temperature of range resulting in an introduction of new species to some marine ecosystems, and it is possible that some species will have changes to bioenergetics such as growth rate and respiration that can alter uptake and metabolism of contaminants (Fossheim et al., 2015; Sappal et al., 2014). Within the Beaufort Sea Shelf, recent modelling work exploring historical changes to the food web from 1970-2021 under climate change found changes to the diversity in the marine ecosystem overall, as well as altered trophic levels, biomass, and consumption rates of some fish and mammals driven by salinity, sea surface temperature, sea ice cover (Sora et al., 2024). Further, some studies have directly linked temporal trends in other contaminants like methylmercury to climate change impacts of reduced sea ice cover in the Arctic and subsequent impacts to primary production and food source dependence from ice-based to pelagic (Braune et al., 2014; Li et al., 2022). Further, modelling work exploring the association of climate change with food web dynamic shifts and contaminant levels has found that climate change is expected to amplify methylmercury and PCB levels in a Northeastern Pacific marine food web by the end of the century (Alava et al., 2018).

To date, there is limited research exploring such climate change-contaminant interactions via food web dynamics in any type of PFAS compound. This study aims to begin filling this gap, by investigating these dynamics of PFOS within the Beaufort Sea Shelf food web in the Canadian Arctic, using the Ecopath with Ecosim mass balance modelling approach.

2.2 Methodology

Ecopath with Ecosim and Ecotracer Modelling Framework

Ecopath with Ecosim (EwE) is an open-source modelling software suite that is widely used to simulate mass-balance models of marine ecosystems and analyze their trophodynamics (Christensen et al., 2024; Christensen & Walters, 2004). It consists of three routines; Ecopath, to establish a static snapshot of the structure and energy flows of the ecosystem, Ecosim, to incorporate time dynamics and environmental changes to the system, and Ecospace, to incorporate a spatio-temporal dynamic component for different regions or habitats in the system (Christensen et al., 2024; Christensen & Walters, 2004; C. Walters et al., 1997). The EwE framework is based on the original Ecopath model developed by J.J. Polovina in the early 1980s to estimate the biomass and food consumption of functional groups, defined as species or a group of species, within the provided context of the ecosystem (Christensen & Walters, 2005; Polovina, 1984). It has been continuously updated and expanded upon since its creation and has been used to describe the structure and function of ecosystems, measure the effects of environmental changes, trace contaminants, and evaluate policies related to the effects of fishing, marine management, and marine protected areas (Christensen & Walters, 2004, 2005).

One of the core routines of EwE, Ecopath, parameterizes the model of an ecosystem as a mass balance system, such that production and consumption are equal. This is done with two 'master equations' for each functional group within the ecosystem, one describing total production and one describing total consumption. In the simplest terms, these equations are:

$$Production = catches + biomass\ accumulation + net\ migration + predation\ mortality + other\ mortality \tag{Eq. 1}$$

$$Consumption = production + respiration + unassimilated\ food \tag{Eq. 2}$$

The equation components are made up of more detailed parameters. However, this provides a summary of the concepts within the mass balance approach of the Ecopath base model (Christensen et al., 2024; Christensen & Walters, 2004; Sora et al., 2022). The second of the two EwE core routines, Ecosim, inherits parameters from the mass balance Ecopath output along with additionally set parameters to simulate trophodynamics under various environmental changes and drivers (Christensen et al., 2024; Christensen & Walters, 2004). For this study, an Ecopath base model was used with Ecosim and Ecotracer. Ecospace was not used.

Ecotracer is sub-routine within the EwE modelling framework that enables tracking a contaminant through the food web and environment (Alava et al., 2018; Booth, Steenbeek, et al., 2020; Walters & Christensen, 2018). In a mass-balanced model, Ecotracer allows for the contaminant to be followed through the ecosystem based on predator/prey interactions defined in the Ecopath base model and toxicokinetic properties determining gains and losses to the system (Booth, Steenbeek, et al., 2020). It has been used to study a variety of environmental contaminants to date, including stable isotopes, radioisotopes, methyl mercury, PCBs, and microplastics (Alava, 2020; Alava et al., 2018; Booth, Walters, et al., 2020; Booth & Zeller, 2005; Li et al., 2022; Walters & Christensen, 2018). Ecotracer can be used to make estimates about the concentration of a contaminant in a species or functional group presently or in future projections, changes in contaminant levels between groups from trophic interactions, and the importance of dietary uptake vs direct uptake of contaminants to species/groups (Booth, Steenbeek, et al., 2020) With these functionalities, it can be applied to examine important ecotoxicological issues like how environmental changes, climate change, and ecosystem structure changes affect contaminant concentrations and biomagnification, and also employed in a policy context where contaminant concentrations along with consumption rates can be used to estimate exposure levels for humans or referenced to regulatory limits which can impact industry, fisheries, and trade (Alava, 2020; Alava et al., 2018; Booth, Steenbeek, et al., 2020; Booth & Zeller, 2005).

An advantage of the EwE modelling approach is the ability to concurrently analyze how ecological, physical, and chemical pressures impact individual key species, trophodynamics, and the food web as a whole (Townhill et al., 2022). These abilities provide a unique opportunity to simulate various environmental scenarios, such as climate changes, to assess their impacts on ecosystem structure and function and provide insight into management options for the future (Townhill et al., 2022).

Model Area

The area described by the model is the Canadian Beaufort Sea Shelf (BSS), shown in Figure 2.1 (Hoover et al., 2021). The BSS covers an area of around 103 000 km^2 of the continental shelf, extending from the Amundsen Gulf on the east to the Mackenzie Canyon on the west, and the Canada Basin to the north (Hoover et al., 2021).

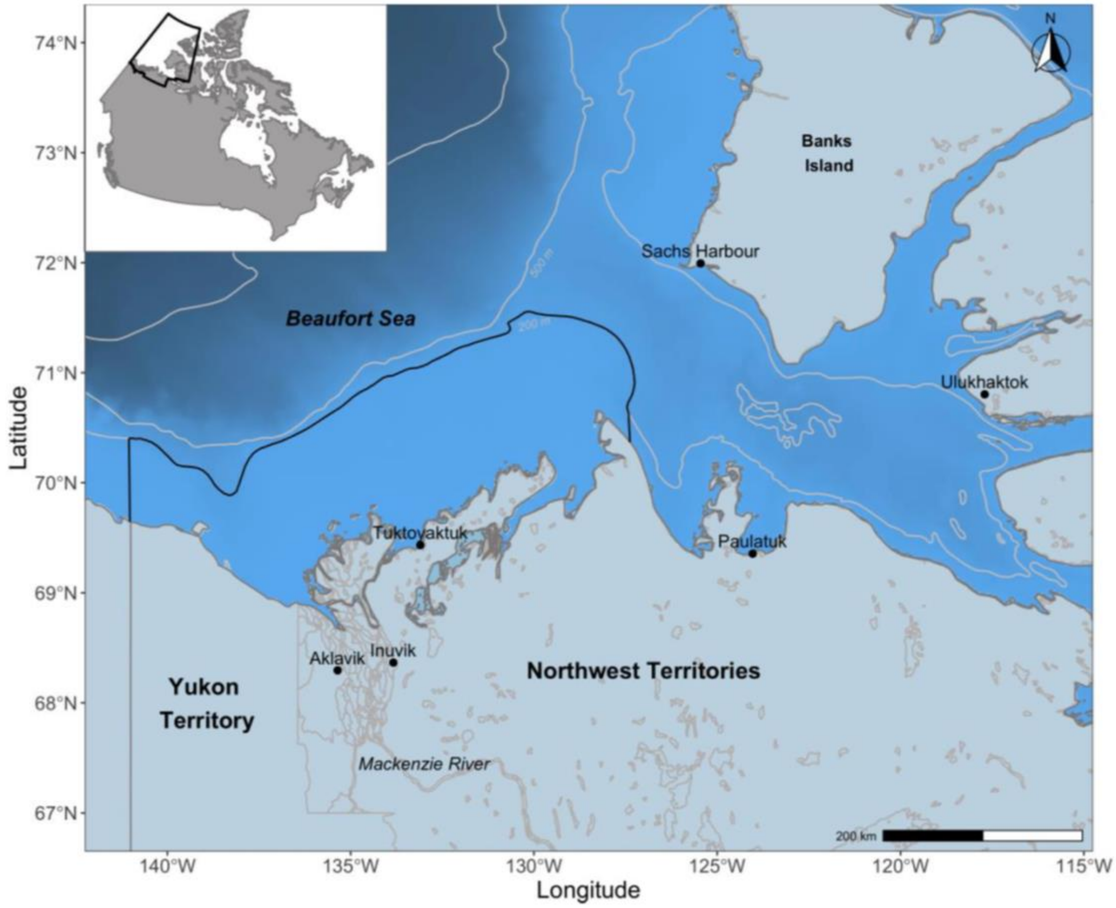

Figure 2.1. *The Beaufort Sea Shelf Model area, outlined in black, and communities within the ISR.* From Hoover et al., (2021).

Arctic shelves make up over 30% of the Arctic Ocean surface area, with the BSS being the largest of the North American shelves in the Arctic, and less than 100m deep throughout most of it (Hoover et al., 2021). It is home to many resident species, such as polar bears and seals, as well as many migratory species, including beluga, bowhead whales, birds, and diadromous char (Hoover et al., 2021). The BSS is an incredibly environmentally significant area; receiving flows into the Mackenzie River basin from the Mackenzie River, the longest river system in North America, and is the site of the first Marine Protected Area in the Canadian Arctic (Hoover et al., 2021; Macdonald et al., 1995). Similar to the rest of the Arctic, climate change has already altered ocean conditions, marine habitats, and ecosystem structures and functions in the BSS that are anticipated to accelerate, with climate models projecting earlier spring ice breakup, longer open-water seasons, upwelling along the Beaufort Sea slope, and changes to primary and secondary production in the food web (Sora et al., 2022, 2024).

The BSS is also located within the homeland of the Inuvialuit, known as the Inuvialuit Settlement Region (ISR) in the Northwest Territories (Hoover et al., 2021). There are six communities within the ISR – Aklavik, Inuvik, Paulatuk, Sachs Harbour (Ikaahuk), Tuktoyaktuk, and Ulukhaktok – all of which rely on the BSS for subsistence hunting and fishing as a vital component of cultural and food security (*Inuvialuit Regional Corporation*, n.d.). Inuvialuit have been stewards of the land and waters from the greater Beaufort Sea for generations and continue to do so within the ISR today. The Fisheries Joint Management Committee and Inuvialuit co-manage many programs with the Department of Fisheries and Oceans for Marine Protected Areas, fisheries management, beluga harvesting, and community-based biomonitoring (*Inuvialuit Regional Corporation*, n.d.).

Beaufort Sea Shelf Food Web

The EwE model for the BSS was built by Hoover et al., (2021) and describes the trophodynamics of 31 functional groups between 1970-2012, as shown in Figure 2.2 (Hoover et al., 2021). The trophic structure described in the model has been verified with stable isotope data, which supports the description of trophic positions by the model (Hoover et al., 2022) A detailed description of the model, including functional groups, including complete species lists, diet composition, biomass (B), production/biomass (P/B), consumption/biomass (Q/B), and ecotrophic efficiency (EE) calculations are published elsewhere (Hoover et al., 2021, 2022).

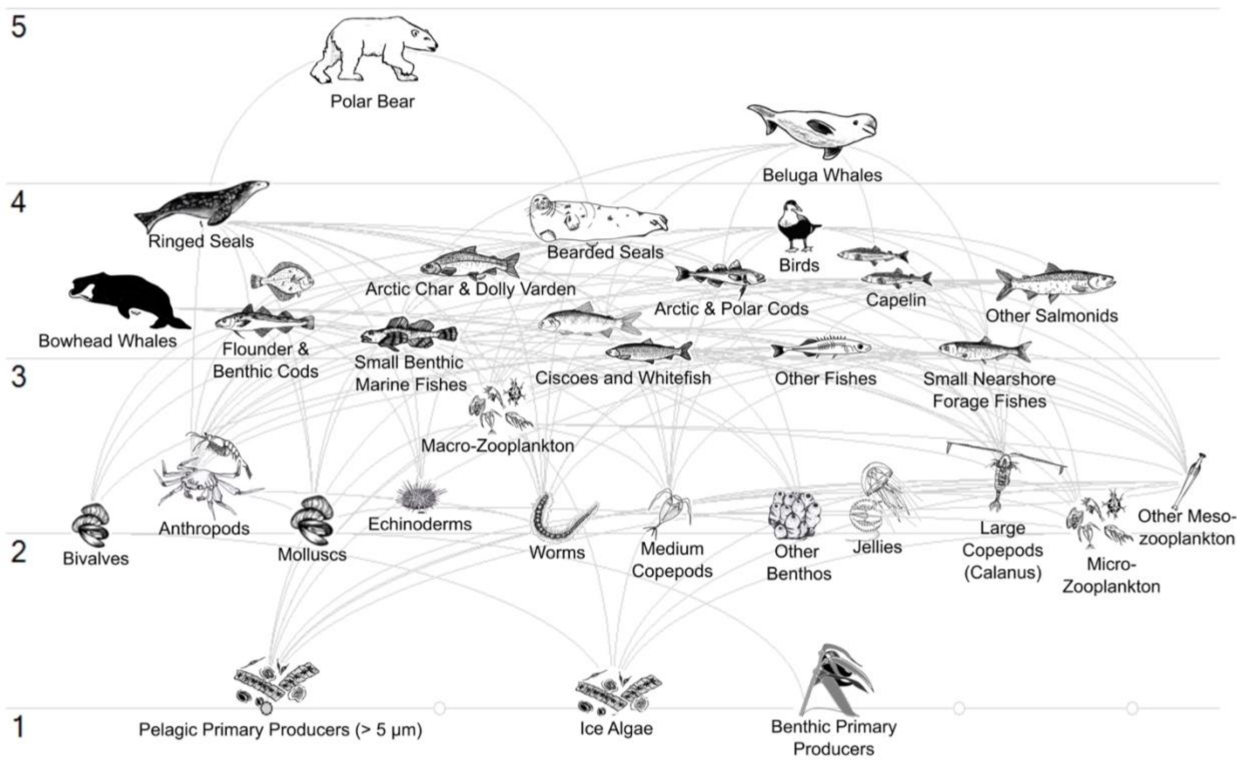

Figure 2.2. *Feeding linkages and trophic levels (y axis) illustrated for the 31 functional groups in the BSS EwE model.* From Hoover et al., (2022).

The model used in this study was adapted from the original 1970-2012 BSS model (Sora et al., 2022), representing the average of the last four years of the original model period, 2008-2012. This 2008-2012 average model has been used with the Ecotracer subroutine to parameterize and simulate the flow of mercury through the BSS ecosystem (Li et al., 2022). The 2008-2012 average model was chosen for this study as it is the most recent baseline period of the ecosystem to use for future-looking projections (Sora et al., 2022).

Parameterizing Ecotracer

Ecotracer requires toxicokinetic parameters for each functional group within the food web to provide an estimate of the static initial conditions in the system (Booth, Steenbeek, et al., 2020).

Ecotracer can provide a dynamic estimate of the movement of a contaminant through the food web over a period of time by simulating contaminant fluxes to give resulting amounts and concentrations (Booth, Steenbeek, et al., 2020). These resulting changes for a given time period is described as:

$$\frac{dA_i(t)}{dt} = \alpha_i - \beta_i A_i(t) \qquad \text{Eq. 3}$$

Where αi, represents the gains (Bq·year-1) in each functional group, βi represents the rate losses (year-1) to each functional group, and Ai represents the amount in each functional group i (Booth, Steenbeek, et al., 2020). Each gains and losses are calculated based on the relationship between various parameters which characterize the movement of the contaminant into and out of the system.

Gains, or intake, is from both the environment and food, and is described by:

$$\alpha_i = u_i B_i C_o + AE_i \sum_{j=prey} Q_{ji} \times \frac{A_j}{B_j} + c_i I_i \qquad \text{Eq. 4}$$

Where C_o represents the environmental concentration ($Bq \cdot km^{-2}$), B_i is the biomass (t) of group *i*, u_i represents the intake/biomass/environmental concentration/year ($km^2 \cdot t^{-1} \cdot year^{-1}$); AE_i is the assimilation efficiency for each group, Q_{ji} is the consumption rate ($t \cdot year^{-1}$) of group *j* by group *i*, A_j is the amount of substance in a group (e.g., Bq), B_j is the prey biomass of each prey item *j* ($Bq \cdot t^{-1}$); c_i is the group biomass concentration ($Bq \cdot t^{-1}$) and I is the immigrating biomass ($t \cdot year^{-1}$).

Losses from the group can be to the environment or to the system, and can be described by:

$$\beta_i C_i = \left(\sum_{j=pred}\left(\frac{Q_{ij}}{B_i}\right) + F_i + MO_i + E_i + m_i + d_i\right) C_i \qquad \text{Eq. 5}$$

Where Qij is the rate of consumption (t·year-1) of group i due to predation by j, Fi is the fishing mortality rate (year-1), mOi (year-1) is other mortality rate (i.e., non-predation mortality), Ei is the emigrating biomass rate (year-1), mi (year-1) is the excretion and/or metabolic rate, and di (year-1) is the physical decay rate. These rates are multiplied by Ci, the amount of contaminant (Bq) in each group i.

With these formulas, the EwE model with subroutine Ecotracer can use the inputted environmental and toxicokinetic parameters for the respective ecosystem to trace contaminants through the food web and provide estimates of contaminant concentrations in each functional group (Booth, Steenbeek, et al., 2020).

Parameterizing Ecotracer involves assigning values to the parameters described previously which inform the gains and losses of a contaminant, in this case PFOS, for each functional group. Some parameters are already characterized in Ecopath from literature and stable isotope studies, such as the group biomass, prey biomass, consumption rate, and mortality rates (Hoover et al., 2021; Sora et al., 2022). The remaining parameters that must be assigned values are split between the environment and biota, respectively the environmental concentration, base volume exchange loss, base inflow rate, and decay rate; and the group concentration, direct absorption rate, and assimilation efficiency. These parameters were defined with estimates from published literature, and where literature was unavailable, calculations based on the EwE equations characterizing the parameter relationships described previously.

Ecotracer Biota Parameters

There are two primary toxicokinetic parameters. First, the direct absorption rate (DAR), also known as an uptake rate in toxicology literature, represents the direct absorption of a contaminant to an organism from the water, such as through respiration (Booth, Steenbeek, et al., 2020). The second is the proportion of contaminant excreted (PCE), which is an inverse of the assimilation efficiency (AE), and represents the amount of a contaminant that is excreted in its original form by an organism back to the environment (Booth, Steenbeek, et al., 2020). Other parameters that were assumed to be negligible include concentration in immigrating biomass, decay rate, and metabolic decay rate. The Ecopath base model does not incorporate immigration and emigration, and Ecospace is not being utilized to introduce a mapped area considering migration in and out of the system external to the defined system of the BSS region represented in the model, so concentration in immigrating biomass is not defined in Ecotracer for this study (Hoover et al., 2021; Sora et al., 2022). The decay rate is a parameter primarily used for radioisotopes, which experience decay in the environment, as the Ecotracer module was originally built for tracing radioisotopes through marine food webs (Booth, Steenbeek, et al., 2020). PFOS is extremely stable in the environment due to the strength of the carbon-fluorine bonds that provide high resistance to hydrolysis, photodegradation, and microbial, so it is assumed to not be changed to other states in the environment and therefore not experience any type of decay akin to that of radioisotopes (Beach et al., 2006; Giesy & Kannan, 2002). Metabolic decay rate is also known as elimination rate, and describes the rate at which a contaminant undergoes biotransformation to another compound by species and is therefore lost to the system (Booth, Steenbeek, et al., 2020). PFOS is not believed to be metabolized or undergo biotransformation to a measurable extent, therefore any PFOS that is not retained by an organism is returned back to the system in its original form and is captured by the proportion of contaminant excreted parameter (Beach et al., 2006; Giesy & Kannan, 2002; Sun et al., 2022).

The PCE values are based on limited published AEs for PFOS in similar species to those in the BSS model. Most studies are lab-based and use aquatic fish species at ambient room temperature; they are not perfectly applicable to the salinity of a marine environment nor the temperature of an Arctic environment. However, it is currently assumed that most fish species experience similar assimilation efficiencies for PFOS (Sun et al., 2022). Thus, the reported assimilation efficiency for rainbow trout (Martin et al., 2003) was used for all fish species based on (Sun et al., 2022). There is a lack of data available for AEs in mammals for most contaminants, including PFOS (Alava et al., 2018; Li et al., 2022). Other model parameterization for methyl mercury and PCBs assume that mammals have the same AE as fish because of the lack of data. Therefore that assumption is also made here for PFOS (Alava et al., 2018; Li et al., 2022). There is one published AE for an invertebrate species, *Daphnia magna*, in the literature, and this was applied to species of similar size and taxonomy (Dai et al., 2013). All other invertebrate species were assigned the same AE as fish, in line with the methods of (Li et al., 2022). The lack of data on AE for various species presents a limitation of the model regarding appropriate representation of the excretion of PFOS back to the environment. However, the current observation of published AEs shows limited variation across species. Therefore, it is anticipated that the approach taken here to assume AE values based on similar species is preferable to not including an AE (Sun et al., 2022). While AE is discussed here, the parameter for direct input into the Ecotracer interface is PCEs, which is the inverse value of AEs. The PCE values for each functional group are outlined in Table 2.2.

The DARs were calculated rather than taken from literature due to the lack of data available from laboratory studies available for representative species in the model. The basic concentration ratio approach used in Ecotracer is adapted to create a formula to determine DARs (Booth, Steenbeek, et al., 2020; Walters & Christensen, 2018). The formula is as follows, with parameters defined in Table 2.1:

$$DAR_i = Z_i \times CR_{io} = \left(\frac{P_i}{B_i}\right) \times \left(\frac{C_i}{C_o}\right) \quad \text{Eq. 6}$$

$$CR_{io} = \left(\frac{C_i}{C_o}\right) \quad \text{Eq. 7}$$

Table 2.1. *Parameters for calculation of Direct Absorption Rates in fish and invertebrates.*

Parameter	Unit	Description	Source
DAR_i	$km^2/t/yr$	Direct absorption rate (or direct uptake rate) of PFOS from seawater	Eq. 6
Z_i	1/yr	Production value for each functional group. Also known as total mortality	BSS Ecopath base model
CR_{io}	km^2/t	Concentration ratio of unit converted sea water concentration to organism concentration	Eq. 7
C_i	g/t	Organism concentration	Literature
C_o	t/km^2	Unit converted seawater concentration given the depth at which sample was taken	Literature

Here, production refers to the amount of tissue elaboration of a population in a given time period, representing a rate of growth for an individual or population (Allen, 1971). The P/B ratio represents how much a group produces relative to its biomass, a function of time with a unit of $year^{-1}$, and related to organism size, metabolic efficiency, and temperature (Allen, 1971; Pauly, 1980). Thus, in a marine ecosystem, the P/B ratio can represent the rate of biochemical processes to convert food to tissues, based on the cell size and water temperature (Allen, 1971; Pauly, 1980). Equation

4 relates these factors incorporated by P/B to the concentration of PFOS in an organism compared to the concentration in its environment, the CR, to determine the rate at which PFOS is taken up from the environment to an organism based on its physiology (size, metabolism) and environment (water temperature), which is a DAR (Christensen et al., 2024; Walters & Christensen, 2018). For species that did not have empirical concentration data available, data for similar species groups (one benthic species used for another benthos) were used for the CR values, and the appropriate Z values from Ecopath were taken to calculate a DAR specific to that species. Using this formula allowed for estimating DARs for many more species than using reported uptake rates from lab studies, as studies reporting species concentrations along with environmental concentrations are much more readily available than toxicokinetic studies. While this approach does allow for advantages in estimating DARs where there are gaps in the toxicokinetic literature and Arctic species literature and may account for important environmental variables such as water temperature and salinity impact on uptake rates, it also has several limitations (Li et al., 2022; Walters & Christensen, 2018). The DAR values calculated using this approach can have greater uncertainty due to the differing methods for sampling or environmental and ecosystem conditions which may change over time, such as water temperature, pH, or predation, which can vary and impact concentration ratios between organisms and the environment that are then integrated into the DAR calculations rather than controlled for as would be done in a laboratory setting (Walters & Christensen, 2018). Nonetheless, this approach does add value by providing a mechanism to calculate DAR values for use in the EwE modelling approach where such data is not yet available and otherwise would not be possible. DARs were only calculated for primary producers and invertebrate functional groups in the model as it is the absorption rate of PFOS directly from water is considered negligible for fish and mammals due to contaminant uptake for those groups being

primarily from the diet (Booth, Steenbeek, et al., 2020; Li et al., 2022). Dietary uptake of PFOS by each functional group is determined by the diet matrix and consumption rates in the Ecopath base model and impacted by the PCE values put into Ecotracer. As such, there is no independent parameter for dietary uptake of PFOS in Ecotracer parameterization like there is for direct uptake from water (Booth, Steenbeek, et al., 2020).

The last parameter for biota in Ecotracer is an initial concentration for each functional group. Initial concentrations can be taken from published literature values for functional groups or from the steady-state model run output. Here, the steady-state output was used as the initial group concentrations, as there was very limited data available for the functional groups that were both collected from species in the Beaufort Sea or the western Arctic and collected within a reasonable time frame that the model represents, ideally 2005-2015. To obtain steady-state concentrations, the environmental parameters provided in Table 2.4 and DAR and PCE parameters in Table 2.2 were put in the Ecotracer module to describe the initial state of the ecosystem. The model was run forward 100 years to reach equilibrium, or steady state, where the movement of PFOS between the functional groups and environment based on the initial environmental state and toxicokinetic parameters is equal. The PFOS concentrations of all functional groups and environment reached a steady state by 75 years. The concentrations of each functional group at steady state are assumed to be representative of empirical group concentrations and were therefore used as the initial concentration parameter for each functional group in Ecotracer.

Table 2.2. *Ecotracer Toxicokinetic Parameters*

Group Name	Initial Concentration (t/t)	Direct Absorption Rate (km^2/t/yr)	Proportion of Contaminant Excreted
Polar Bear	3.01E-11	0	0.279
Beluga	5.82E-10	0	0.279
Bowhead	2.00E-09	0	0.279
Ringed Seal	2.74E-11	0	0.279
Bearded Seal	5.20E-11	0	0.279
Birds	5.36E-13	0	0.279
Char & Dolly Varden	8.49E-12	0	0.279
Cisco & Whitefish	3.10E-11	0	0.279
Salmonids	2.58E-11	0	0.279
Herring & Smelt	3.99E-11	0	0.279
Arctic & Polar Cod	9.10E-11	0	0.279
Capelin	1.14E-11	0	0.279
Flounder & Benthic Cod	1.26E-11	0	0.279
Small Benthic Marine Fish	1.40E-11	0	0.279
Other Fish	2.09E-11	0	0.279
Arthropods	3.41E-11	2.34E-06	0.25
Bivalves	3.28E-12	1.01E-07	0.25
Echinoderms	1.32E-11	9.21E-08	0.25
Mollusks	6.12E-12	1.43E-07	0.25
Worms	8.31E-12	1.60E-07	0.25
Other Benthos	6.34E-12	1.26E-07	0.25

Jellyfishes	1.95E-11	1.10E-05	0.25
Macro-Zooplankton	9.04E-12	4.38E-06	0.25
Medium Copepods	6.98E-11	1.16E-04	0.17
Large Copepods	1.87E-10	3.24E-05	0.25
Other Meso-Zooplankton	6.58E-11	1.47E-04	0.17
Micro-Zooplankton	2.16E-11	7.74E-05	0.17
Producers > 5 um	1.11E-12	2.90E-06	0
Producers < 5 um	2.91E-12	5.75E-06	0
Ice Algae	9.61E-13	2.49E-06	0
Benthic Plants	6.71E-13	6.86E-07	0
Pelagic Detritus	3.16E-12	0	0
Benthic Detritus	1.74E-14	0	0

Ecotracer Environmental Parameters

For environmental conditions, the initial concentration of PFOS in BSS seawater was taken from published field data, using an average of 28 samples taken from six locations within the Beaufort Sea in 2012 (Yeung et al., 2017). There is currently very limited data available for the amount of PFOS entering the Beaufort Sea each year. As a result, a method used by (Li et al., 2022) for parameterizing the same BSS model for mercury was adopted to set the initial environmental conditions to best reflect the PFOS in the Beaufort. The initial seawater concentration and base inflow rate to the same value as observed 2012 concentrations, and the base volume exchange loss to 1, to calibrate the transport of PFOS in and out of the BSS based on the observed seawater concentration rather than an estimate based on extremely limited data. The following equation was used to calculate the initial seawater concentration, with parameters defined in Table 2.3:

$$C_i = C_a = \frac{C_v}{0.2} \qquad \text{Eq. 8}$$

Table 2.3. *Parameters for calculation of initial seawater concentration.*

Parameter	Unit	Description	Source
C_i	t/km^2	Initial environmental concentration of BSS seawater	Eq. 8
C_a	t/km^2	Unit converted seawater concentration assuming 200m depth of the Beaufort Sea (0.2 km)	Eq. 8
C_v	t/km^3	Average observed concentration of PFOS in Beaufort Sea	Literature (Yeung et al., 2017)

It is possible that transport of PFOS to the BSS from the Mackenzie River is underestimated using this method. However, it is not possible to develop a more specific method because of the lack of monitoring data available for PFOS input via the Mackenzie River. The environmental parameters inputted to the Ecotracer interface are provided below in Table 2.4.

Table 2.4. *Ecotracer Environmental Parameters in baseline year, 2010.*

Parameter	Unit	Value
Initial Concentration	(t/ km^2)	2.80E-06
Base Inflow Rate	(km^2/t/yr)	2.80E-06
Decay Rate	(/yr)	0
Base Volume Exchange Loss	(/yr)	1

Model Bias

Model performance was evaluated by comparing the modelled concentrations at steady state with observed concentrations for respective functional groups to assess how well the model reproduces the natural ecosystem. A ratio between the modelled (M) and observed (O) PFOS concentrations and a normalized mean bias was determined using equations 9 and 10, following the approach (Li et al., 2022).

$$\textit{M/O Ratio} = \frac{M}{O} \qquad \text{Eq. 9}$$

$$\textit{Normalized Mean Bias} = \frac{\sum_1^n (M-O)}{\sum_1^n O} \qquad \text{Eq. 10}$$

Published data, included in Table 2.5, was used for the observed values, with a preference for samples collected from the Beaufort Sea or the western Arctic and between the years 2000-2020, which is +/- 10 years from the model baseline year of 2010. Due to a lack of data availability for PFOS, some observed concentrations were used for species in other Arctic regions or from earlier than the year 2000 to still allow for a comparison between the modelled and observed data, even if such values were not perfectly matched for date, species, and location.

Climate Change Scenarios

The climate change scenarios were run in Ecosim for a time period of 2010-2100. Climate change scenarios were developed using data from the Earth System Model 4.1 (ESM4) developed by the NOAA-Geophysical Fluid Dynamics Laboratory (GFDL) for the IPCC AR6 Shared Socioeconomic Pathways (SSP) 1-2.6 and 5-8.5 (Dunne et al., 2020; Lee et al., 2021). SSP1-2.6 describes a scenario where global policy action results in significant greenhouse gas emission reduction keeping global warming levels below 2°C by the end of the century, and is used for the 'low emissions' climate change scenario (Chen et al., 2021). SSP5-8.5 describes what is colloquially called a worst-case scenario, where limited global action is taken to curb greenhouse gas emissions resulting in global warming levels above 4°C by the end of the century, and is used for the 'high emissions' climate change scenario (Chen et al., 2021). These two scenarios are used in climate change modelling frequently to represent the bookends of 'ideal' climate change mitigation policies and no climate change mitigation policies (Chen et al., 2021; Lee et al., 2021). Ideal is presented here in quotation marks as the previous 'ideal' global warming scenario was limiting warming to 1.5°C by the end of century as outlined in the Paris Agreement, however this can be considered unrealistic given the pledges made by most countries, which if followed, will not achieve 1.5°C of warming by the end of century (Chen et al., 2021). The current trajectory as of 2023, based on nationally determined contributions under the Paris Agreement, is between 2.5-2.9 °C of warming by the end of the century. Thus, the low emissions scenario represented by SSP1-2.6 requires additional carbon emissions reductions to achieve (United Nations Environment Programme, 2023). Nevertheless, SSP1-2.6 is used for the low-emissions scenario, which would keep warming below 2°C by the end of the century. In addition to the two climate change scenarios, a baseline reference scenario was run to serve as a reference without climate change forcing. This scenario represents the average state at baseline, 2010, based on the 20-year average data from 2000-2020.

There is no downscaled data available for the model area. Therefore, the extracted data from the GFDL-ESM4 model was for a larger area encompassing most of the Beaufort Sea, including the whole BSS model area. This area is shown in a rough schematic in Figure 2.3, with the red lines delineating the area covered by the coordinates: 68.5, -148.5; 68.5, -110.5; 79.5, -148.5; 79.5, -

110.5. In addition to the larger area encompassing the BSS model, this geographic area was chosen to capture the majority of the ISR, as well as a large portion of the beluga range within the Beaufort Sea, given the migratory nature of this species and its importance as both a high-trophic level mammal and important country food in the Inuvialuit diet (Loseto et al., 2018).

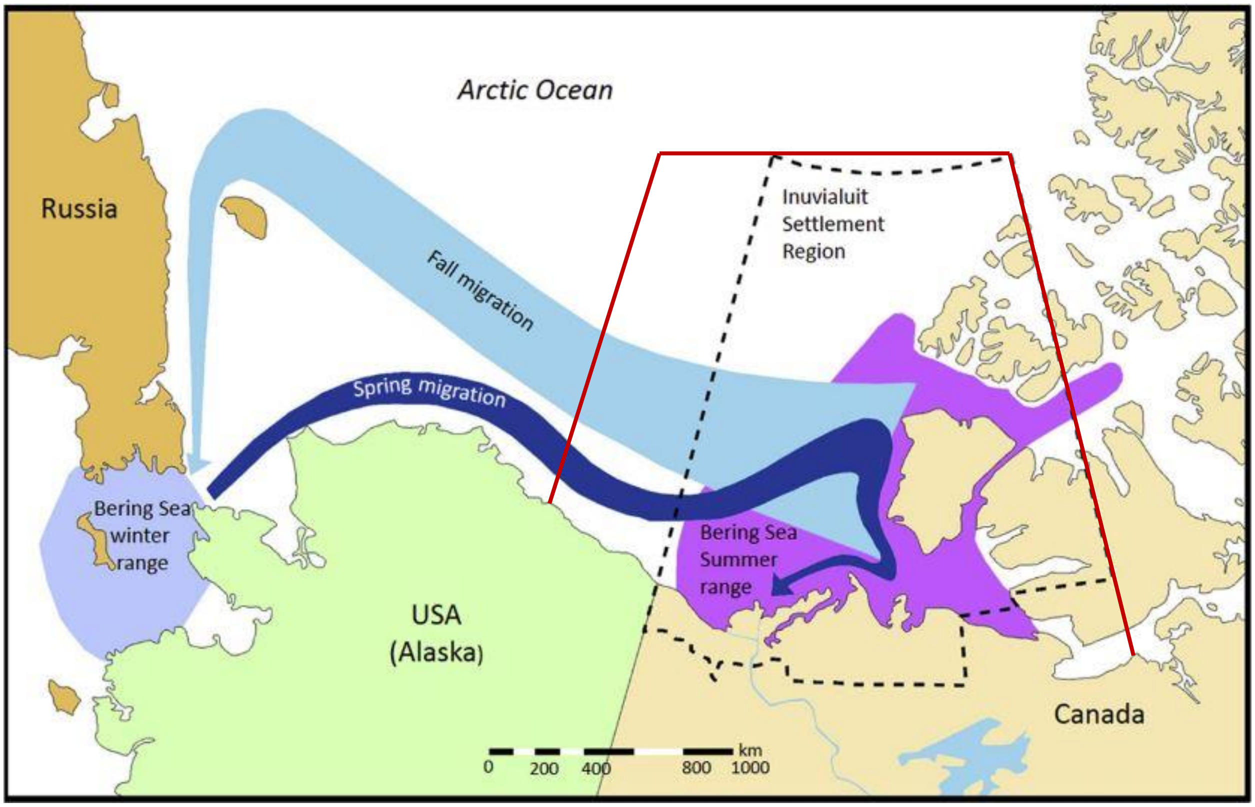

Figure 2.3. *Visualization of the geographical area (red line) for which sea surface temperature and sea ice cover data were extracted from the GFDL-ESM4 model for EwE climate change scenarios, in reference to beluga home range and migration patterns*. Adapted from Loseto et al., (2018).

GFDL-ESM4 predictions were extracted for this area for each SSP1-2.6 and SSP5-8.5, for sea surface temperature (SST) and sea ice cover (SIC), from the years 2010-2100 (Dunne et al., 2020). These projected data were corrected using observational data, based on a 20-year average of observed data from 2000-2020 for SST and SIC values in the region (British Atmospheric Data Centre, 2010). The data was then processed to establish a change index from the baseline year of

2010, rescaled to 1, and then implemented as a multiplier via various functions available in Ecosim. The index of change from the baseline year for SST and SIC under each climate change scenario is shown in Figures 2.4 and 2.5.

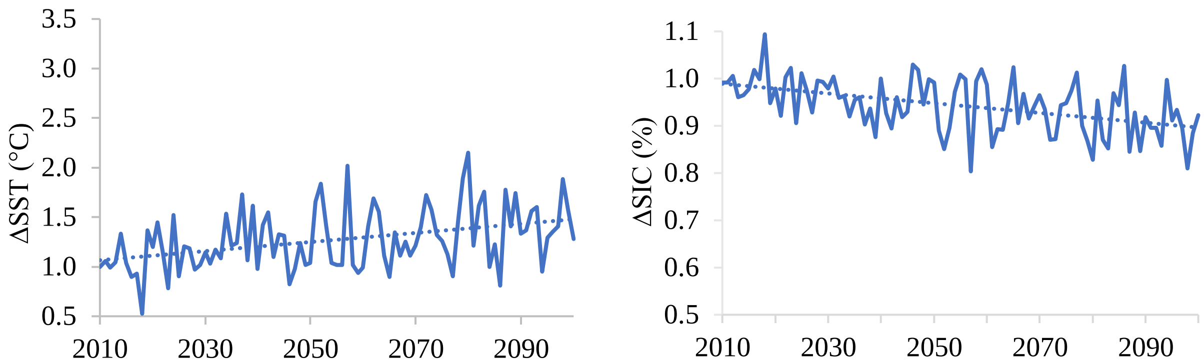

Figure 2.4. *Relative change in sea surface temperature (°C) and sea ice cover (% total ice cover) under SSP1-2.6 from 2010-2100.*

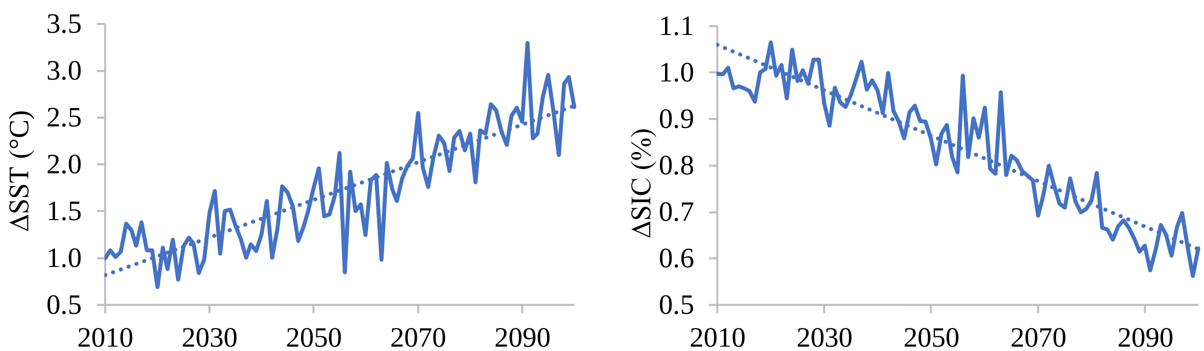

Figure 2.5. *Relative change in sea surface temperature (°C) and sea ice cover (% total ice cover) under SSP5-8.5 from 2010-2100.*

Implementing Ecosim Scenarios

The impacts of SST and SIC as climate drivers were implemented in the model using three different mechanisms: forcing functions, environmental responses, and mediation functions. The implementation approach draws on several uses of each of these mechanisms from previous studies

(Ainsworth et al., 2011; Alava et al., 2018; E. J. Gillies et al., 2024; Guénette et al., 2014; Hoover et al., 2021; Li et al., 2022; Sora et al., 2022). A conceptual framework of the Ecosim scenarios for SST and SIC that are implemented for each climate change scenario is presented below in Figure 2.6, adapted from Gillies, (2022).

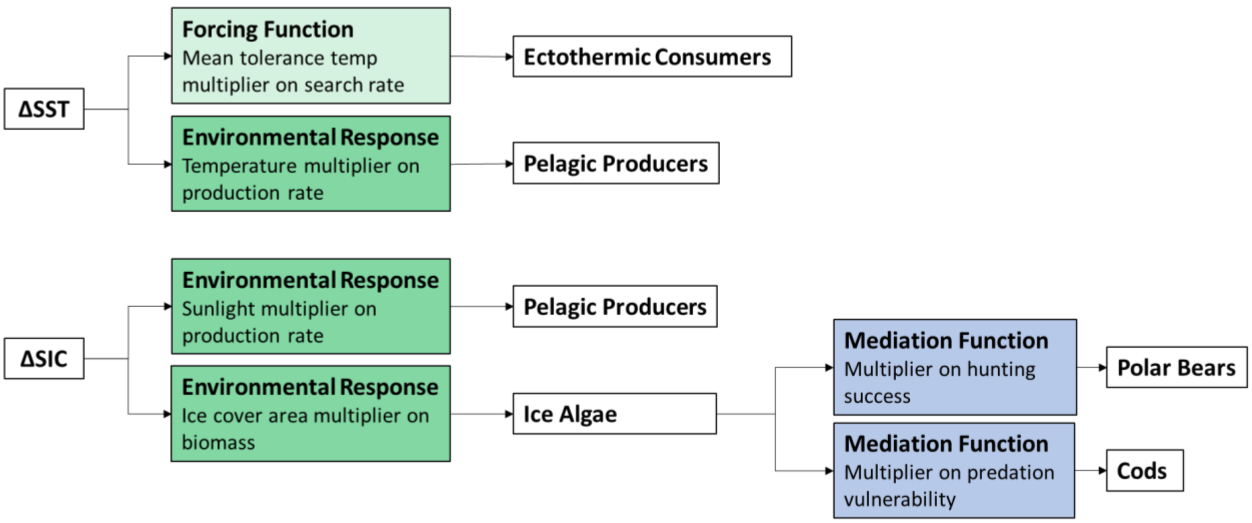

Figure 2.6. *Schematic of the Ecosim scenario approaches utilized to implement the climate change scenarios in the EwE model*. Adapted from Gillies, (2022).

To implement the effects of SST changes under each scenario in the model, two approaches were used based on the relative change time series shown in Figures 2.4 and 2.5. The first was the use of forcing functions to directly force consumer group biomass, and the second was the use of environmental response functions to force producer biomass. To force consumer groups, the time series of relative change from 2010-2100 for each climate change scenario was used in conjunction with mean tolerance temperatures (MTT) for functional groups of ectothermic species to create unique forcing functions for each group (Alava et al., 2018). The MTT for each ectothermic consumer functional group was used to create a normal distribution curve, which was integrated with the relative change in SST to create a forcing function that acts as a multiplier on the search rate, or predator success (Ainsworth et al., 2011; Alava et al., 2018; Guénette et al., 2014). This approach assumes that environmental changes alter a consumer's efficiency as a predator, where they either become more or less efficient depending on the environmental changes experienced and the species MTT, resulting in increased or decreased growth and ultimately altering the consumption to biomass ratio (Q/B) for a functional group (Ainsworth et al., 2011; Alava et al., 2018; Guénette et al., 2014). Thus, the species-specific forcing functions were applied to functional groups in the model for ectothermic consumers, fish and invertebrates. A time series of relative SST from baseline was applied to mammals and birds, assuming that the direct impact of SST on consumption to biomass is negligible for these species given they are endothermic and therefore do not have MTTs (Ainsworth et al., 2011; Alava et al., 2018). Figure 2.7 shows an example of the forcing function for the Arctic char and dolly varden functional group under both climate change scenarios, where the values are a multiplier on biomass each year as a function of the relative change in SST and species' MTT. MTT values can be found in Appendix B, Table B-1.

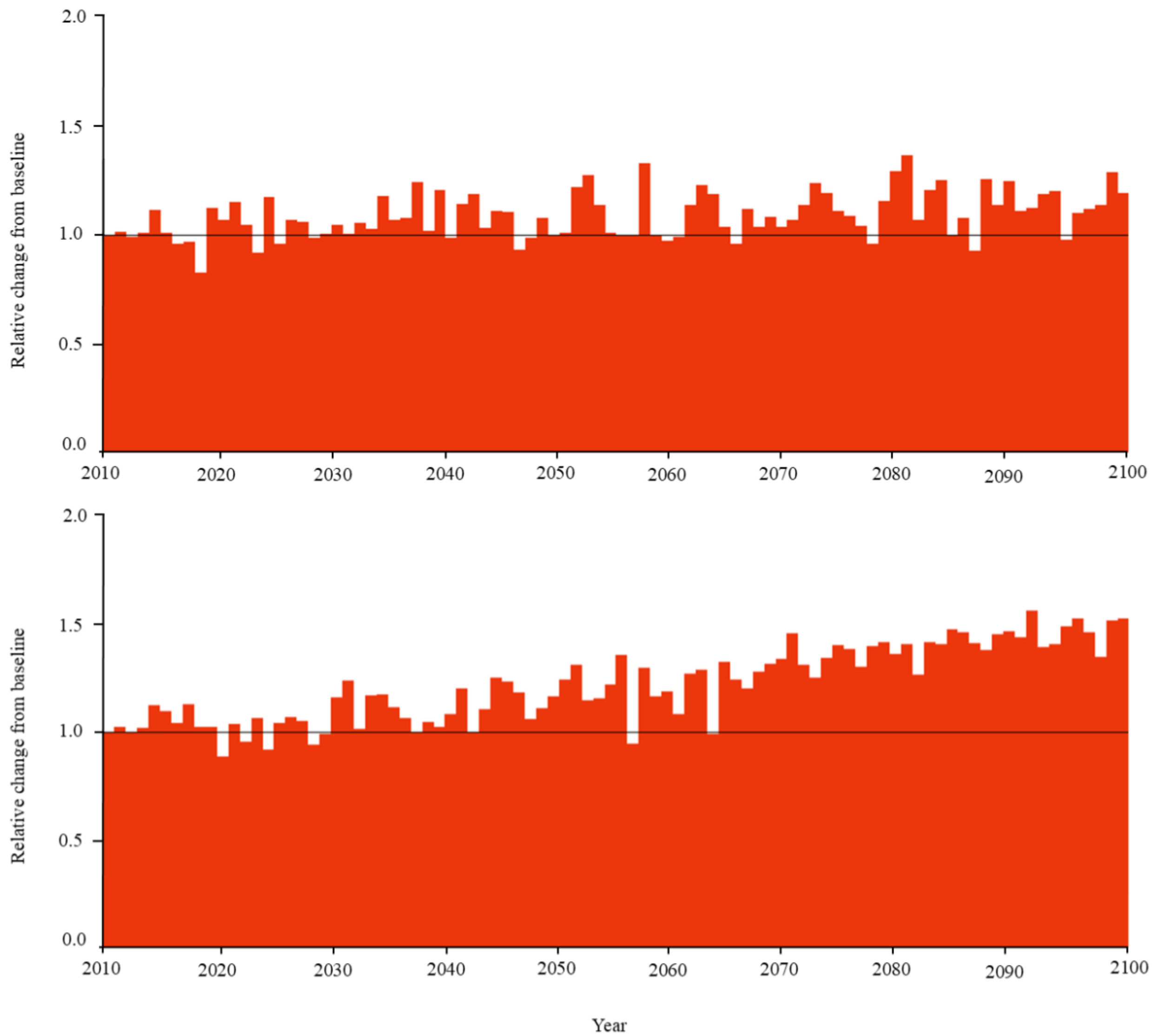

Figure 2.7. *Forcing functions to implement SST changes under SSP1-2.6 (upper panel) and SSP5-8.5 (lower panel) for the Arctic char and dolly varden functional group.*

To implement the effects of SST change on producer groups, the time series of relative change in SST was implemented in conjunction with environmental response functions to indirectly force biomass of the pelagic producers' functional group. This approach uses defined environmental response functions based on a supposed optimal range of environmental conditions, outside of which production efficiency drops. Such functions are developed for primary producers, therefore modify the efficiency of primary production based on changes in SST. The environmental response function implemented here was developed (Gillies et al., 2024) based on (Steiner et al., 2019) for small and large pelagic primary producers in the BSS, and is shown in Figure 2.8. The fundamental relationship assumes that primary production increases with SST increases, where primary production increases are proportional to SST increases between relative SST of 0.5 (50% of the 2010 annual average temperature) and 1.5 (150% of the 2010 annual average temperature). These thresholds are based on primary production research within the Arctic, where there have been observed linear increases in primary production associated with SST increases to about 5°C (Gillies et al., 2024; Steiner et al., 2019). However, there is uncertainty in the observed relationship of primary production to SST and optimal zones of primary production. Hence, an entirely linear relationship between primary production and SST increases is not justifiable (Gillies et al., 2024; Steiner et al., 2019). Based on the best available research, the thresholds set for this relationship are relative changes between 0.5-1.5 in SST from baseline, within which there is a linear relationship between pelagic primary producers and SST. Together, the forcing functions and environmental response functions were implemented to simulate the effects of predicted changes in SST on both consumer and producer groups in the BSS ecosystem under the two climate change scenarios.

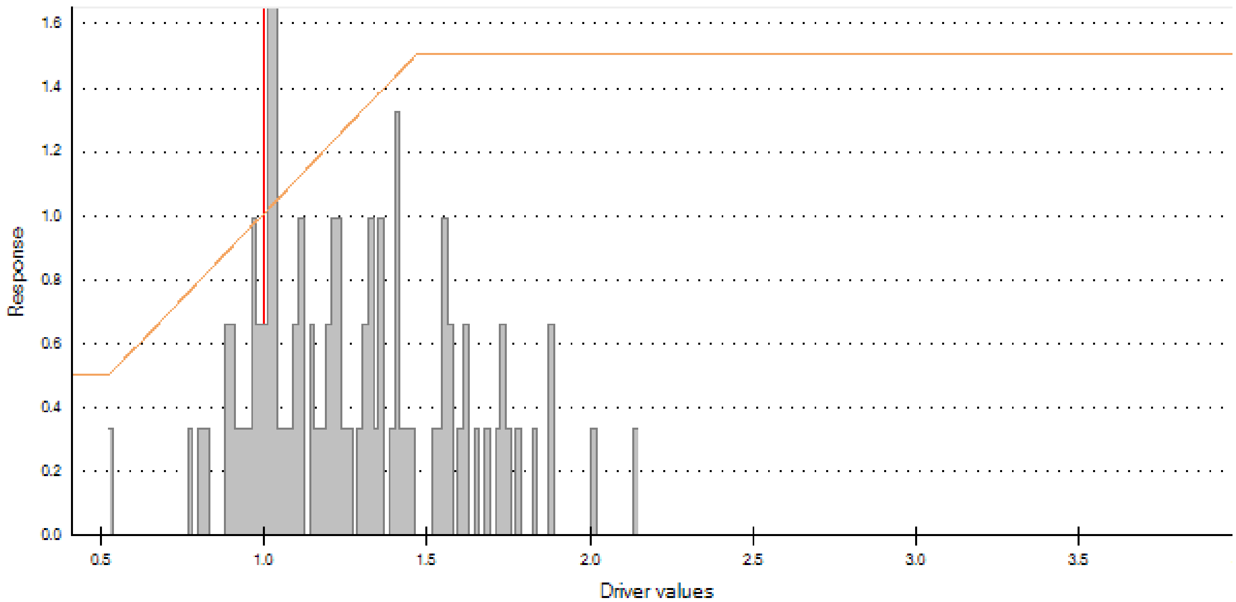

Figure 2.8. *Environmental response function of pelagic producers to sea surface temperature, where the response is a multiplier on the baseline level of production and the driver value is the relative sea surface temperature compared to baseline.*

To implement the effects of SIC changes in the model, environmental response functions were used to define the impacts of SIC change on producers and mediation functions were used to simulate the impacts on select consumer groups. The environmental preference functions were also developed by (Gillies et al., 2024) using the work (Steiner et al., 2019), following similar assumptions as used for SST. Two environmental preference functions were defined for SIC impacts on producers, one for impacts to pelagic primary producers and one for impacts to ice algae, given the different ways each is impacted by SIC change. The environmental response function defining the relationship between SIC and pelagic primary producers was based on sunlight, where less SIC leads to more sunlight reaching the water column, therefore increasing primary production and vice versa. This function is shown in Figure 2.9. Like the SST environmental response function, the thresholds of 0.5 and 1.5 represent the change from baseline, wherein there is a linear relationship between the change in driver and change in production. Here, the relationship assumes that as SIC increases from 0.5, or a 50% reduction in SIC from baseline, there is a proportional decrease in production. After an increase in SIC by a factor of 1.5, so 50% more ice cover compared to baseline, the relationship is no longer linear and there is no defined change in primary production. Similar to SST, this is due to uncertainty in the relationship between SIC and pelagic primary production, where sunlight is an important variable but not the only one mediation production, and most relationships like these have an optimal zone based on organism tolerance windows even if they are not well characterized by literature therefore are usually not entirely linear.

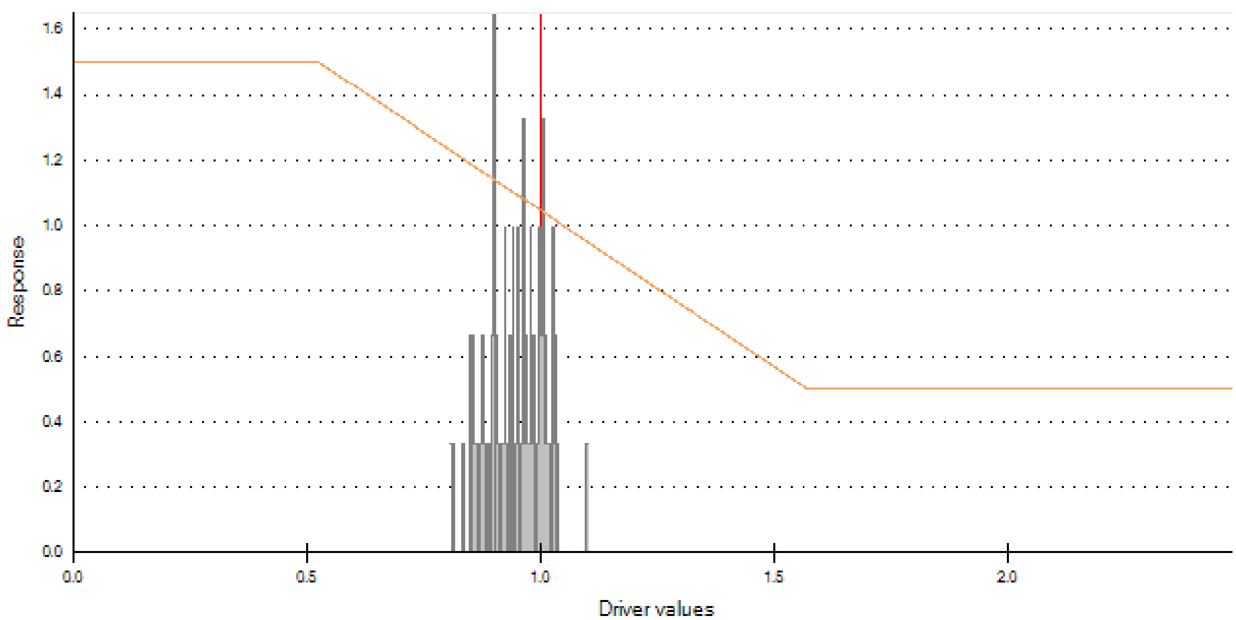

Figure 2.9. *Environmental response function of pelagic producers to sea ice cover, where the response is a multiplier on the baseline level of production and the driver value is the relative sea ice cover compared to baseline.*

The last environmental response function is for the ice algae response to change in SIC, also defined by (Gillies et al., 2024) based on (Steiner et al., 2019). The relationship here is between ice algae biomass and SIC, where ice algae are producers found within sea ice as a main part of their life cycle. Therefore, the assumption is that ice algae production increases proportionally with increases in SIC. The lower threshold is set at 0 rather than 0.5, as it assumes that with no sea ice cover, there is no ice algae biomass, which is due to the heavy reliance of ice algae on SIC. Until a relative increase in SIC of 1.5, the relationship between ice algae production and SIC is assumed to be linear. After 1.5, the relationship plateaus and is no longer linear. This is shown in Figure 2.10. The assumptions here mirror those in the functions for pelagic producers to SST and SIC, where there is an assumed optimal range for this relationship rather than one that is entirely linear, and linear increases are not consistent with the observed biomass trends of ice algae.

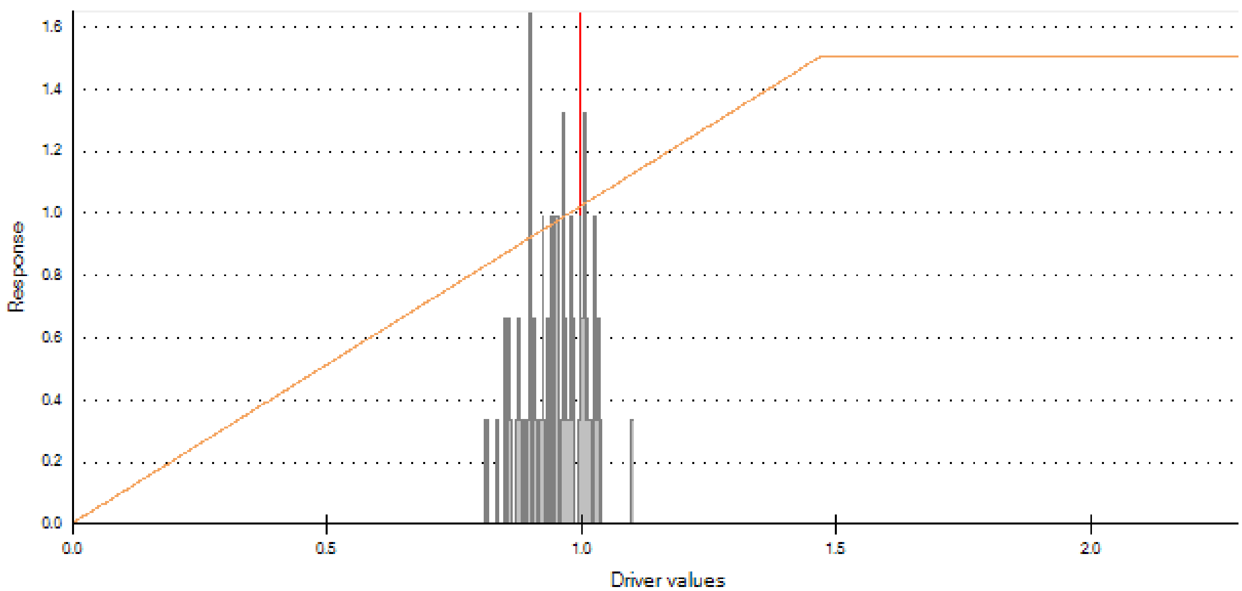

Figure 2.10. *Environmental response function of ice algae to sea ice cover, where the response is a multiplier on the baseline level of production and the driver value is the relative sea ice cover compared to baseline.*

Mediation functions were used to implement the effects of SIC change on two consumer groups that have uniquely defined relationships to sea ice extent: polar bears and cods. A mediation function uses one group as a 'mediator' for the impacts to another group. For these functions, ice algae were used as the mediating group, assuming a linear relationship between ice algae biomass and SIC (Hoover et al., 2021). So, if ice algae biomass decreases by 30%, it is assumed that this has been caused by a proportional decrease in SIC of 30%. These mediation functions were developed by (Hoover et al., 2021) based on the relationship of each group to sea ice. Polar bears use sea ice as an area for hunting, so less sea ice means a smaller area to hunt and less efficient predation. This relationship is illustrated in Figure 2.11.

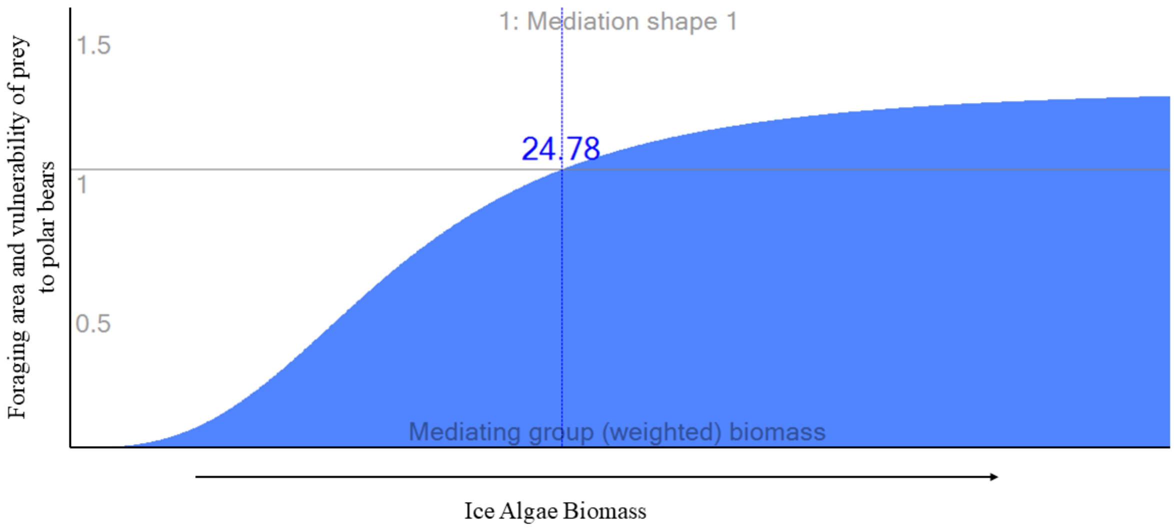

Figure 2.11. *Mediation function defining response of polar bear predation success (y-axis) to ice algae biomass (x-axis). The intersecting grey lines indicate the baseline situation defined by the Ecopath base model.*

For cods, the crevasses in the sea ice underwater are used to hide from predators like seals, so more ice cover means more hiding places, and a decreased vulnerability to predation (Hoover et al., 2021). This relationship is shown in Figure 2.12. In both cases, these mediation functions are very similar to the environmental response functions. However, the driver is the biomass of another functional group – ice algae – rather than an environmental forcing function.

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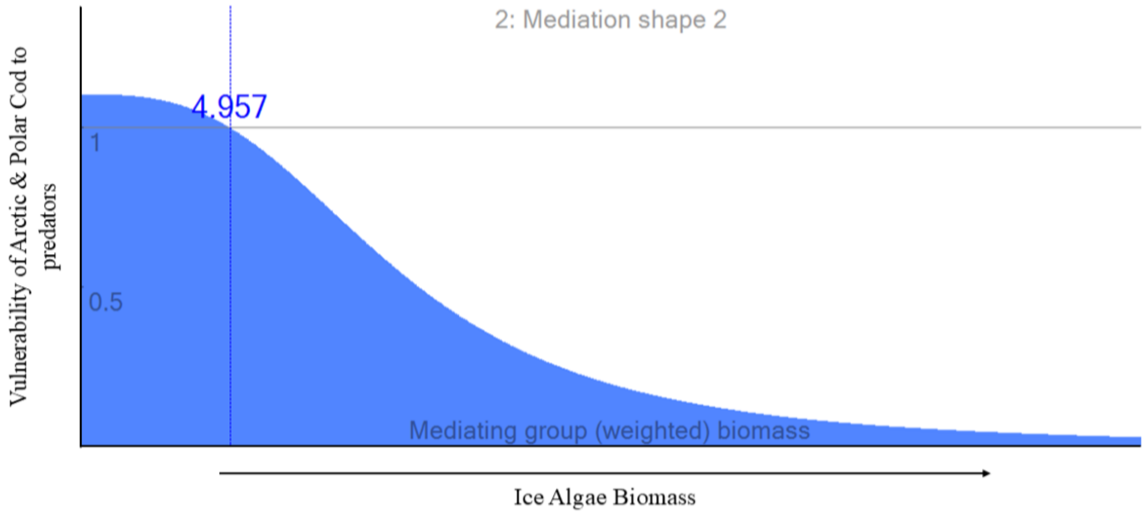


Figure 2.12. *Mediation function defining response of Arctic and polar cod vulnerability (y-axis) to ice algae biomass (x-axis). The intersecting grey lines indicate the baseline situation defined by the Ecopath base model.*

Model Output Analysis

Trophic magnification factors (TMF) were calculated for the simulations under each climate change scenario, following the method of Alava et al. (2018). TMFs are commonly used to quantify the biomagnification of a contaminant in a food web (Alava et al., 2018). The TMFs were calculated as the antilog of the regression slope, per equation 11:

$$\log [\text{PFOS}] = a + b\text{TL} \qquad \text{Eq. 11}$$

A TMF greater than 1 ($b > 0$) signifies that the contaminant biomagnifies in the food web, a TMF less than 1 indicates trophic dilution is taking place, and a TMF of 0 indicates no change in concentrations across trophic levels (Alava et al., 2018).

2.3 Results and Discussion

Evaluation of Model Performance

The model output produces average concentrations for each functional group population. The simulated concentrations are comparable to the observed concentrations of each group with available, shown in Table 2.5. As this is the first attempt at using the EwE with Ecotracer modelling approach for PFOS, there are no previous studies to compare model bias values with directly. However, previous studies simulating methylmercury concentrations have reported what is considered to be poor model bias from difficulties parameterizing (M/O = 0.016 – 0.056) and much-improved model bias (M/O = 0.088 – 0.86). Here, the range of M/O = 0.013 – 1.74. There is a relatively large variation here, for several reasons.

Table 2.5. *Model bias values for species with available data.*

Group	Modeled ng/g	Observed ng/g (Mean)	Model Bias = M/O	Normalized mean bias =Σ(M-O)/ΣO	Observed ng/g (Mean or range)	Sample size
Polar Bear*	17.7	128	0.138	N/A	128[a]	19
Beluga†	17.1	11.3	1.513	0.51	9.86[b]	9
					12.7[b]	9
Ringed seal	1.01	5.55	0.182	-0.82	2.5-8.6[c]	5
Bearded seal	2.26	1.30	1.738	N/A	1.30[c]	1
Cods	0.11	0.50	0.228	-0.77	0.3-0.7[c]	5
Cisco & Whitefish	0.06	0.32	0.187	N/A	0.32 (<0.06 - 0.59)[d]	9
Large copepods	0.02	0.39	0.059	N/A	0.39 (<0.06-1.51)[d]	5
Macro-zooplankton	0.04	2.66	0.013	-0.97	2.66 (2.12, 3.19)[d]	2
		0.10	0.350		<0.2-0.2[c]	3

Note. All samples collected between 1999-2006, from the Beaufort Sea with exceptions from Greenland* and Hudson Bay[ᶧ]. Observed values from: (Greaves et al., 2012)[a]; (Kelly et al., 2009)[b]; (Powley et al., 2008)[c]; (Tomy et al., 2009)[d].

The model overpredicts PFOS concentrations in beluga compared to what has been observed (M/O = 1.51, normalized mean bias=0.51). This is consistent with the beluga concentrations predicted by the BSS model parameterized for methylmercury (M/O = 1.74, normalized mean bias=0.74) (Li et al., 2022). In the case of the mercury overprediction, the model predicted mercury concentrations in the main prey of beluga (cods, whitefish, and cisco) with high accuracy compared to what is observed, so it was anticipated that the model overestimated mercy intake from food, rather than a systemic overestimation within the model as shown in Figure 2.13 (Li et al., 2022). This is attributed to the foraging nature of beluga, which only feed within the Beaufort Sea during summers, and feed during winters in the Bering strait, which is not accounted for in the base Ecopath model (Hoover et al., 2021; Li et al., 2022; Loseto et al., 2018). Mercury concentrations are historically higher in the Beaufort than the Bering, so it is expected that beluga food sources for half of the year are in areas with lower mercury concentrations, which is not accounted for in the model, leading to the model simulating higher concentrations than observed (Li et al., 2022). This is possibly also happening with respect to PFOS, as the simulated PFOS concentrations in cods, whitefish, and cisco are lower than what is observed (Table 2.5), suggesting that the discrepancy between modelled and observed beluga concentrations may also be due to beluga foraging behaviour. However, there is weaker evidence than exists for mercury regarding differences in PFOS concentrations between the Beaufort Sea and the Bering Strait. This is in part due to a lack of spatio-temporal PFOS data that would allow for comparisons between regions within the same time frame. From the data available, PFOS seawater concentrations sampled in 2005 in the Bering Strait and the Chukchi Sea are slightly higher than the Beaufort Sea (14-27 pg/L vs 9.1-11 pg/L). However, it is thought that sediment or in-vivo biotransformation of PFOS precursors may play an important role in PFOS uptake by biota (Armitage et al., 2009; Benskin et al., 2012; Lin et al., 2020). There are limited studies available. However, one study with samples from 2014-2016 reports slightly higher PFOS concentrations in Bering Sea sediments relative to parts of the Chukchi Sea, indicating a potential trend of higher PFOS concentrations in sediment compared to areas closer to the Beaufort Sea (Lin et al., 2020) and another study with samples from 2018-2021 reports concentrations of 1.1 and 2.8 ng/g ww in Beaufort Sea saffron cod and whitefish, respectively, and PFOS was not detected in the same species sampled from the Chukchi Sea, also suggestive of a trend toward higher PFOS levels meaningful for update in biota in the more western parts of the beluga range compared to the Eastern (Fraley et al., 2023). However, given the lack of data available for PFOS concentrations in seawater, surface sediments, fish, and beluga, further information would provide better insight as to the reason for the discrepancy between the modelled concentrations and observed concentrations in beluga. There was also a challenge with the empirical data for beluga, as the samples in the western Arctic were only for liver tissue. PFOS accumulates in the liver; therefore, concentrations are much higher and not representative of the whole-body burden and therefore are not appropriately comparable to the model output. Other studies have shown that PFOS concentration values tend to be similar between Hudson Bay and Beaufort Sea beluga (Kelly et al., 2009). As such, the empirical data used here is from Hudson Bay beluga.

The model bias for ringed seals is very high (M/O = 1.74). This is in part due to a lack of data for ringed seals, as there is only one single sample for the whole Arctic. As a result, the PFOS concentration from the one sample could either be higher or lower than an average would be, which could cause a large model bias. In Table 2.5, the observed concentration is 1.3 ng/g, and the modelled is 2.26 ng/g, so while the M/O indicates that the modelled concentration is almost double the observed, the difference is limited to approximately 1 ng/g. Due to the limited sample size for estimating the range of ringed seal PFOS concentrations, the model bias here should be interpreted with caution.

There is also an area of uncertainty with the model bias for polar bear concentrations. The value (M/O = 0.138) suggests a reasonable comparison, however, comparing the modelled vs observed concentrations themselves, the observed value is 10-fold higher than the modelled value. It is not clear why this is; however, there are several potential explanations. The observed samples were from Greenland, where biota typically have much higher PFOS concentrations relative to the Canadian Arctic (Greaves et al., 2012; Smithwick et al., 2006). However, this is usually a difference of only 2-3 times, which does not account for the entirety of the 10-fold difference (Greaves et al., 2012; Smithwick et al., 2006). It is possible that there is some form of precursor biotransformation taking place, which has previously been suggested as a tentative theory to explain the very high PFOS concentrations observed in polar bears relative to other marine mammals (Greaves et al., 2012). It is possible that this model bias analysis offers some support for this theory, as the model does not replicate these high, observed PFOS polar bear concentrations. Biotransformation of PFOS was not included in the Ecotracer parameterization for any species and the BSS food web is well characterized by the base Ecopath model, therefore much higher observed concentrations in polar bears than those modelled may indicate a species-specific toxicokinetic driver, such as a biotransformation pathway. However, this cannot be determined from this modelling alone, and the reason for a large discrepancy between the modelled and observed PFOS concentrations in polar bears remains unclear.

The modelled fish concentrations are also underpredicted compared to the observed values (M/O = 0.228 & 0.187), which is consistent with the BSS model and was also observed with mercury concentration simulations (Li et al., 2022). This was attributed to potentially underestimating seawater methyl mercury concentrations or uptake in lower trophic levels, which may also be occurring with PFOS (Li et al., 2022). Given the lack of data for DARs and associated uncertainty, it is possible that underestimations in lower trophic level species' uptake of PFOS from seawater can explain the underestimation in fish. It is also possible that the lack of data regarding PFOS uptake from sediment contributes to this underestimation as well. However, the lack of data regarding empirical PFOS concentrations also limits the model bias calculation so it is possible that the small sample size is also a contributing factor.

The model also underpredicts the PFOS concentrations in invertebrates (M/O = 0.059, 0.013, 0.350), consistent with the performance for mercury (Li et al., 2022). The modelled concentrations are less consistent with observed values than fish. However, the sample size is also very limited, and there is significant variation in the observed concentrations. The concentrations in large copepods are not detected (<0.06) to 1.51 ng/g, and similarly for macrozooplankton are not detected (<0.2) to 3.19 ng/g, both of which are large ranges. It is possible that the sampling locations played a role in the large difference in observed macrozooplankton PFOS concentrations. Two samples were taken for *Calanus hyperboreus* and *Themisto libellula* very close to shore, right at the mouth of the Mackenzie Estuary, and show much higher concentrations (2.19 – 3.19 ng/g ww), compared to the other samples for *Calanus hyperboreus, Themisto libellula,* and *Chaetognatha* taken further out in the Beaufort Sea just one year earlier, ranging from <0.2 – 0.2 ng/g ww (Powley et al., 2008; Tomy et al., 2009). It is possible that there is PFOS transportation to the Beaufort Sea via the Mackenzie River leading to higher concentrations within the Mackenzie

Estuary. PFOS transport via the Mackenzie was not captured in the Ecotracer module due to a lack of data regarding potential sources of PFOS from the Mackenzie River, which may be causing an underestimation of PFOS concentrations in seawater, which may also explain the discrepancy in modelled and observed PFOS concentrations in invertebrates.

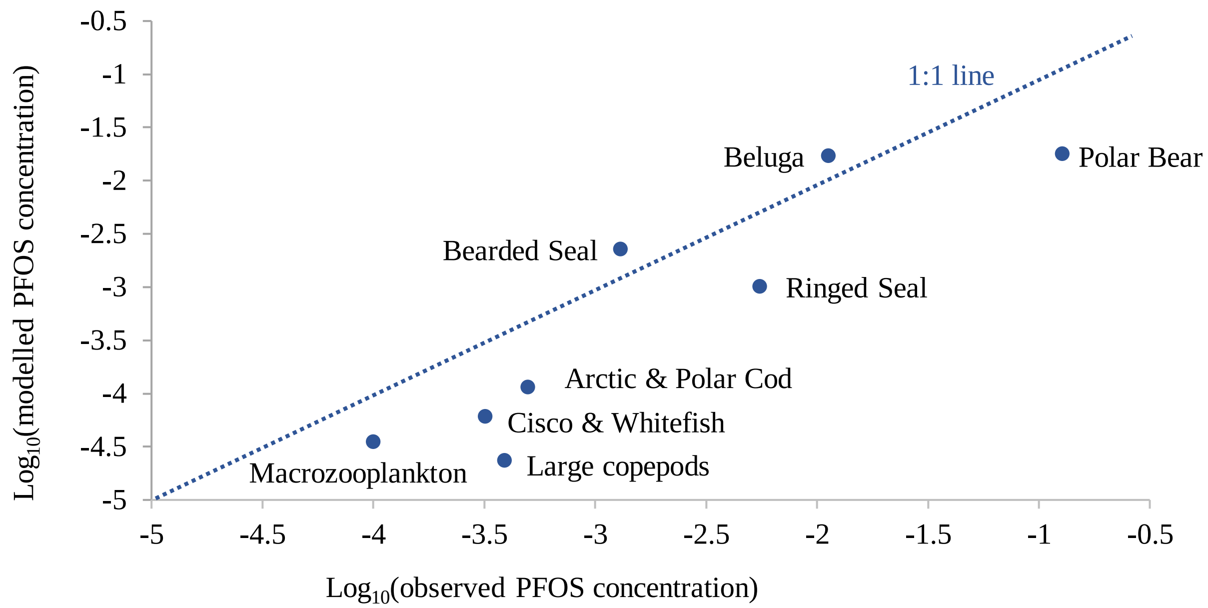


Figure 2.13. *Comparison between log transformed modeled and empirical PFOS concentrations (mg/kg wet weight) for species with available data in the Beaufort Sea shelf food web.*

While the model performance is comparable to observed concentrations based on the available data, and there are justifications for some of the larger model biases, there are still many avenues for improving the model performance. This is the first EwE model to simulate PFOS concentrations, in part because of the limited toxicokinetic data available to be used in parameterizing the model. Further details were provided in the Ecotracer parameterizing section as to the data gaps and assumptions made to mechanistically parameterize the direct absorption rates for many species. However, significant limitations of this model are the lack of toxicokinetic data available with which to characterize possible biotransformation of PFOS within biota, the assumption that seawater concentrations of PFOS remain constant and therefore do not account for differing PFOS sources from atmospheric deposition, ocean currents, and the Mackenzie River, and lastly the assumption that direct uptake of PFOS is only from seawater, without factoring in uptake from surface sediments as well. With more data emerging on environmental PFOS transport and concentrations as well as better understanding of PFOS toxicokinetics representing more species and impacts of colder seawater temperatures in the Arctic, more detailed Ecotracer parameterization will likely facilitate improved performance of future models.

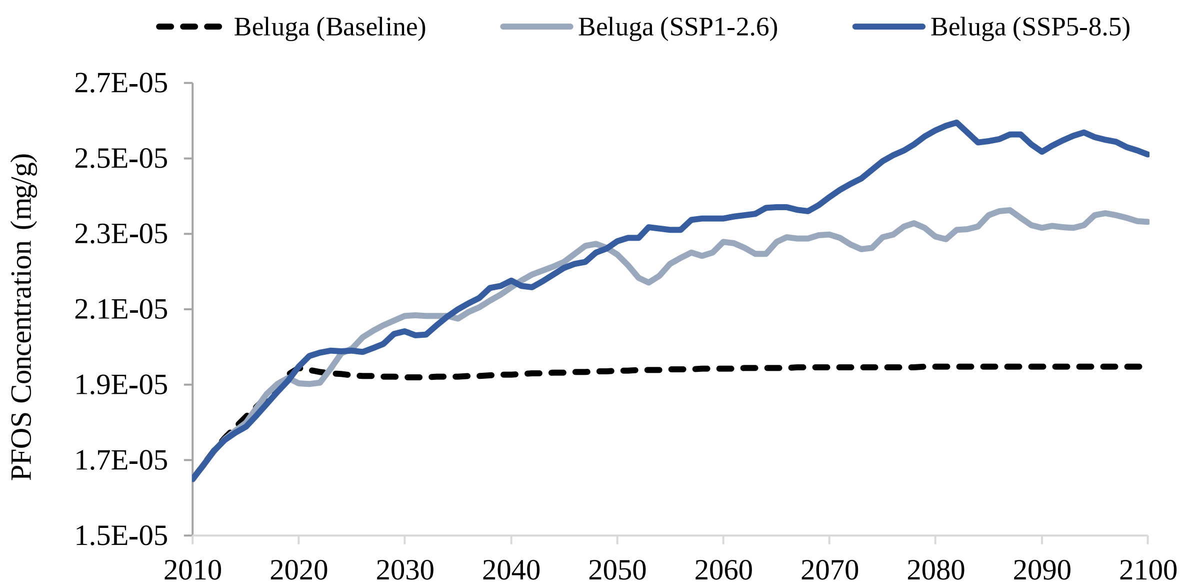
Projected PFOS Concentrations

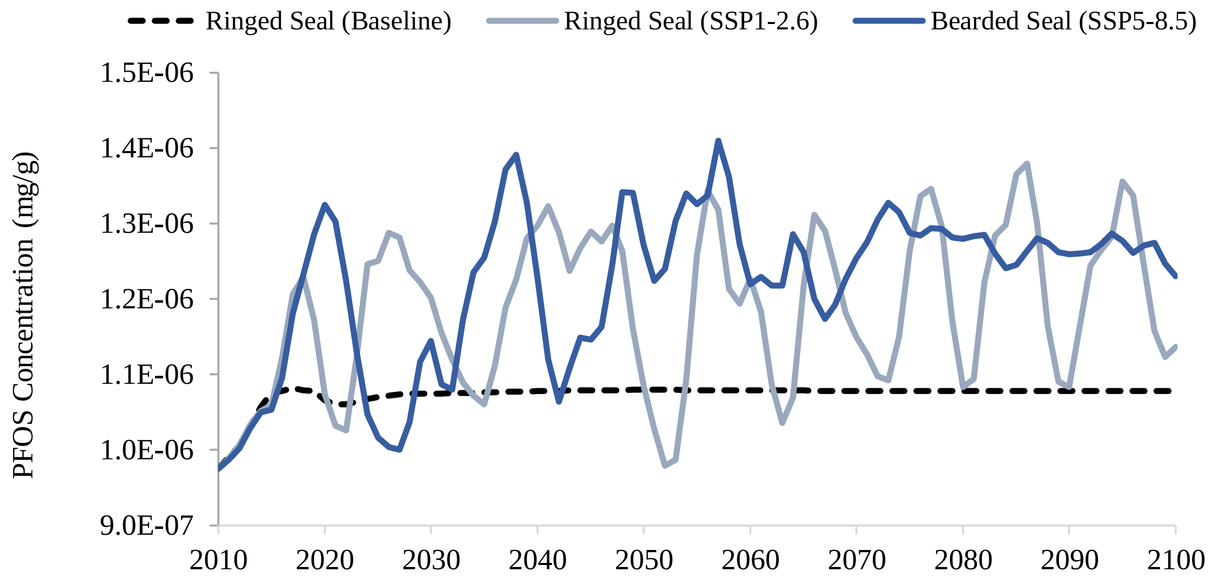
The model projected climate change-driven increases in PFOS concentrations in the BSS food web over the 2010-2100 time period. The average concentrations in the last thirty years of the model period (2070-2100) are shown in Table 2.6. PFOS concentrations were higher under both climate change scenarios compared to the baseline for all consumer species, with the concentrations being greater in the high-emissions scenario relative to the low-emissions scenarios. PFOS concentrations in primary producer species were lower under both climate change scenarios relative to baseline. While greater concentrations were generally observed under the high emissions scenario, SSP5-8.5, there was a limited difference in PFOS concentrations between the two climate change scenarios. The differences in PFOS concentrations between scenarios were more pronounced in higher trophic levels, which was expected due to the biomagnification potential of PFOS.

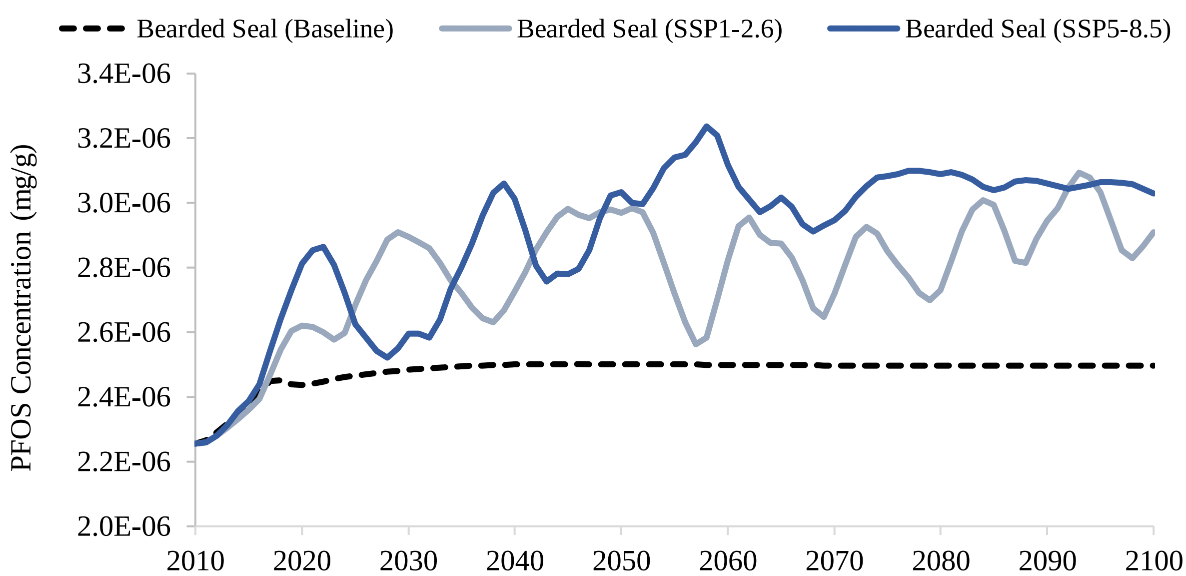
Table 2.6. *Percentage difference in each climate change scenario relative to baseline, using the last 30 years of the model period as representative for end of century (2100).*

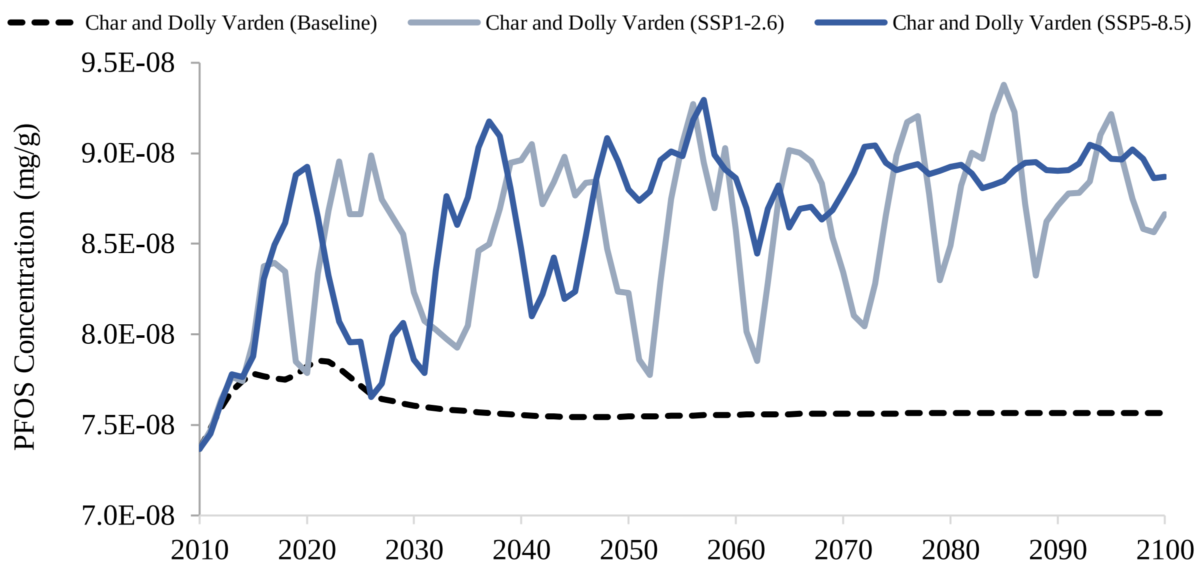
Functional Group	Climate Change Scenario	% Difference in 2100	95% CI
Beluga	2.6/Baseline	19	1.0
	8.5/Baseline	30	1.8
Ringed Seal	2.6/Baseline	13	6.5
	8.5/Baseline	18	1.4
Bearded Seal	2.6/Baseline	16	3.1
	8.5/Baseline	22	0.9
Char and Dolly Varden	2.6/Baseline	16	3.3
	8.5/Baseline	18	0.6
Cisco and Whitefish	2.6/Baseline	9	2.8
	8.5/Baseline	11	0.6
Arctic and Polar Cod	2.6/Baseline	10	3.2
	8.5/Baseline	12	0.6
Large Copepods	2.6/Baseline	6	4.1
	8.5/Baseline	10	0.6
Pelagic Producer	2.6/Baseline	-10	3.9
	8.5/Baseline	-16	0.5
Ice Algae	2.6/Baseline	-14	5.1
	8.5/Baseline	-21	2.5

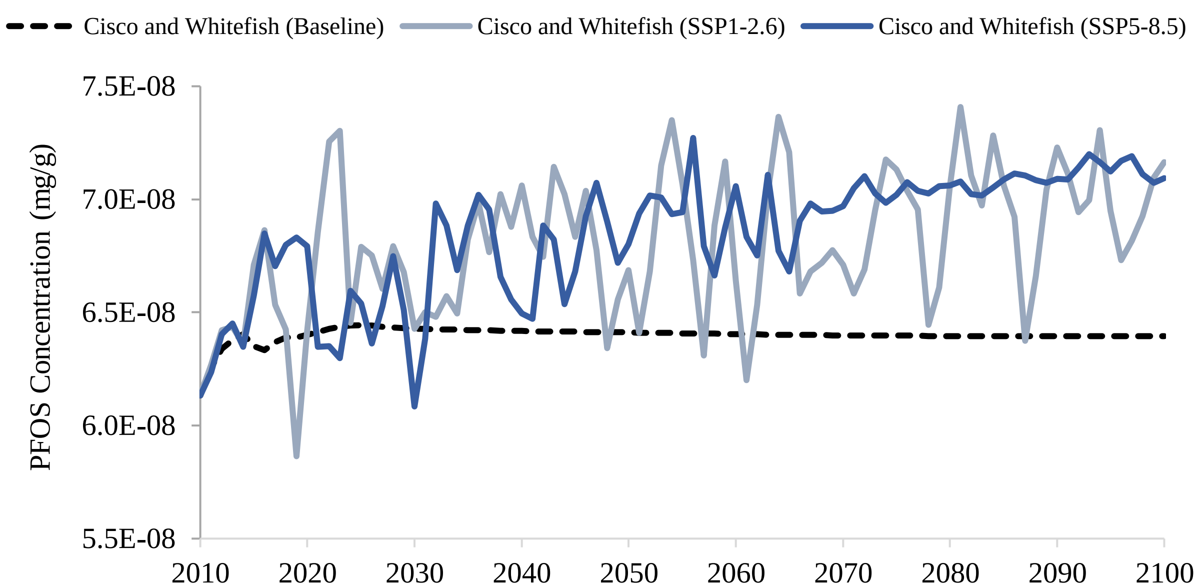
There are two trends observed across species, shown in Figure 2.14. One is the greater variation in PFOS concentrations under SSP1-2.6 relative to SSP5-8.5, which can be attributed to the combination of greater variation in SST and SIC changes year to year from 2010-2100. The second is a divergence in PFOS concentrations between SSP1- 2.6 and SSP5-8.5 around 2070. This is primarily driven by the decrease in SIC under SSP5-8.5, which begins to decline more around 2065. The resulting effect is a sudden decline in ice algae biomass by 2070, followed by a notable increase in biomass of all other species and decreases in variation of consumer diets, all of which contribute to steadier PFOS concentrations from 2070-2100. While the impact of this drop in SIC just before 2070 impacts all species, the drivers for changes are based on various species' positions in the food web and interactions with both SST and SIC changes.

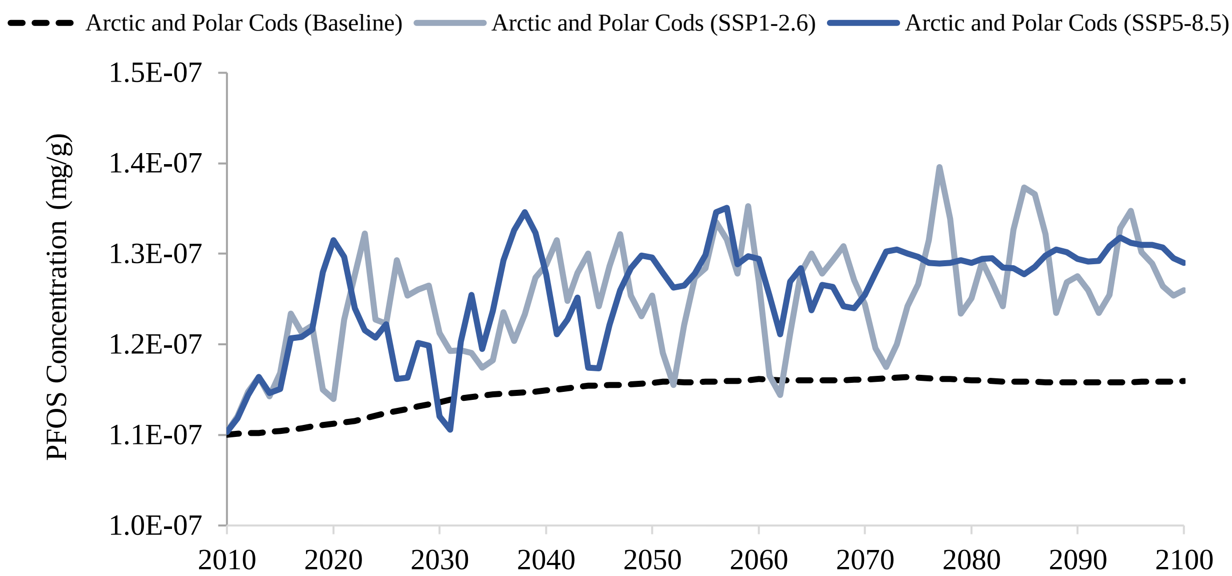


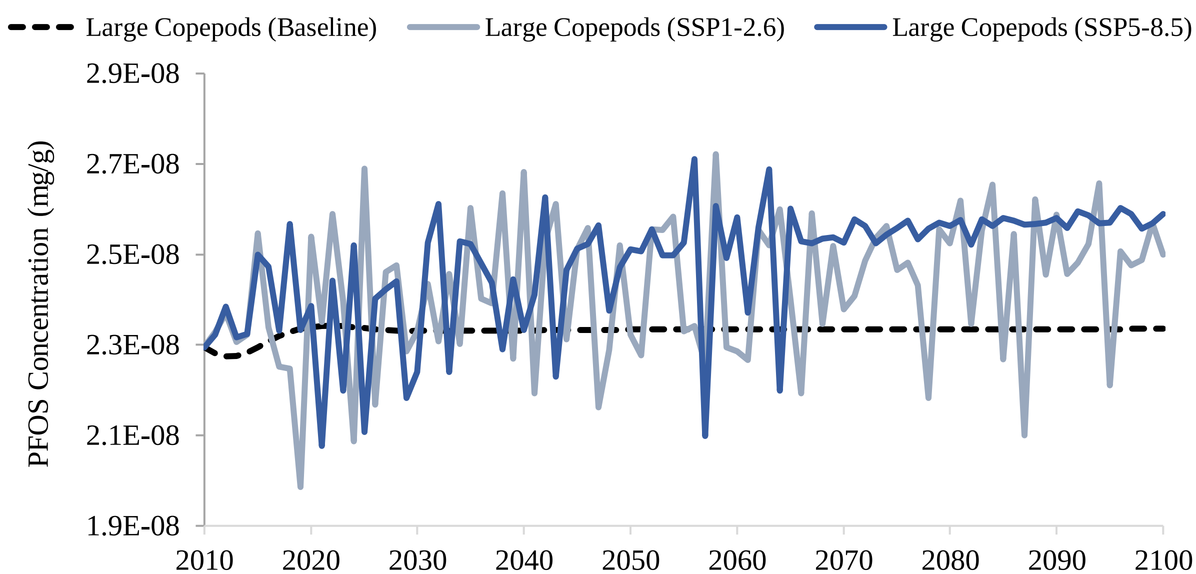

Ringed Seal (Baseline)
Ringed Seal (SSP1-2.6)
Bearded Seal (SSP5-8.5)
PFOS Concentration (mg/g)
1.5E-06
1.4E-06
1.3E-06
1.2E-06
1.1E-06
1.0E-06
9.0E-07
2010
2020
2030
2040
2050
2060
2070
2080
2090
2100

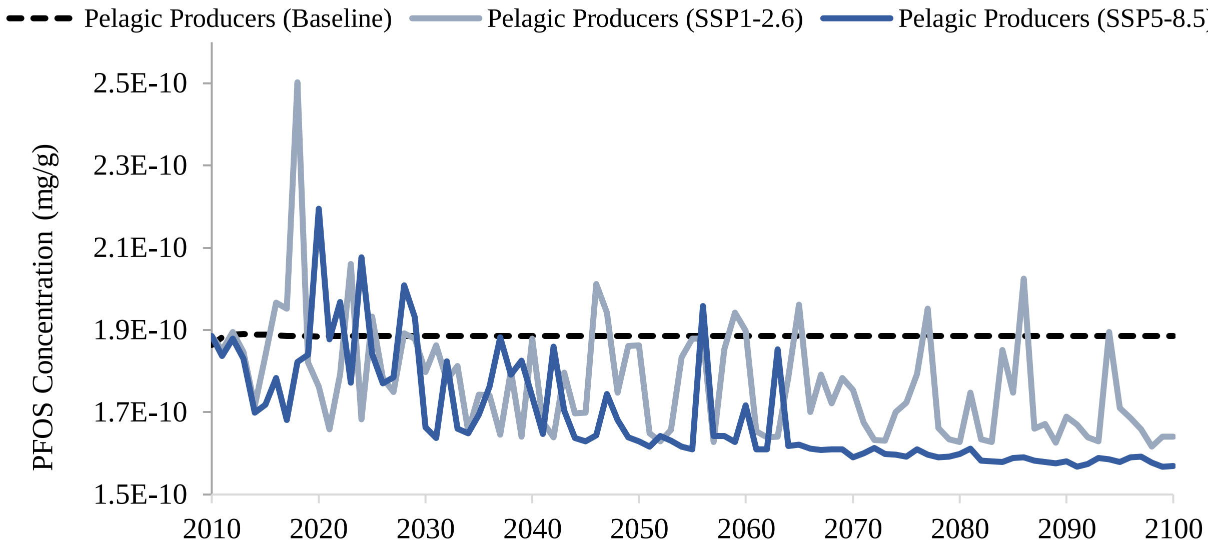

Bearded Seal (Baseline)
Bearded Seal (SSP1-2.6)
Bearded Seal (SSP5-8.5)
PFOS Concentration (mg/g)
3.4E-06
3.2E-06
3.0E-06
2.8E-06
2.6E-06
2.4E-06
2.2E-06
2.0E-06
2010
2020
2030
2040
2050
2060
2070
2080
2090
2100


Char and Dolly Varden (Baseline)
Char and Dolly Varden (SSP1-2.6)
Char and Dolly Varden (SSP5-8.5)
PFOS Concentration (mg/g)
9.5E-08
9.0E-08
8.5E-08
8.0E-08
7.5E-08
7.0E-08
2010
2020
2030
2040
2050
2060
2070
2080
2090
2100


Cisco and Whitefish (Baseline)
Cisco and Whitefish (SSP1-2.6)
Cisco and Whitefish (SSP5-8.5)
PFOS Concentration (mg/g)
7.5E-08
7.0E-08
6.5E-08
6.0E-08
5.5E-08
2010
2020
2030
2040
2050
2060
2070
2080
2090
2100


Arctic and Polar Cods (Baseline)
Arctic and Polar Cods (SSP1-2.6)
Arctic and Polar Cods (SSP5-8.5)
PFOS Concentration (mg/g)
1.5E-07
1.4E-07
1.3E-07
1.2E-07
1.1E-07
1.0E-07
2010
2020
2030
2040
2050
2060
2070
2080
2090
2100


Large Copepods (Baseline)
Large Copepods (SSP1-2.6)
Large Copepods (SSP5-8.5)
PFOS Concentration (mg/g)
2.9E-08
2.7E-08
2.5E-08
2.3E-08
2.1E-08
1.9E-08
2010
2020
2030
2040
2050
2060
2070
2080
2090
2100



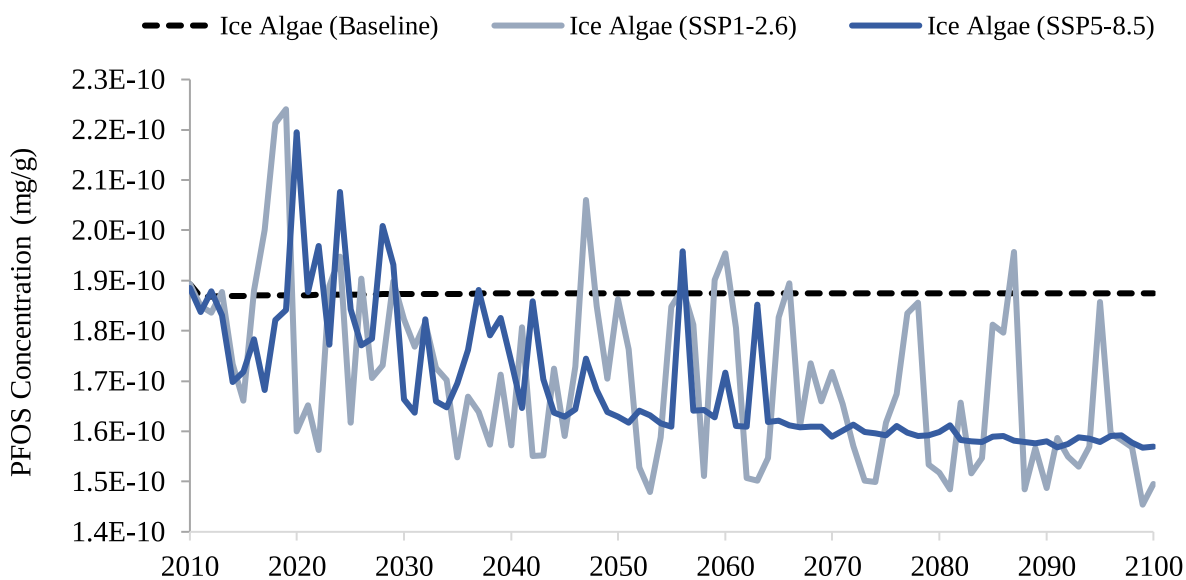


Figure 2.14. *Projected PFOS concentrations in beluga, ringed seal, bearded seal, Arctic char and dolly varden, cisco and whitefish, large copepods, pelagic producers, and ice algae, by species, under two climate change scenarios, SSP1-2.6 and SSP5-8.5, and baseline, from 2010-2100.*

Projected PFOS concentrations in beluga were most affected under the climate scenarios relative to baseline. This is likely due to the effect of biomagnification of PFOS on higher trophic level species, as well as the general increase in biomass under both climate change scenarios. Beluga had the lowest variation in PFOS concentrations over the model period, despite also experiencing the divergence between SSP1-2.6 and SSP5-8.5 around 2070. For beluga, this is likely driven by a greater rate of increase in biomass after 2060 and an increase in the amount and consistency of Arctic and polar cod consumption after 2070 under SSP5-8.5. The proportion of cods in the beluga diet ranges year to year from 25-58% of the diet in 2010-2070 and from 43-48% year to year in 2070-2100, during which there is a greater and sustained increase in cod biomass and predation mortality to beluga. This change in abundance and vulnerability of cods to beluga predation is driven by the decreases in SIC, which after 2070 exponentially increases the vulnerability of cods to predation from the decreased ability to hide within sea ice. Increased consumption of cods drives a higher concentration of PFOS in beluga, as cods have a higher PFOS concentration by one order of magnitude than other species in the beluga diet: cisco, whitefish, herring, and smelt.

Ringed and bearded seals also had greater variation in PFOS concentrations over the model period under SSP1-2.6 compared to SSP5-8.5, and concentrations under both scenarios were higher than baseline. Similar to beluga, this can be attributed to an increase in predation of cods under SSP5-8.5 relative to SSP1-2.6 due to vulnerability of cods with decreased SIC. Ringed seals consume cods more consistently, as well as Arctic char and dolly varden, and decrease consumption of macro-zooplankton, contributing to the increased PFOS concentration under SSP5-8.5. This effect is less pronounced than beluga, most likely due to the greater variation in the diet of ringed seals, which is roughly 15-20% cods versus 40% for beluga. Bearded seals decrease consumption of arthropods in favour of cods, which also make up 15-20% of the diet, similarly driving the increase in PFOS concentrations post-2070 under SSP5-8.5.

PFOS concentrations in Arctic and polar cod had greater variation under SSP1-2.6 as consistently observed, however were much less impacted by the divergence between scenario concentrations after 2070. This is likely due to beluga and seals being more directly influenced by the vulnerability of cods to predation, whereas cods are the group driving this at higher trophic levels. Despite the increased predation, the biomass of the Arctic and polar cod group did increase under both climate change scenarios and more so under SSP5-8.5. This increase is at a lesser magnitude than other pelagic fishes, particularly after 2070, which can explain the smaller differences between climate change scenarios observed in cods. The increased biomass also likely prevented significant competition between beluga and seals for cods as prey, with those groups having increased their consumption of cods. The increased biomass is also the likely driver of increases to PFOS concentrations due to the increase in production associated with increased SST, rather than a change in diet composition, which remains as the total diet composition remains relatively similar between climate change scenarios. Similarly, the Arctic char, dolly varden, cisco, and whitefish functional groups saw increased PFOS concentrations driven by increased biomass, therefore production from increased SST, as there was minimal change to dietary composition between climate change scenarios.

Large copepods followed a similar trend to the pelagic fishes, with more varied concentrations under SSP1-2.6, steadier under SSP5-8.5, and both trending higher than baseline. There was greater overlap with baseline concentrations relative to fishes, likely due to a lower amount of PFOS accumulation in copepods as lower trophic level organisms. The trend of steadier PFOS concentrations seen after 2070 under SSP5-8.5 was again observed here, though at a relatively greater magnitude than fishes. This was driven by a change in dietary composition due to the significant drop in ice algae biomass after 2060. Large copepods increase the proportion of microzooplankton in the diet by approximately 10% to replace the loss of ice algae, which in turn drives the increase in PFOS concentration as microzooplankton have higher concentrations as a higher trophic level functional group than ice algae as producers.

Contrary to trends in higher trophic levels, PFOS concentrations in both pelagic producers and ice algae decreased under both climate change scenarios relative to baseline. The most likely driver for this was a marked change in biomass from climate change experienced by both groups, though each is driven by a different climate factor. As the concentration of PFOS in each functional group is given in PFOS (t) / biomass (t), both PFOS uptake and biomass change drive the concentration. Under both climate change scenarios, pelagic producer biomass increased steadily and notably over the model period, due to the combined effects of both increased SST driving production and increased sunlight from decreased SIC. Under both climate change scenarios, the cumulative amount of PFOS in pelagic producers increased, however this increase was outpaced by the increase in pelagic producer biomass. As a result, the concentration of PFOS in this functional group decreased over time under both climate change scenarios. Inversely, the decrease in ice algae PFOS concentration was driven by a decrease in biomass under both climate change scenarios. The decrease in SIC under both climate change scenarios resulted in a decrease in ice algae biomass to nearly zero, which in turn reduced the production of the group as a whole and led to a decrease in cumulative PFOS concentration independent of biomass. Thus, unlike pelagic producers, this decrease in PFOS concentration was a function of decreased PFOS levels in ice algae as a group and not a discrepancy between the rates of increased PFOS within the group and increased biomass of the group. PFOS concentrations under SSP5-8.5 were lower and had much less variation year to year as a result of the much greater and steady decline in SIC, a nearly 40% decrease over the model period under this climate change scenario as shown in Figure 2.5. Under SSP1-2.6, the decrease in PFOS concentration was not as great and there was much more variation year to year resulting from the magnitude of SIC decrease by approximately 10%, as shown in Figure 2.4.

There are several possible contributing factors to the minimal differences between PFOS concentration levels under the two climate change scenarios. The most likely is the interaction between the climate changes to SST and SIC under each scenario and the corresponding impacts on trophodynamics. The observed increases to SST and decreases to SIC were greater under SSP5-8.5 than SSP1-2.6 as shown in Figures 2.4 and 2.5. Under SSP1-2.6 the relative change from 2010 to 2100 is a factor of around 1.5 for SST, and 0.9 for SIC. Under SSP5-8.5, the relative change from 2010 to 2100 is a factor of around 2.5 for SST, and 0.6 for SIC. These changes from baseline largely fall within the ability of species to adapt, as defined by the MTT integrated into the forcing functions for SST on consumers, the environmental response functions for SST and SIC on primary producers, and the mediation functions for polar bear predation success and vulnerability of cods to predation. As such, the impacts of changes to SST and SIC under each climate change scenario did drive differences in concentrations. However, these differences were not large as they were primarily within the range of species' environmental preferences. Even with ice algae biomass decreasing to almost zero by end of century under SSP5-8.5, the corresponding increase in pelagic producer biomass served as a replacement in the dietary composition for zooplankton and copepods, the species which consume ice algae. Due to the similar concentrations in pelagic producers and ice algae, this change did not contribute to a large difference in PFOS concentrations in the zooplankton or copepod groups, and subsequent species at higher trophic levels. An additional possibility is that the environmental response function for pelagic producers to SST may have diminished the difference of effects between the climate change scenarios. The environmental preference function was defined for a positive, linear relationship of SST driving pelagic producer production between 0.5- and 1.5-times baseline (Figure 2.8). At a relative change in SST above 1.5, the corresponding increase to production remained at a factor of 1.5, due to the uncertainty in pelagic producer response above this threshold. However, it is likely that production does increase beyond this factor of 1.5 as increases have been observed up to around 5°C though not in a consistent or linear manner (Gillies, 2022; Steiner et al., 2019), so it is possible that the impacts of SST increase under SSP5-8.5 were not fully captured and may have contributed to a smaller difference between the two climate change scenarios.

These results are comparable to similar studies modelling projected impacts of climate change on contaminants, particularly methyl mercury and PCBs (Alava et al., 2018). The PFOS concentrations in higher trophic level species compared to lower ones are consistent across modelling approaches, indicative of the ability of these contaminants to biomagnify in marine food webs. The effects of projected climate changes here are consistent with the projected effects on mercury in a Pacific Northeast food web, where concentrations trend higher under climate change scenarios compared to baseline, where high-emission scenarios trend higher than low-emission scenarios; however, the effects here are greater compared to baseline than for mercury (Alava et al., 2018). This study also projected decreases in PCB concentrations in higher trophic level species such as chinook salmon and southern resident killer whales, which is inconsistent with the projected PFOS increases here. This is likely due to the inclusion of PCB environmental concentration changes in the Pacific Northeast model, which accounts for decreases in PCB levels due to their phase-out over the past decades. Given the assumption here that environmental PFOS concentrations remain the same, this discrepancy is also justified. The projections here of decreased PFOS concentrations relative to baseline in both producer functional groups is inconsistent with the projections for both methyl mercury and PCBs in the Pacific Northeast. This is potentially due to the significant role of SIC in the BSS as an Arctic ecosystem driving the drop in ice algae biomass and subsequent increase in primary production biomass associated with decreases in SIC under climate change. However, these results remain unusual and would be valuable to compare to any future studies examining projected climate changes in Arctic marine ecosystems.

Trophic Magnification of PFOS

The estimated trophic magnification factor (TMF) of the whole food web with polar bears as the apex predator is 15.32, without polar bears is 14.74, without polar bears or migratory mammals is 12.84, and without mammals is 10.04, shown in Table 2.7. These TMF values and strong correlation (Figure 2.15) indicate greater magnification of PFOS concentrations in higher trophic level species.

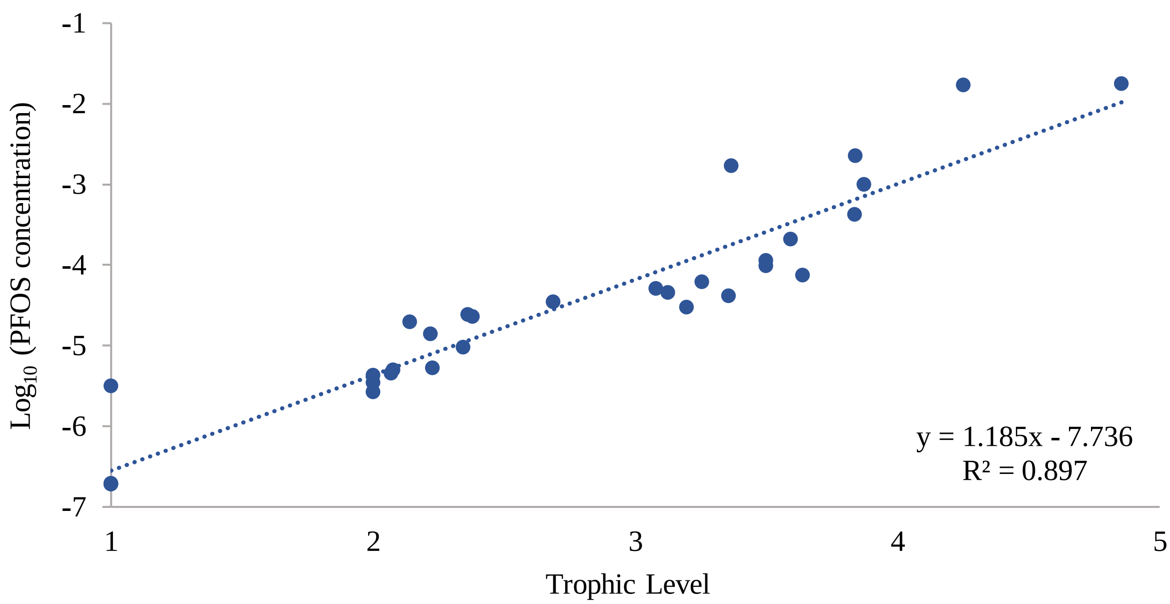


Figure 2.15. *PFOS trophic magnification in the BSS food web under steady state.*

Table 2.7. *Trophic magnification factors of PFOS in the BSS food web under SSP1-2.6, SSP5-8.5, and baseline climate change scenarios.*

Baseline	**SSP1-2.6**	**SSP5-8.5**	**Species included**
15.3	16.6	17.3	All
14.7	16.0	16.3	Minus polar bear
12.8	13.9	14.2	Minus polar bear + beluga
11.8	12.8	13.0	Minus polar bear + whales
10.0	10.8	11.0	Minus mammals

These TMFs are similar, but slightly higher than, those calculated for mercury within the BSS, which are 11.1, 9.8, and 7.8, for the food web without polar bears, without polar bears and migratory mammals, and without mammals (Li et al., 2022). This is slightly lower than the TMF of 17.4 for PFOS in a Hudson Bay marine food web (Kelly et al., 2009), and is higher than a range of other TMFs estimated for other marine food webs such as those in the St. Lawrence seaway (1.6) and Baffin Island (3.1); and for bottlenose dolphin food webs within the Atlantic Ocean (3.9) and Lake Ontario (5.5) (Houde et al., 2006; Martin, Whittle, et al., 2004; Munoz et al., 2022; Tomy et al., 2009). The much higher TMF estimated here is most likely due to the large range in trophic levels included in the analysis (1 – 4.8), encompassing algae up to polar bears, which is also the range covered in the Hudson Bay ecosystem TMF of 17.4, in contrast to the other TMFs which span a lower range of trophic levels (Kelly et al., 2009). The higher TMFs estimated here suggest that polar ecosystems may biomagnify PFOS more efficiently than lower latitude ecosystems in part due to the greater range in trophic levels, as is understood to be the case for mercury (Li et al., 2022).

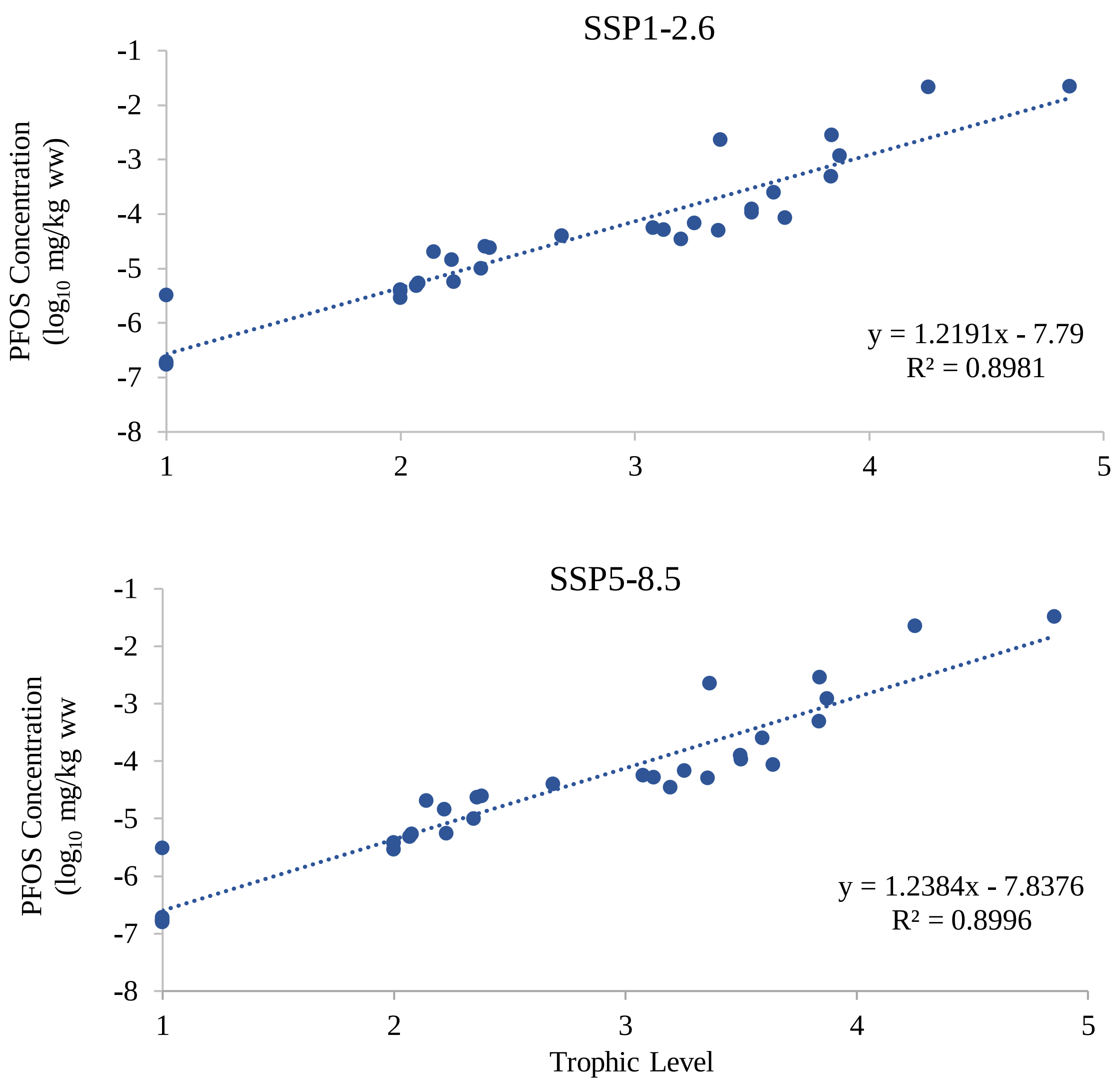


Figure 2.16. *PFOS trophic magnification in the BSS food web under SSP1-2.6 (upper panel) and SSP5-8.5 (lower panel).*

There is a negligible difference between the TMFs under the two climate change scenarios, both when compared to each other and to the baseline scenario. This is consistent with the minimal differences in PFOS concentrations shown between the climate scenarios. In addition to the drivers of the minimal concentration differences, it is possible that this can also be attributed in part to the decreases in PFOS concentration in both pelagic producers and ice algae under both climate change scenarios. The decreased concentrations in primary producers do not lead to a bio dilution effect given the bioaccumulation potential of PFOS. However, it is possible that this reduces the magnitude of biomagnification, causing the TMFs to be similar for each climate change scenario and baseline. This negligible effect of climate change on TMFs is consistent with climate change modelling of both PCBs and methyl mercury in the Pacific Northeast, where TMFs for each were similar under climate change scenarios and baseline (Alava et al., 2018). This was attributed in part to decreases in biomass of key species under both climate change scenarios toward the end of the century, theorizing that a possible maximum level of biomagnification potential due to contaminant oversaturation could result in any additional trophic magnification going undetected (Alava et al., 2018). Given the increases in biomass of all species in the BSS food web under the climate change scenarios, with the exception of ice algae, it is unlikely that this same mechanism can be attributed to the lack of difference in PFOS TMFs in the BSS food web. This is not unexpected, however, due to the differences in the marine ecosystem environments and effects of climate change impacts. The colder sea temperatures of the Arctic may delay the occurrence of SST ranges that are uninhabitable for many species relative to more southern marine ecosystems by end of century, and both contaminant concentrations in primary producers increase under climate change scenarios in the Pacific Northeast ecosystem therefore are unlikely to play a significant role in TMF differences between climate change scenarios (Alava et al., 2018; Fossheim et al., 2015; Sappal et al., 2014).

Limitations and Future Opportunities

Modelling approaches have inherent uncertainty built in, partly due to parameters set in the attempt at reconstructing natural systems, the complexity of which can rarely be captured by computer systems, also due in part to limitations in data availability. Table 2.8 summarizes the key assumptions made in the Ecotracer parameterization and Ecosim scenarios, which are discussed in depth earlier in the methods section. The limitations respecting the Ecotracer and Ecosim portions of the model are discussed here in the context of interpreting results and areas for future research. There are also limitations arising from parameterizing the Ecopath base model, which are mentioned, with full details available in previous publications (Hoover et al., 2021, 2022).

Table 2.8. *Summary of key assumptions made in Ecotracer and Ecosim.*

Assumption	**EwE Parameter**
PFOS input to the environment is constant	Ecotracer – Base inflow rate
PFOS does not degrade in the environment	Ecotracer – Decay rate
No PFOS entering system via other organisms	Ecotracer – Concentration in immigrating biomass
PFOS does not undergo biotransformation	Ecotracer – Metabolic decay rate
The assimilation efficiency for mammals is the same as fish	Ecotracer – Proportion of contaminants excreted
Direct uptake from water similar between similar taxonomic species of invertebrates and producers	Ecotracer – Direct absorption rate
Direct uptake of PFOS from water is negligible for fish and mammals	Ecotracer – Direct absorption rate
No MTT required for birds and mammals as they are endothermic	Ecosim – Forcing function
Positive, linear relationship between forcing functions and production of consumers	Ecosim – Forcing function
Positive, linear relationship between SST and pelagic producer production between 0.5- and 1.5-times the baseline SST	Ecosim – Environmental response
Negative, linear relationship between SIC and pelagic producer production between 0.5- and 1.5-times the baseline SIC	Ecosim – Environmental response

Positive, linear relationship between SIC and ice algae production between 0- and 1.5-times the baseline SIC	Ecosim – Environmental response
Positive, exponential relationship between SIC and the foraging area and vulnerability of prey to polar bears	Ecosim – Mediation function
Negative, exponential relationship between SIC and the vulnerability of cods to predation	Ecosim – Mediation function

The Ecosim scenarios were constructed to simulate predicted climate changes over the period of 2010-2100. The primary limitations that arise from this portion of the model are in the way the parameters were chosen to characterize these climate changes. SST and SIC were chosen as the two climate change factors as most important factors in the Arctic. However, other biogeochemical factors such as pH, salinity, and oxygenation are also altered by climate change and play a role in ecotrophic dynamics of the BSS food web (Sora et al., 2024). These were not incorporated in this model due to time limitations; however, future work would benefit from incorporating these variables into Ecosim scenarios to force climate change within the EwE modelling approach. Further, some species are changing their habitat ranges as a result of climate change, and these geographical shifts are not captured in the current EwE modelling approach. An additional routine within the modelling software known as Ecospace is able to capture geographic aspects of ecosystem and food web dynamics, was not used due to time limitations, but could further enhance the way in which climate change parameters are captured in future models. This is especially pertinent given that beluga are a migratory species, as well as being an important country food for ISR communities. They have recently been reported in areas outside of their historic range, suggesting there may already be changes to their migratory pattern or home range as a result of climate change (Loseto et al., 2018).

Additional limitations that arise from data gaps include how the SST and SIC impacts on the food web were defined within the model. The impacts of SST used MTT for ectothermic species within the food web. However, the data MTTs in Arctic species to create the forcing functions was much more limited than other similar approaches in more southern marine food webs such as the Pacific Northeast (Ainsworth et al., 2011; Alava et al., 2018; Guénette et al., 2014). The relationship between increasing SST and impacts on ectothermic marine species may not be as well characterized due to this lack of data, specifically that the impacts of increasing SST may be underestimated as the species available are often ones with a very large habitat area therefore a large range of MTT, and any impact of warming temperatures on these organisms may not be fully captured by the large MTT range.

Lastly, the biogeochemical aspects of climate change are not currently integrated into the Ecosim scenarios but likely also play a role in the climate change-contaminant interactions in the BSS. Transportation of PFOS is not taken into account within the model due to the lack of data on PFOS loading to the Beaufort Sea. However, this would be important to consider. The Mackenzie River likely plays a large role in PFOS transportation due to glacial and snow melt as well as transport from any other more inland sources of PFOS given the expanse of its watershed, therefore characterizing what PFOS transportation takes place, if there is a dilution effect from the large amount of freshwater, would all be valuable to consider. Long-range transportation would be more difficult to quantify but also valuable, as changing wind patterns and ambient air temperatures could play an important role in the transportation of PFOS to the BSS either from increasing volatilization of precursors for transport and/or increasing transport and deposition overall from changing wind and precipitation patterns.

A lack of data availability is the primary source of limitations in parameterizing Ecotracer. While PFOS is one of the most studied compounds in the larger class of PFAS, its toxicokinetics are not as well researched compared to other contaminants like PCBs or mercury. As such, one of the most important parameters, direct absorption rates, were estimated using concentration ratios from empirical data rather than laboratory studies with fewer potential sources for error. In some cases, this may prove to be an advantage where the literature reports were for Arctic organisms. Therefore, the impact of colder water temperatures on uptake rates would be factored into those calculations; this was only done for a few species and remained a source of error. Additionally, there are very limited values for assimilation efficiencies derived from lab studies, so values for one fish species (rainbow trout) and one invertebrate (*Daphnia magna*) were used for all species based on the best assumptions available for transferability between species (Li et al., 2022; Sun et al., 2022).

Elimination rate and biotransformation are also gaps in research; these processes may be taking place but are not well characterized in species. Due to a lack of data the influences of these processes were assumed to be negligible. However, as discussed earlier, there are discrepancies in some sampling studies between expected concentrations of PFOS in various tissues and species, such as very high concentrations in polar bears and evidence of accumulation in blubber leading, suggestive of some biotransformation of PFOS itself may be taking place in some species such as mammals, and this was not accounted for in the model due to the lack of toxicokinetic data. It could also reflect the biotransformation of precursors, which were also not included in the model due to the lack of data as well as challenges integrating more than one contaminant into the model at a time. As research continues this will be valuable additions to parameterizing the model to better capture the toxicokinetics of PFOS and role of precursors in its bioaccumulation.

An additional limitation here that is present and may remain is the impact of water temperature on the toxicokinetics, where lab derived values are often done in both aquatic species and at ambient temperatures, which are not always representative of the uptake rates for Arctic marine species. PFOS concentrations in and uptake from sediment is also not accounted for in the current Ecotracer model, which presents a limitation in accounting for the role of benthos in PFOS uptake to the food web.

Lastly, sensitivity analyses were not run due to time constraints, limiting the ability to fully interpret what toxicokinetic parameters are most influential on PFOS concentrations to better characterize the trophodynamic relationships related to PFOS movement through the food web. Future studies could incorporate this to paint a fuller picture of the BSS and what factors are most important for PFOS accumulation to the food web and various species.

2.5 Conclusion

This modelling study is the first to examine how future climate changes may impact PFOS concentrations and amplification in a marine food web. Results indicate that climate change is likely to cause increases to PFOS concentrations in consumer species within the Beaufort Sea Shelf food web between 2010-2100 relative to a baseline scenario of no climate change. PFOS concentrations were generally higher under SSP5-8.5, representative of a high-emissions climate change scenario relative to SSP1-2.6, a low-emissions climate change scenario. However, these differences between climate change scenarios were marginal, and smaller than the differences compared to baseline. The changes in PFOS concentrations due to climate change were driven by both SST and SIC impacts to diet composition and changes in species biomass. Amplification of PFOS concentrations is anticipated to be greater in species at higher trophic levels given the high

TMFs, therefore concentrations in beluga are likely to be the most impacted by climate change. These results provide a first step in understanding the climate change-food web-contaminant dynamic at play for PFOS, which has been understudied relative to other POPs such as PCBs. Given the status of PFAS as a class of chemicals of emerging concern, these results provide insight into how this group of contaminants may be impacted by future climate changes, with specific insight into PFOS as one of the most commonly found types of PFAS in the environment. Future studies should continue to add in additional climate change considerations, PFOS transportation, and PFOS toxicokinetics, as they become available, to improve our understanding of these climate-pollution interactions of PFOS and PFAS.

CHAPTER 3. POTENTIAL CLIMATE CHANGE IMPACTS ON PFOS DIETARY EXPOSURE AND HEALTH RISK IN THE INVUIALUIT SETTLEMENT REGION

3.1 Introduction

Per- and polyfluorinated alkyl substances (PFAS) are a group of over 4000 chemicals commonly used in various consumer and industrial products due to their hydrophobic and oleophobic properties (Giesy & Kannan, 2002; Sunderland et al., 2019). These consumer product uses include carpets, furniture, outdoor gear, food packaging, and cookware, while industrially, they are commonly found in aqueous film-forming foams and at airports for spraying down planes (Sunderland et al., 2019). PFAS have been manufactured and sold since the 1940s, therefore combined with their wide range of uses have been released to the environment steadily from both the manufacturing process and use of products. PFAS are also incredibly stable and persist in the environment, therefore also accumulate in environmental compartments over time (Blum et al., 2015; Giesy & Kannan, 2002).

Perfluorooctane sulfonate (PFOS) is considered a ‘legacy’ PFAS as one of the oldest and most common compounds in the greater class. In the last two decades it has been regulated due to its environmental persistence and potential human health effects associated with exposure, resulting in its addition to the Stockholm Convention for Persistent Organic Pollutants in 2009 (Blum et al., 2015; Idowu et al., 2013). Being an Annex B compound, it is still produced, however only in small amounts, therefore the PFOS released to the environment is largely from product uses rather than large sustained manufacturing processes (Idowu et al., 2013). Despite this regulation, PFOS is effectively ubiquitous in the environment, having been detected in some of the most remote locations in the world, including the Canadian Arctic (Giesy & Kannan, 2001, 2002; Muir et al., 2019). Exposure to PFOS is also associated with a wide range of adverse health effects on the immune system, reproduction, liver and thyroid function, neurodevelopment, and potentially some cancers (Fenton et al., 2021; Health Canada, 2021; Sunderland et al., 2019; Zeng et al., 2019). Its carcinogenicity was evaluated in December 2023, and it has been classified as Group 2B carcinogen, meaning it is possibly carcinogenic to humans based on the evidence available (Zahm et al., 2024). As such, PFOS remains a target for biomonitoring and health risk assessment to minimize adverse health effects (Health Canada, 2021).

It is likely that between 95-100% of the Canadian population has detectable levels of PFOS in their bodies (Health Canada, 2021). Most exposure for Canadians comes from drinking water or seafood consumption, which is especially relevant for coastal and Indigenous communities whose residents may consume more seafood than the general population (Health Canada, 2021; Sunderland et al., 2019; Tittlemier et al., 2007). In Northern Canada, PFOS has been detected and quantified in marine species such as beluga, ringed seal, bearded seal, and Arctic char, all of which are commonly consumed as country foods across Inuit Nunangat (Kenny, Hu, et al., 2018; Kenny & Chan, 2017; Ostertag, Tague, et al., 2009). Biomonitoring programs in Nunavik, northern Quebec, have also measured PFOS in many country foods and have found associations between PFOS serum concentrations in Inuit women and consumption of marine country foods, indicating marine country foods as a likely source of exposure to PFOS for Inuit (Caron-Beaudoin et al., 2020; Dallaire et al., 2009). Country foods are an incredibly important part of Inuit diet, both for their nutritional and cultural value (Kenny, Hu, et al., 2018).

Due to its presence in all environmental compartments within the Arctic, negative health impacts, and exposure from consumption of marine country foods, PFOS is a contaminant of high interest and is considered a contaminant of emerging concern by the Arctic Monitoring and Assessment

Programme (AMAP, 2017; Muir et al., 2019). This is especially pertinent given potential interactions of contaminants with climate changes, such as biogeochemical processes that can impact contaminant transportation and movement through environments, as well as food web structure and function changes that can be associated with climate change as well (Alava et al., 2017; Fossheim et al., 2015; Guénette et al., 2014). This relationship between climate change and contaminants can be a driver for changes in contaminant levels, sometimes increasing them, which is a further area of concern in the Arctic as it warms at a rate roughly three times that of the global average (Alava et al., 2017; AMAP, 2021; Bush & Lemmen, 2019).

Given the health concerns associated with PFOS exposure, potential climate change-contaminant interactions in marine food webs, and source of exposure via marine country food consumption, this study aims to characterize potential future PFOS concentrations in key country foods within the Inuvialuit Settlement Region (ISR). Modelling with the Ecopath with Ecosim (EwE) approach was used to estimate the impacts of climate change on PFOS concentrations in Beaufort Sea Shelf marine species, as described in Chapter 2. Here, the projected concentrations are used to estimate future dietary exposure to PFOS from commonly consumed country foods, and how these exposures compare to the health-based value determined by Health Canada. The objective of this study is to provide a forward-looking estimate of how climate change may impact exposure to PFOS from country foods in the ISR, and how that may translate to associated health risks.

3.2 Methods

PFOS Concentrations & Data Transformation

The raw output from the EwE model was first processed before the calculations were undertaken to estimate dietary exposures and hazard quotients. The concentration units from EwE are tonne of PFOS per tonne of biomass (t/t); they were therefore converted to more appropriate units and generalized to first organism- and then tissue-level concentrations. Only certain tissues are consumed as country foods, therefore the modelled concentrations were processed to be representative of the specific tissues consumed as country foods for each species. The approach here assumes that the change in concentration projected by the model is constant across tissues, therefore any change to overall organism PFOS concentration is of a proportional magnitude to any change to tissue PFOS concentrations.

First, the empirical PFOS concentration of the tissue consumed as country food was determined. This was done by using published concentrations for the appropriate tissue or, if published data for the specific tissue collected in the Beaufort Sea in the 10 years before or after 2010 was not available, I determined a tissue-to-tissue ratio based on observed concentrations for a different Arctic region and applied this to available Beaufort data. For example, the two forms of beluga most commonly consumed as country food are meat and muktuk, namely muscle and blubber. There are no published data of beluga muscle or blubber concentrations in the Beaufort Sea or western Arctic within 10 years of the model baseline year of 2010; only concentrations for liver are available. However, there are published PFOS concentrations of beluga muscle, blubber, and liver in the Eastern Arctic within 10 years of 2010. A concentration ratio of liver to muscle and liver to blubber was calculated using the Eastern Arctic data. Then, those ratios were applied to the liver concentrations in the Beaufort Sea to estimate muscle and blubber PFOS concentrations as best approximations. Then, an index of change from the baseline year of 2010 was created for the projected beluga PFOS concentrations for each baseline and climate change scenarios. The index of change was then used along with the estimated tissue concentrations to calculate projected concentrations for the baseline and climate change scenarios that would be representative of the specific tissues consumed as country foods. These projected concentrations for each tissue/country food were then used in the dietary exposure calculations.

Dietary Data Sources

The dietary consumption data for the ISR was collected during the International Polar Year Inuit Health Survey (IPY-IHS), which took place from 2007-2008 (Egeland, 2010). Dietary intake data for country foods published in the IPY-IHS Final Report for the Inuvialuit Settlement Region were used to determine which country foods were most commonly consumed in the ISR and the respective daily consumption rates (Egeland, 2010). Additional data published for the whole of Inuit Nunangat in Laird et al., (2013)), was used where data was not available in the IPY-IHS ISR Final Report. As the dietary intake data described here has already been published, its use is considered secondary data analysis and not subject to research ethics board approval.

Dietary Exposure Estimates & Hazard Quotients

Per the IPY-IHS final report for the ISR, the most commonly consumed marine country foods within the ISR, in order from greatest to least, are: Arctic char, whitefish, dried beluga meat, and beluga oil. In lieu of beluga oil, calculations were performed for beluga muktuk, another popular country food in the ISR (Kenny, Wesche, et al., 2018). This was done as it is difficult to approximate the PFOS concentration estimates for beluga oil from the EwE model output, which was transformed to tissue-specific concentrations for the respective tissues consumed as country foods using observed tissue concentration ratios in the published literature. Since oil is not a tissue type and therefore not a typical sample considered in biomonitoring studies, there are no observed PFOS concentrations in beluga oil that could be used to approximate a modelled PFOS concentration. Instead, beluga blubber was used. Blubber is not typically consumed alone, but as muktuk, which includes both the skin and blubber of a whale. There are also no observed concentrations of PFOS in beluga skin. Therefore, the modelled PFOS concentrations for beluga blubber and dietary intake data for beluga muktuk were used to calculate dietary exposure estimates and hazard quotients for beluga muktuk.

Dietary exposure estimates (DEE) were calculated for each country food under the two climate change scenarios, SSP1-2.6 (low emissions/high mitigation policies) and SSP5-8.5 (high emissions/low mitigation policies), and baseline (no climate change). This was done using equation 12, adapted from the UN Food and Agriculture Organization, to determine the amount of PFOS consumed per unit of body weight (Mbabazi, 2011).

$$DEE = \Sigma \frac{(\text{Concentration of chemical in food} \times \text{Food consumption})}{\text{Body weight}} \qquad \text{Eq. 12}$$

The ‘concentration of chemical in food’ value is the projected PFOS concentration from the EwE model for each country food under the climate change and baseline scenarios. The values used were an average (Mean +/- SD) of the last 30 years of the model period, 2070-2100. An average of the last 30 years of the model was used to account for year-to-year fluctuations and uncertainty in PFOS concentrations, so the time period of 2070-2100 was used as ‘end of century’. The ‘food consumption’ values were the dietary intake values form the IPY-IHS for each of the four country foods. Lastly, the ‘body weight’ value used was 70kg, the average weight of an adult human used by Health Canada to calculate tolerable daily intake (TDI) limits for PFOS (Health Canada, 2018). This is slightly lower than the average adult body weight from the biometric data reported in the IPY-IHS final report for the ISR, which is roughly 72.34kg (Egeland, 2010). Given these values are quite close, 70kg was chosen for the DEE calculations to be most consistent with the Health Canada TDI units. Additionally, using 70kg may offer a more conservative approach to calculating hazard quotients, as it is slightly lower than the reported average body weight in the ISR as per the IPY-IHS.

The calculated DEEs for each country food under the three climate change scenarios were then compared to Health Canada's TDI. To compare the DEE and TDI, hazard quotients were calculated as described in equation 13 (Ostertag, Tague, et al., 2009).

$$Hazard\ Quotient = \Sigma\ \frac{\text{DEE}}{\text{Health Canada TDI}} \qquad \text{Eq. 13}$$

Health Canada has established three TDI values for PFOS, based on endpoints for hepatocellular hypertrophy (liver function), thyroid hormone T3, T4, and TSH levels (thyroid function), and incidence of hepatocellular adenomas and carcinomas (cancer risk) (Health Canada, 2018). These are, respectively, 0.000066, 0.0001, and 0.0011, mg/kg bw/day (Health Canada, 2018). The most conservative value is for liver function, which was used here to provide one TDI value that is considered protective for all three measured health risks from PFOS; therefore, it is the most comprehensive value to use (Health Canada, 2018). Hazard quotients (HQ) are used to provide an indication of the daily exposure to a contaminant that is occurring relative to a known value under which exposure is considered safe, in this case, the TDI. An HQ < 1 indicates that the daily exposure is lower than the daily limit therefore, unlikely to be a concern for health, and an HQ >

1 indicates the daily exposure is over the daily limit and may be at a level associated with negative health impacts and therefore of concern for health risks.

3.3 Results and Discussion

Concentrations of PFOS in marine country foods

The reported dietary intake values from the IPY-IHS for the ISR are presented in Table 3.1. The country food consumed in the highest amount is Arctic char, which is one order of magnitude higher, followed by whitefish, dried beluga meat, and beluga muktuk. Beluga muktuk is made up of the skin and adjacent layer of blubber (Laird et al., 2013).

Table 3.1. *Daily consumption amounts for each of the four most consumed country foods in the ISR, from the IPY-IHS.* Data accessed from Egeland, (2010) and Laird et al., (2013).

Country Food		**Consumption (g/day)**
Arctic char	meat	116.0
Whitefish	meat	26.0
Beluga	meat (dry)	15.8
	muktuk	11.0

The projected PFOS concentrations in each of the country foods based on the model projections under two climate change and baseline scenarios are presented in Table 3.2. The concentrations were generally higher under SSP5-8.5, lower under SSP1-2.6, and lowest with no climate change. While these differences between concentrations under each climate change scenario and baseline were small, they were all within the same order of magnitude. Projected concentrations of PFOS in marine country foods were consistent with those reported in empirical studies, shown in Table 2.5. Concentrations were higher in species at higher trophic levels, and liver concentrations were markedly higher than in other tissues, which is in line with the observation that PFOS biomagnifies in food webs and accumulates in the liver (Houde et al., 2006; Martin et al., 2003; Tomy et al., 2009). The projected concentrations in country foods under baseline are similar to observed concentrations reported for country foods across Nunavut in the early 2000s, for dried beluga meat (3.6E-06, 1.6E-06 mg/g), beluga muktuk (4.0E-07 mg/g), and Arctic char (5.0E-07 mg/g) (Ostertag, Tague, et al., 2009). While similar, the projected results here are higher, which can be expected given the ability of PFOS to biomagnify and the roughly 100-year time difference between the collection of the samples tested for the observed concentrations and the modelled values here.

Table 3.2. *Projected PFOS concentrations in specific tissues consumed as country foods by 2100.*

Species	**Tissue**	**Projected PFOS concentration (mg/g)**					
		Baseline		SSP1-2.6		SSP5-8.5	
		Mean	SEM	Mean	SEM	Mean	SEM
Arctic Char	muscle	4.9E-07	6.5E-10	4.8E-07	3.4E-09	4.9E-07	6.5E-10
Whitefish	muscle	4.1E-07	7.9E-12	3.6E-07	2.4E-09	3.7E-07	5.2E-10
Beluga	muscle (dry)	3.6E-06	1.8E-10	4.4E-06	9.5E-09	4.7E-06	1.6E-08
	skin+blubber	1.8E-06	9.1E-11	2.2E-06	4.7E-09	2.4E-06	8.2E-09

Dietary Exposure Estimates

The projected dietary exposure estimates (DEE) based on the projected PFOS concentrations and regional dietary intake data are presented in Table 3.3. Consistent with the projected concentrations, there was also minimal difference between the DEEs of the two climate change scenarios and baseline. As the absolute differences between scenario concentrations were relatively small, when calculated with dietary intake values, the final exposures were largely within the same order of magnitude for each climate change scenario and baseline. The highest total PFOS exposure was from dried beluga meat consumption, followed by Arctic Char, beluga muktuk, and whitefish. While Arctic char was the country food consumed in the largest amounts (116 g/day compared to 15 g/day for the next country food), the PFOS exposure was still very low as Arctic char have much lower concentrations than beluga, by about one order of magnitude, due to their different trophic levels within the ecosystem. The highest exposure from country foods was expected to be from consuming dried beluga meat, but only by a small margin. This is reasonable, as the drying process leads to a much higher concentration compared to the other country foods which are not dried, but rather eaten either cooked or raw such as beluga muktuk. The DEEs here are comparable to previously published dietary exposure estimates for country food consumption in Nunavut (Ostertag, Tague, et al., 2009). These ranged from 2.0E-07 to 2.4E-06 mg/kg bw/day, using a conservative approach (Ostertag, Tague, et al., 2009). While it is not possible to directly compare the two DEEs as the estimates done in Nunavut were determined for a combination of market and country foods based on 24-hr recall, it is valuable to see the DEEs from modelled concentrations of PFOS in country foods here falling within the lower range of previously estimated dietary exposures. This also suggests that the increases in PFOS concentrations observed from climate change impacts on country foods likely have a negligible effect on dietary exposures.

Table 3.3. *Projected dietary exposure estimates and standard error mean (SEM) using mean projected PFOS concentrations under climate change scenarios and baseline.*

Country Food		Projected Dietary Exposure Estimates (mg/kg bw/day)					
		Baseline	**SEM**	**SSP1-2.6**	**SEM**	**SSP5-8.5**	**SEM**
Arctic char	meat	8.1E-07	1.54E-11	7.9E-07	4.78E-02	8.1E-07	1.07E-09
Whitefish	meat	1.5E-07	6.13E-12	1.4E-07	3.66E-03	1.4E-07	1.95E-10
Beluga	meat (dry)	8.2E-07	4.12E-11	1.0E-06	2.43E-02	1.1E-06	3.72E-09
	muktuk	2.9E-07	1.4E-11	3.5E-07	7.41E-10	3.7E-07	1.29E-09

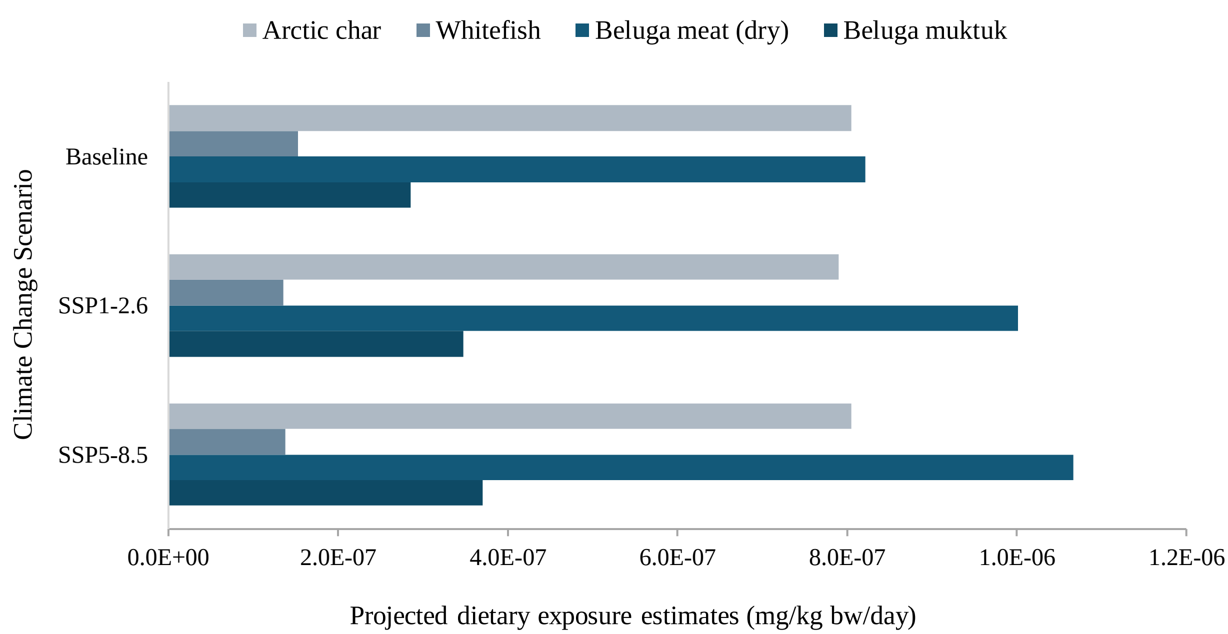


Figure 3.1 *Dietary exposure estimates of country foods under each climate change scenario.*

Hazard Quotients

The hazard quotients for each country food are presented in Table 3.4. All were well below 1, by 2-3 orders of magnitude, indicating that the PFOS exposure from all country foods evaluated here were well below the TDI value set by Health Canada. The HQs followed the same trend as exposure estimates, as expected. Given the very low HQs for each country food, it is unlikely that PFOS exposures from country foods as projected in 2100 will be a risk to health at the levels consumed in the early 2000s as reported in the IPY-IHS. A visualization of the difference between the exposure estimates for PFOS from each country food with the TDI is presented in Figure 3.2, showing the large gap between projected exposures by the end of the century and the current TDI value. This is consistent with previous dietary exposure and risk assessments for Inuit in Nunavut and the general Canadian population (Ostertag, Chan, et al., 2009; Ostertag, Tague, et al., 2009; Tittlemier et al., 2007). This is especially notable as these previous risk assessments were based on observed PFOS concentrations, and took place prior to the development of the current TDI value from Health Canada, therefore is suggestive that health risk from PFOS over time is not likely to differ to a degree that raises health concerns.

Table 3.4. *Projected hazard quotients (HQ) by 2100 using mean projected PFOS concentrations for the four highly-consumed country foods.*

Country Food		**Projected Hazard Quotients (TDI = 6.60E-05 mg/kg bw/day)**		
		Baseline	**SSP1-2.6**	**SSP5-8.5**
Arctic char	meat	1.2E-02	1.2E-02	1.2E-02
Whitefish	meat	2.3E-03	2.0E-03	2.1E-03
Beluga	meat (dry)	1.2E-02	1.5E-02	1.6E-02
	muktuk	4.3E-03	5.3E-03	5.6E-03

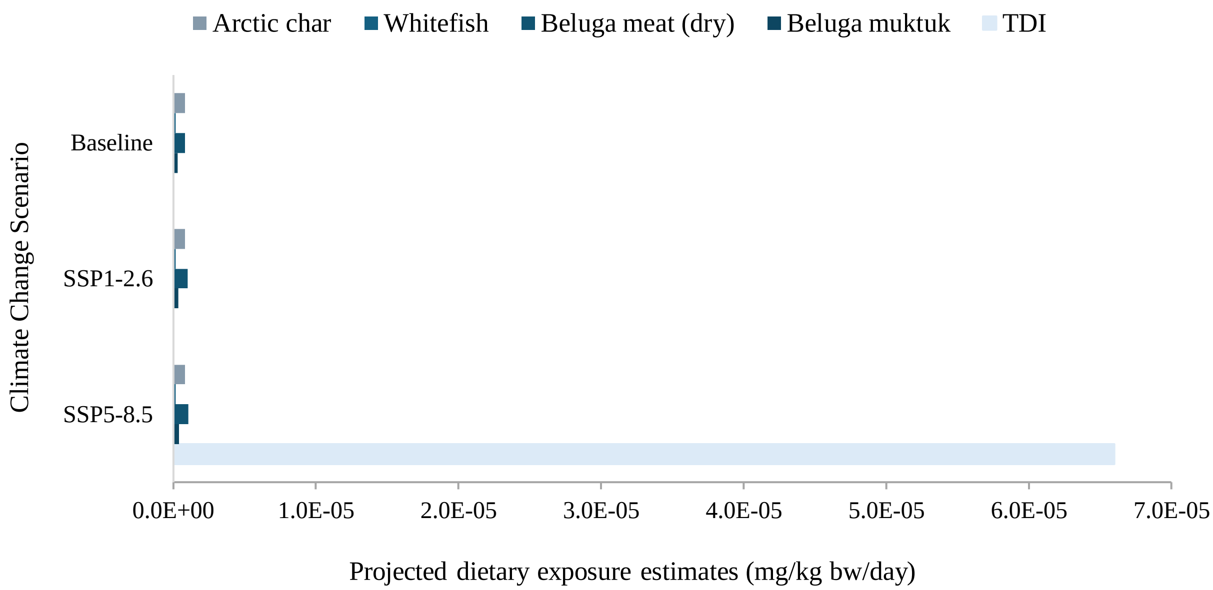


Figure 3.2 *Projected dietary exposure estimates of country foods under each climate change scenario relative to the Health Canada TDI for PFOS.*

To examine exposure estimates and HQ under higher consumption rates of each country food, DEEs and HQs were also calculated for each country food assuming 250g/day of intake, or a standard cup, to compare with the TDI values. Even with these consumption values, the HQs for each country food are still below 1. They may, however, be entering a level that could warrant monitoring for beluga, where the HQ values are less than one order of magnitude below 1. While 250g/day is likely an unrealistic amount of country food consumption for any of the four country foods evaluated, being 2-10 times higher than the reported consumption in the IPY-IHS, it is valuable to estimate whether the HQs are approaching 1 to warrant further investigation should there be any increases in country food consumption.

Table 3.5. *Projected DEE and HQ assuming 250 g/day consumption of each country food.*

Country Food		Projected Dietary Exposure Estimates (mg/kg bw/day)			Projected Hazard Quotients (TDI = 6.60E-05 mg/kg bw/day)		
		Baseline	**SSP1-2.6**	**SSP5-8.5**	**Baseline**	**SSP1-2.6**	**SSP5-8.5**
Arctic Char	meat	1.4E-06	1.7E-06	1.7E-06	0.02125	0.02508	0.02543
White fish	meat	1.1E-06	1.3E-06	1.3E-06	0.01713	0.01904	0.01928
Beluga	meat (dry)	1.3E-05	1.5E-05	1.6E-05	0.19062	0.22838	0.24661
	muktuk	6.3E-06	7.5E-06	8.1E-06	0.09510	0.11394	0.12304

It is worth noting how Health Canada's TDI values compare to other similar health-based values in different jurisdictions. Overall, Health Canada's TDI values for PFOS are higher and more conservative than those set by the United States, New Zealand, Australia, Denmark, and the European Union (EU). Only the United Kingdom has a higher TDI for PFOS, set by the Committee on Toxicity (COT) at 0.0003 mg/kg/day, which has not been updated since 2009 (Drinking Water Inspectorate, 2021). Food Standards Australia New Zealand (FSANZ) has set a TDI of 0.00002 mg/kg/day for PFOS (Wee & Aris, 2023). The European Food Safety Administration (EFSA) introduced a tolerable weekly intake equivalent to a TDI of 0.00000186 mg/kg/day for PFOS, however revised this in 2020 and proposed a tolerable weekly intake for four PFAS (PFOS, PFOA, PFHxS, and PFNA) equivalent to a TDI of 0.00000063 mg/kg/day (EFSA CONTAM Panel et al., 2020; EFSA Panel on Contaminants in the Food Chain et al., 2018; Wee & Aris, 2023). The US EPA recommends a reference dose of 0.00002 mg/kg/day for PFOS, while the Agency for Toxic Substances and Disease Registry (ATSDR), an agency under the CDC, in 2019 drafted a minimum risk level of 0.000002 mg/kg/day for PFOS (Wee & Aris, 2023). Health Canada's most conservative TDI, 0.00006 mg/kg/day, is higher but within the same order of magnitude as TDI or equivalents established by the Danish Environmental Protection Agency (DK EPA), US EPA, and FSANZ, but is 1 and 2 orders of magnitude higher than what is set by the EFSA and ATSDR, respectively.

Table 3.6. *Comparison of TDI and equivalent for PFOS set by other jurisdictions.*

Jurisdiction	Agency	TDI-Equivalent (mg/kg bw/day)	Year
Canada	HC[a]	0.0011 0.0001 0.00006	2018
United Kingdom	COT[b]	0.0003	2009
Denmark	DK EPA[c]	0.00003	2015
Australia, New Zealand	FSANZ[d]	0.00002	2018
United States	US EPA[d]	0.00002	2016
United States	CDC-ATSDR[d]	0.000002	2019
European Union	EFSA[e]	0.00000063	2020

Note. (Health Canada, 2018)[a], (Drinking Water Inspectorate, 2021)[b], (Larsen & Giovalle, 2015)[c], (Wee & Aris, 2023)[d], (EFSA CONTAM Panel et al., 2020)[e].

To assess how health risk may vary based on the most conservative HBV from other regions, the EU's EFSA HBV of 0.00000063 mg/kg bw/day was used to calculate HQs for each country food based on dietary exposure estimates by the end of the century. This results in HQs above 1 for two of the country foods; dried beluga meat (1.3-1.7) and Arctic char (1.3). The HQs for beluga muktuk and whitefish remain below 1, ranging from 0.45-0.59 and 0.21-0.24, respectively. While these HQs do suggest that there is a health risk associated with consuming beluga meat and Arctic char in the reported amounts, the risk should also be further characterized in the context of the dose-response relationships that t the toxicological reference values were derived.

The Health Canada TDI is based on adverse liver impacts of PFOS exposure alone, using hepatocellular hypertrophy as the critical effect based on studies done in rats and human-equivalent points of departure (Health Canada, 2018). The value is derived from the highest dose at which there were no observed adverse effects on the liver, and is generalized for the whole population (Health Canada, 2018). EFSA value is based on immunological effects of exposure to four PFAS, one of which is PFOS, specifically a depressed immune response to vaccinations in 12-month-old infants as the critical effect. A benchmark dose lower limit was determined using the combined blood serum levels of four PFAS associated with decreased antibody titres to vaccines (EFSA CONTAM Panel et al., 2020). This benchmark dose was then used to determine the corresponding daily dietary intake of the four PFAS by the mother, assuming the infant was breastfed for 12 months (EFSA CONTAM Panel et al., 2020). Thus, the EFSA HBV may have less uncertainty from the use of human studies and is more conservative as it is both based on multiple PFAS and on effects for the most vulnerable population (EFSA CONTAM Panel et al., 2020). Health Canada's TDI may have more uncertainty being based on animal-to-human extrapolation, and is less conservative being for only PFOS and not a vulnerable population (Health Canada, 2018).

As a low HBV for PFAS exposure, this EFSA value does result in a significant portion of the European population having exposure to the four PFAS well above the HBV. The mean reported dietary exposures to PFOS for adults across Europe range from 0.29-5.94 ng/kg bw/day (EFSA CONTAM Panel et al., 2020), while those calculated in this study for each ISR country food range from 0.14-1.08 ng/kg bw/day (whitefish, SSP1-2.6 and dried beluga meat, SSP5-8.5). As a theoretical maximum, the estimated combined exposure from all four country foods under SSP5-

8.5 is 2.38 ng/kg bw/day, which is below the European maximum of 5.94 ng/kg bw/day. It is important to note that these ranges are not directly comparable, as the exposure estimates for European adults reflect many food categories closer to the whole diet, and the exposure estimates for Inuvialuit reflect the individual country foods as there are regional variations in which are consumed frequently across the ISR (EFSA CONTAM Panel et al., 2020). However, it is valuable to compare exposures across these different populations subject to different HBVs, as it indicates that the projected PFOS dietary exposures from Inuvialuit country foods by the end of the century, even under climate change, are within the lower range of current, total dietary exposure to PFOS for European adults. PFOS concentrations in each country food are also comparable to the ranges of similar food types in Europe, such as livestock meat (0.028 – 0.17 ng/g), game meat (0.94 – 1.59 ng/g), and animal fats and vegetable oils (0.004 – 0.11 ng/g) comparable to beluga meat (0.63 ng/g) and beluga muktuk (1.8 ng/g), and fish such as cod and whiting (0.47-1.05 ng/g) and salmon and trout (0.31-0.83 ng/g), compared to Arctic char (0.41 ng/g) and whitefish (0.34 ng/g) (EFSA CONTAM Panel et al., 2020). The comparable dietary exposure estimates and PFOS concentrations in similar food types suggest that the health risks from PFOS exposure in country foods defined by the EFSA HBV are similar to those incurred by the general European population from dietary exposure to PFOS.

A 2023 study evaluated PFAS exposure for Inuit in Greenland from country food consumption using the EFSA HBV, providing an opportunity to compare the application of the EFSA HBV to Inuit country foods. The study reported that the average dietary exposure estimates for Inuit adults from the two country foods evaluated, polar bear and ringed seal meat, both exceeded the EFSA HBV (Sonne et al., 2023). The PFOS exposures for each were also within the same range as those in the ISR, polar bear meat (1.32 ng/kg bw/day) and ringed seal meat (0.68 ng/kg bw/day), though the exposure from polar bear meat is higher than any of the four ISR country food given how high PFOS concentrations in polar bears are relative to other marine mammals (Greaves et al., 2012; Sonne et al., 2023). Similar amounts of beluga meat and ringed seal meat are consumed in each respective location, and the projected PFOS concentration in beluga meat (0.89 ng/g ww) is also lower than what is reported in ringed seal meat (2.37 ng/g ww). However, beluga meat is consumed dried in the ISR; therefore, the dry weight concentration (3.64 ng/g dw) contributes to the higher dietary exposure than ringed seal (Sonne et al., 2023). The study does not report HQs, however applying the same methodology above using the dietary exposure estimates and EFSA HBV results in an HQ of 2.1 for polar bear meat and 1.1 for ringed seal meat, compared to the climate change scenario ranges of 1.3-1.7 for beluga meat and 1.3-1.3 for Arctic char. This study suggests that using the EFSA HBV, the health risks associated with country food consumption for Inuvialuit in the ISR are comparable to those associated with country food consumption for Inuit in Greenland, or slightly lower considering that country foods like beluga muktuk and whitefish with lower PFOS concentrations and HQs below 1 are consumed frequently in the ISR and country foods with high concentrations like polar bear are not.

Therefore, the dietary exposures to PFOS from consuming the four Inuvialuit country foods are comparable to exposures in the general European population and Greenlandic Inuit. The health risks from dietary exposure to PFOS are therefore also comparable between the general European population, Inuit in Greenland, and Inuvialuit in the ISR. However, overall Inuvialuit exposures are on the lower end, especially given that exposures from beluga muktuk and whitefish consumption do not exceed the EFSA HBV. These comparisons are useful for risk communication, as they indicate that the differing health risks as described by the HQs calculated using the HC HBV compared to the EFSA HBV are primarily a function of the evidence, methodologies, and risk tolerance reflected in the HBVs, rather than significant differences in PFOS exposures and health risks across different populations. As such, the HQs above 1 for beluga meat and Arctic char using the EFSA HBV should be considered within the context that a large part of the European population, as well as Inuit in Greenland, have comparable health risks associated with PFOS using the same HBV. Further research would be valuable to better understand how market food available in the ISR compares to country foods, in addition to monitoring future developments in guidance from Health Canada regarding PFOS health risk as more research emerges.

Limitations

There are several limitations associated with these results and the use of regional data and ecological modelling in conducting dietary exposure estimates. The data in the final report for the ISR represent averages from all six communities within the ISR and does not account for the unique communities' contexts and differences in country food consumption patterns. As a result, the dietary exposure estimates are for the ISR as a whole and cannot be assigned or attributed to any one community. As an example, the communities in the western part of the ISR are on the outer part of beluga territory, so beluga are not as frequently harvested compared to other communities or as other country foods like Arctic char, whereas in Inuvik and Tuktoyaktuk, beluga are harvested each fall and are therefore a more common staple in the diet for these communities, and residents consume them in higher amounts (Lea et al., 2023; Loseto et al., 2018; Ostertag et al., 2019). Additionally, there may be age and gender differences in country food consumption patterns, which are also not taken into account in the dietary exposure estimates.

The intention of these values was to assess potential future dietary exposures and hazard quotients to get a range of where exposures and risks may be under different climate change scenarios at a regional level, as information for planning purposes that may be used by Inuvialuit governance organizations such as the Inuvialuit Regional Corporation (IRC) or wildlife management bodies like the Inuvialuit Game Council and Fisheries Joint Management Council. The analysis here is valuable for these purposes; however, it cannot necessarily be applied to more specific contexts given the nature of the data used. This is important to be aware of when considering the meaning and applicability of results. With the second iteration of an Inuit Health Survey currently underway, there may be an opportunity for future studies to incorporate more refined dietary exposure analyses and risk assessments using the projected PFOS concentrations from the EwE model (*The Qanuippitaa? National Inuit Health Survey*, 2021).

Second, it is important to consider the limitations of modelling approaches when being used in the context of human dietary exposures and health risks. Models are very useful tools; however, they can never be perfect replicates of reality and therefore carry inherent limitations. In addition to the inherent uncertainty, models also include assumptions and constraints given the defined parameters being examined. These are discussed in depth in Chapter 2, but are revisited here in the context of dietary exposure estimates. The EwE model used here includes a number of assumptions for each different portion of the model. The toxicokinetic and environmental data available for PFOS is much more limited than other contaminants previously modelled with Ecotracer sub-routine in EwE, therefore many assumptions were made such as PFOS uptake rates of species for which there was no data available, and that PFOS concentrations would remain constant in the Beaufort seawater given the lack of data regarding PFOS transport and cycling in the environment. The climate change scenarios also only represent changes to sea surface temperature and sea ice cover, and do not include other relevant climate change factors like seawater salinity, pH, or oxygen concentration (Sora et al., 2022, 2024). The EwE approach also does not consider geographical changes to the ecosystem from climate change, such as changes in habitat range or foraging area, which could be taken into account by using the Ecospace routine in the model (Hoover et al., 2021). Given recent observations of beluga in Ulukhaktok, which was historically outside of the beluga range within the Beaufort region, integration of Ecospace may be of much greater importance to capture the climate change impacts on this ecosystem than previously imagined (Loseto et al., 2018). Lastly, the EwE model was built using data from published literature, which reflects almost exclusively the state of Western scientific knowledge on the BSS ecosystem. Including Inuvialuit knowledge in the model itself and the scenarios would be incredibly value and important to reflect the ecosystem structure, function, and changes over time in the model in a way that Western science cannot capture. Should this be an opportunity to co-develop with Inuvialuit to include in one or any of the Ecopath, Ecosim, or Ecospace model components, it would greatly strengthen the model and could address data gaps within Western scientific literature.

3.5 Conclusion

PFOS is a persistent organic pollutant that is found across the Arctic, including the Beaufort Sea, which has contributed to dietary exposures for Inuvialuit from consuming country foods. Here, it was found that climate change is unlikely to cause meaningful increases in PFOS exposure via country food consumption in the Inuvialuit Settlement Region by the end of the century, despite small increases in PFOS concentrations in marine species. The dietary exposures estimated from the modelled PFOS concentrations in marine country foods were comparable to those in Nunavut for both country and market foods in the early 2000s, as well as to Europeans in the 2020s (EFSA CONTAM Panel et al., 2020; Ostertag, Chan, et al., 2009). The hazard quotients based on Health

Canada’s TDI for PFOS indicate that the dietary exposure estimates are well below the limit for preventing adverse health effects, suggesting the future health concerns from PFOS dietary exposures via country foods is low. However, given the limitations of a modelling approach to estimate PFOS concentrations in marine species, regional-level dietary data, and limitations in Health Canada’s TDI, more detailed dietary analyses, measures of internal PFOS doses (i.e. blood serum concentrations) and continued marine wildlife biomonitoring would be valuable to more thoroughly assess likely future health risks from PFOS in marine country foods. Continued research to better understand the sources and transport of PFOS to the Arctic, the toxicokinetics of PFOS, and more specific and up-to-date dietary intake data for marine country food consumption in the Inuvialuit Settlement Region would improve the certainty of results in similar future studies.

CHAPTER 4. CONCLUSION

4.1 Results Sharing and Risk Communications

An important portion of this study involves sharing the results with the local communities within the Inuvialuit Settlement Region. As described in Chapter 1, a research license was obtained from the Aurora Research Institute. Through the application process, the project description was shared with various regional and local governance organizations, including the IRC, HTCs for each of the six communities, and territorial government As such, these groups will be contacted again to share results of the study.

To do this, a one-page research summary was created to summarize the project, findings, and implications for Inuvialuit communities (Figure A-3). The summary was created using plain-language communications principles, specifically tailored to the intended audience. The intention was to balance the level of detail, avoiding technical language and details given the non-scientific audience, while also recognizing the extensive knowledge that harvesters have about the land and ecosystems, which is greater than non-Inuvialuit researchers from the south. Therefore, the aim was to present the research in a way that was accessible to individuals not as non-experts in western scientific methods, but who are experts in the ecosystem and country foods at the heart of the research. Any further information about the research that may be requested, such as this thesis or future publications, will be shared as well with the government(s) or organization(s) interested.

At the center of the results sharing was risk communication. This builds on the historical context of contaminants research in the North and the implications for communities. In the past, there have been instances where researchers have made missteps in their results communications to communities. The lessons learned within the field as well as the nature of this research informed the strategy for communicating results in a way that is clear about the purpose of the study, the results, the limitations of it, and the implications. Front of mind was the PFAS as a contaminant class, which are currently receiving attention and concern within both scientific and general media landscapes, the use of a modelling approach which fills a different role within this field of research compared to empirical or field studies like biomonitoring, and the use of modelling projections in a health risk context with country foods. The research summary aimed to communicate the results in a way that aimed to answer the questions: why did we do this study, what did we do, what did we find, and what does this mean? Key points were to share that modelling is a tool to make predictions about possible scenarios in the future, rather than a tool that can tell us with certainty about what will happen. As such, the value in this study is to get a sense of what the future might look like for PFOS in country foods and health risks of that based on the knowledge we have in the present. The second key point was to highlight that the results suggest the potential future risk from PFOS in country foods is very low, so even though the language centers around language of 'health risk', that this does not mean the risk is automatically high, just that it is being evaluated. Ultimately, the goal is to share the results with communities such that they are empowered with more information and their own understanding of the research on contaminants and country foods.

This approach was done with guidance from and in partnership with the Country Foods for Good Health project. This study is a large collaboration between many researchers and Inuvialuit communities to conduct community-based biomonitoring work of contaminants in country foods, among them PFOS, and to understand the results and risk communication preferences of communities so that it can be designed with the specific circumstances and needs of each community in mind.

Upon submission of this thesis, a 6-month embargo will be placed on its publication to the university site, to leave time for communities to receive the results and reach out with any further questions, if relevant or desired. Many communities are very small and receive a high volume of information from not only researchers but also government and non-government entities. Therefore, extra time is intended to be mindful of those capacity limitations, with the knowledge that communities may not be able to review results for a few months after they are shared. Manuscripts submitted for publication will also be delayed for the same purpose, ultimately to make the best effort to ensure that communities have the chance to see the results before they are shared publicly.

4.2 Implications

This research can be valuable for informing a variety of future research, policy, and community-based work in the future. With results showing climate change-driven increases in PFOS in Beaufort Sea species, the research has the potential to inform the direction of community-based biomonitoring work in the ISR. This could involve continuing monitoring efforts, or adapting the focus of biomonitoring to various species based on the modelling results and needs of the communities as time goes on. The limitations highlighted in the model parameterization also provide insight into data gaps for modelling purposes, which may be valuable to guide future monitoring studies.

The use of Ecotracer output from EwE also presents a unique opportunity for informing policy around country food consumption guidelines. The ability to integrate both trophodynamic and environmental drivers of ecosystems in mapping contaminant movement through a food web is incredibly valuable. As more information becomes available on country food consumption from the second iteration of the Inuit Health Survey, further information about climate change, and more research on the health effects of PFOS exposure via diet, this type of modelling approach may be a valuable tool for evaluating the climate change-contaminant relationship at a regional level as it relates to dietary exposure to contaminants and potential risks from country food consumption.

The results from this current research provide insight into the risk of PFOS in the future given the environmental concentrations, climate policy, and country food consumption around the year 2010. These results indicate that the estimated exposures by the end of the century are within the acceptable range based on current guidance from Health Canada. This is a positive finding given the importance of country foods for Inuvialuit, and sharing this research and future insight will hopefully provide communities with assurance on country food consumption now and into the future. Comparison to other jurisdictions also highlights the importance of revisiting these results as guidance around PFOS exposure changes in the future with emerging evidence. This may inform decisions by policymakers and communities within the ISR on not only country food harvesting and consumption planning, but also climate change adaptation, as this research aligns directly with two of the six thematic areas outlined in the ISR Climate Change Strategy: food and wellness, and ecosystem health and diversity (Inuvialuit Regional Corporation, 2021). Ultimately, the aim is to increase the availability of information about climate change, country foods, and contaminants within the ISR to empower decision-makers in the region to make informed plans for the future.

Contribution to the Field

This research offers several novel perspectives on ecological modelling, ecotoxicology, and environmental health. As PFAS have never been modelled using the EwE framework, this study provides a basis for further work using EwE for this class of compounds. It also provides novel insight into the climate change-food web-contaminant dynamic with PFOS, which has not yet been explored in any PFAS and cannot be inferred from existing knowledge on other POPs due to the more hydrophilic properties of PFAS and subsequent difference in toxicokinetics. Lastly, this study is unique in its direct use of EwE modelling output for human health risk assessment. EwE is commonly used in the context of fisheries management and marine protection, with one prior study directly studying impacts of climate change on mercury exposure and health risk in the Faroe Islands (Booth & Zeller, 2005). This study provides further insight into the application of EwE in this context, as well as the application to both the Canadian Arctic and PFOS as a contaminant. It also aligns with ongoing research projects, modelling contaminants in the Beaufort Sea Shelf ecosystem and community-led biomonitoring for many contaminants including PFOS, adding linkages between biomonitoring and modelling as methodological approaches to contaminant and health research.

4.3 Limitations and Future Directions

The full limitations associated with the modelling approach and use of model output for dietary exposure estimates of PFOS are discussed in full detail in each respective chapter. Overall, the primary limitations of this study arise from the modelling approach and the inherent uncertainties, parameter limitations, data limitations and assumptions made in building an ecosystem model. The EwE modelling framework does allow for the concurrent evaluation of multiple drivers on an ecosystem which is a great strength of this approach. However, it remains constrained by limitations in data availability, which impact the Ecopath, Ecosim, and Ecotracer routines, as discussed previously, as well as the extent of the modelling capabilities that can be employed in just one study. Many other routines within the framework, such as Ecospace and Ecosampler, could be employed to address the uncertainties associated with the lack of geographic factors and to better characterize the uncertainty that is inherent to the model. The climate change scenarios could be expanded to include more climate drivers such as salinity and oxygenation to improve the ability of the model to reflect the environment within set parameters. The Ecotracer parameterizing could be substantially improved with more toxicokinetic and environmental data as it emerges, particularly regarding the role of precursors in PFOS partitioning in the environment and biotransformation, the role of the Mackenzie River in transporting PFOS to the BSS, and the role of PFOS uptake from sediment. Further, integration of local Inuvialuit knowledge of the ecosystem and species within it into the Ecopath model, Ecosim climate change scenarios, and if used the Ecospace subroutine would be valuable to address the many limitations of constructing such models using published data, which reflects primarily Western scientific knowledge on the ecosystem and misses the knowledge held by land-users on both the nature of the Beaufort Sea ecosystem and how it is changing. This work builds on a wealth of earlier modelling work within the BSS to continuously update the Ecopath base model itself, characterize the impacts of environmental and climate drivers on trophodynamics, and in using Ecotracer to map mercury within the BSS ecosystem (Gillies et al., 2024; Hoover et al., 2021, 2022; Li et al., 2022; Sora et al., 2022, 2024; Steiner et al., 2019). This extensive work contributing to the EwE modelling suite in the BSS presents many opportunities for future directions in this work, both for contaminants as well as ecosystem dynamics in the face of climate change impacts.

The use of IPY-IHS dietary data also presents a limitation in the context of projecting future dietary exposure estimates, given the likely changes in country food consumption patterns that can be impacted by both climate change, such as changes in harvesting, and other external factors, like changes to community demographics or access to country foods, which can all influence diet and are not captured by the methods used here. There is potential here for integration of both more up-to-date data with the second iteration of the Inuit Health Survey underway, which will provide current data, as well as a second data set to make comparisons on changes over time to diet within the ISR at a population level. It is also possible that future studies may use different scenarios on country food consumption based on what is anticipated by communities based on their local context and knowledge of opportunities, which may also present the potential for community-level data rather than or in addition to a regional level.

Overall, the first use of EwE in evaluating future climate change-contaminant-food web interactions for PFOS presents many opportunities for future directions to better understand the impacts of these changes and implications for human health in the Beaufort Sea region and surrounding Inuvialuit communities.

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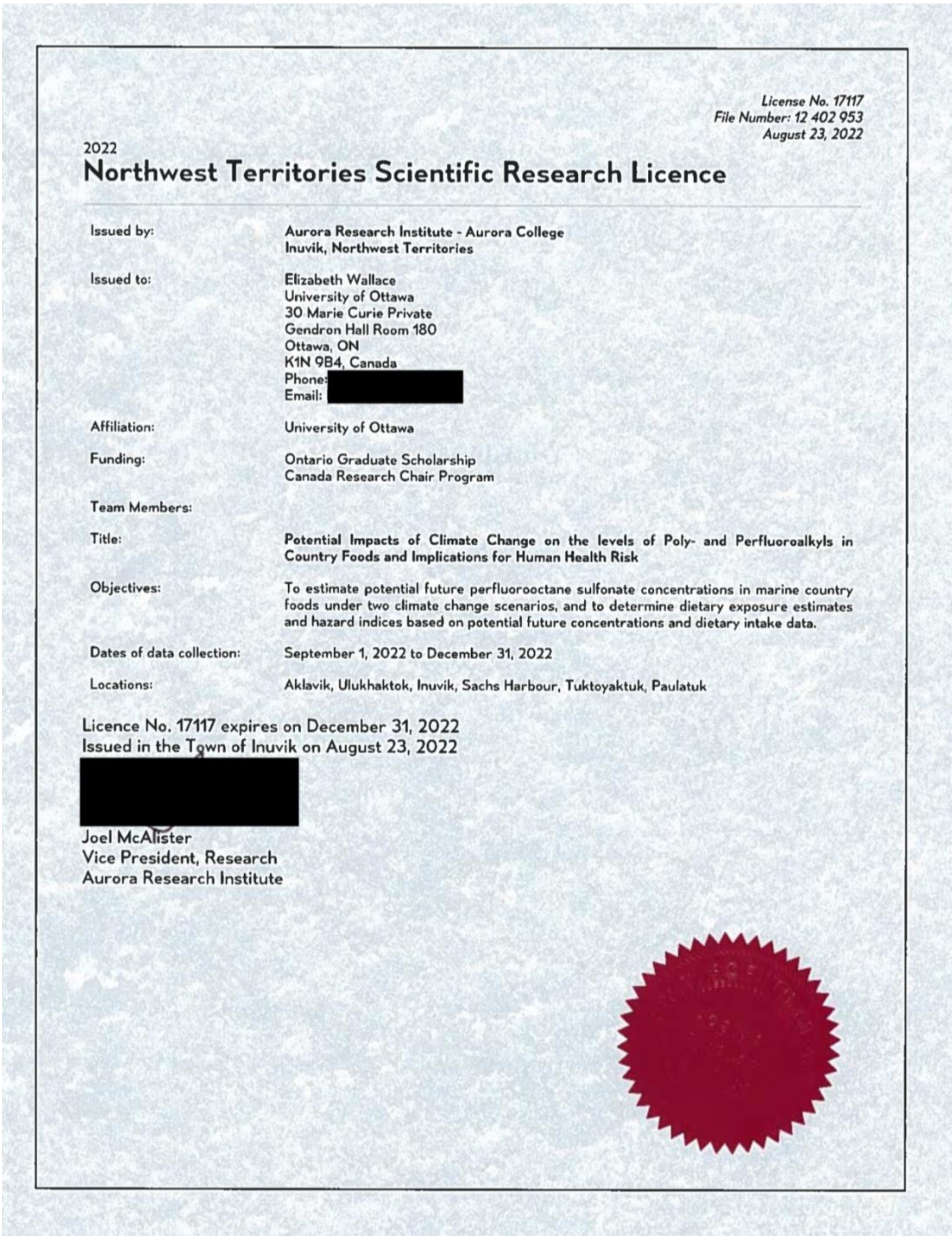
APPENDICES

Appendix A: Research License and Community Engagement



License No. 17117
File Number: 12 402 953
August 23, 2022

2022
Northwest Territories Scientific Research Licence

Issued by:	Aurora Research Institute - Aurora College Inuvik, Northwest Territories
Issued to:	Elizabeth Wallace University of Ottawa 30 Marie Curie Private Gendron Hall Room 180 Ottawa, ON K1N 9B4, Canada Phone: [redacted] Email:
Affiliation:	University of Ottawa
Funding:	Ontario Graduate Scholarship Canada Research Chair Program
Team Members:	
Title:	Potential Impacts of Climate Change on the levels of Poly- and Perfluoroalkyls in Country Foods and Implications for Human Health Risk
Objectives:	To estimate potential future perfluorooctane sulfonate concentrations in marine country foods under two climate change scenarios, and to determine dietary exposure estimates and hazard indices based on potential future concentrations and dietary intake data.
Dates of data collection:	September 1, 2022 to December 31, 2022
Locations:	Aklavik, Ulukhaktok, Inuvik, Sachs Harbour, Tuktoyaktuk, Paulatuk

Licence No. 17117 expires on December 31, 2022
Issued in the Town of Inuvik on August 23, 2022

[redacted]

Joel McAlister
Vice President, Research
Aurora Research Institute

Figure A-1. *Research License from the Aurora Research Institute.*

INUVIALUIT
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May 25, 2022

Elizabeth Wallace, MSc Candidate
Dr. Laurie Chan
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30 Marie-Curie Private, Gendron Hall Room 180
Ottawa, ON., K1N 9B4

Re: Letter of Support — "Potential Impacts of Climate Change on the levels of Poly- and Perfluoroalkyls in Country Foods and Implications for Human Health Risk".

The Inuvialuit Regional Corporation (IRC) was established with the overall responsibility of managing the affairs of the Western Arctic Claims Settlement Act as outlined in the Inuvialuit Final Agreement (IFA) of 1984. The IFA provided the Inuvialuit with ownership of 91,000 square kilometers (35,000 square miles) of land including 13,000 square kilometers (5,000 square miles) with subsurface rights to oil, gas, and minerals. These lands must be continually monitored amidst changes due to climate, development, contaminants, and land use.

The mandate of the IRC is to continually improve the economic, social, and cultural well-being of the Inuvialuit through implementation of the IFA and by all other available means. The IFA contains three principles expressed by the Inuvialuit and recognized by Canada: (a) to preserve Inuvialuit cultural identity and values within a changing northern society; (b) to enable Inuvialuit to be equal and meaningful participants in the northern and national economy and society; and (c) to protect and preserve the Arctic wildlife, environment, and biological productivity.

Legacy Persistent Organic Pollutants (POPs), formerly produced in industrialized regions of the world, are rapidly impacting the Inuvialuit lands through atmospheric, oceanic, and river system transportation. Future effects of climate change on perfluorooctane sulfonate (PFOS)- a type of POP- in food webs and the implication for human health is not well understood. Currently, these contaminants accumulate and bio-magnify in Arctic food webs, reaching the greatest concentrations in top-of-the-food chain marine mammals. Recognizing the threat of POPs, the IRC continues to maintain focus on contaminants through outreach and engagement initiatives with the Inuit Research Advisor (IRA), and the Northern Contaminants Program (NCP)'s Northwest Territory Regional Contaminants Program (NWTRCC).

The IRC is issuing this Letter of Support to the University of Ottawa, for the proposed project "Potential Impacts of Climate Change on the levels of Poly- and Perfluoroalkyls in Country Foods and Implications for

Human Health Risk". A research plan and proposal have been thoroughly reviewed, and the IRC sees the value of this project.

The action from the University of Ottawa will contribute to the knowledge mobilization, transparent communication, and education outcomes of the Inuvialuit strategies by raising regional awareness of potential future contaminant impacts in the Inuvialuit Settlement Region (ISR). The University of Ottawa will also strengthen engagement and participation by including Inuvialuit knowledge holders in the ISR to validate their contaminants research findings. The project will contribute to the principles of the IFA by providing Inuvialuit pertinent information that focuses on the protection and preservation of the Arctic wildlife and environment through knowledge transfer on a local and regional scale.

The overall outcome of the "Potential Impacts of Climate Change on the levels of Poly- and Perfluoroalkyls in Country Foods and Implications for Human Health Risk", will be to address the potential effects of climate change on PFOS in the Beaufort Sea food web and the implications for human health risk. The objective of this project is to provide information on the potential impacts of climate change on contaminants in country foods to enable communities to make informed health decisions and climate change adaptation plans. The researchers will study the effects of potential future PFOS levels on country foods in the Beaufort Sea under two climate change scenarios; and will determine potential future dietary exposure estimates and hazard indices under these climate change scenarios. Through partnerships, we hope to promote more action and support to collaborative work on contaminants research in our region and across the Canadian Arctic.

Sincerely, [redacted]

Bob Simpson
Director, Government Affairs
Inuvialuit Regional Corporation

Figure A-2. *Letter of support from the IRC included in the ARI research license application.*

Climate Change Impacts on PFOS in the Beaufort Sea

Key Messages

- PFOS levels in Beaufort Sea species will likely increase in the future because of climate change
- These increases in PFOS levels are very small
- The increases are so small there is minimal change to PFOS exposure from country foods
- The future health risk from PFOS in country foods is very low

Research Team

Elizabeth Wallace, Master's student
Dr. Laurie Chan, Professor

University of Ottawa

Contact
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Why do this study?

- PFOS is a type of environmental contaminant known as a 'forever chemical'. It is a concern because it can cause negative health effects and does not break down easily in the environment.
- PFOS production has decreased to almost zero in the past 20 years because of international regulation. Despite, it is still present in the environment, including the Arctic, because it does not break down.
- Climate change can impact levels of contaminants, like PFOS, in marine species.
- It can do this by changing marine food webs, like the type or amount of a food source for different species.
- We wanted to study these changes to explore impacts to PFOS levels in Beaufort Sea country food species.

What did we find?

We modelled future climate changes and PFOS levels in the Beaufort Sea from 2010-2100. **We found that climate change impacts to the Beaufort Sea food web will likely cause small increases to PFOS levels in Beaufort Sea species by 2100, compared to baseline (no climate change).**

We used two scenarios, low and high, to compare different levels of future climate change. **We found that PFOS levels are similar between the low and high climate change scenarios by 2100, and both scenarios cause small increases compared to baseline (no climate change).**

We used the modelled PFOS levels to estimate possible future PFOS exposures from 4 country foods in the ISR: Beluga muktuk, dried beluga meat, Arctic char, and whitefish. **The estimated PFOS exposures from all 4 country foods increased a small amount due to climate change, between 10% to 30%, compared to baseline (no climate change).**

We compared the estimated PFOS exposures from each country food to Health Canada's exposure limit, to assess the possible health risks in the future. **The estimated PFOS exposures for each country food were much lower than Health Canada's limit (~100 times lower), for both climate change scenarios and baseline (no climate change).**

It is important to note that our results are only possible future outcomes. Models are not perfect replicates of reality, so while they are useful tools to consider what might happen in the future based on the information we have now, they do not provide absolute predictions.

What do these results mean?

Our results suggest that future health risk from PFOS in country foods is low. Even though climate change will likely cause small increases to PFOS levels in Beaufort Sea species, these increases are small enough to not cause a meaningful change in PFOS exposure from country foods.

Figure A-3. *Copy of Research Summary.*

Appendix B: EwE Input

Table B-1. *Mean temperature (thermal) tolerance (MTT) and standard deviation (SD) in for marine fish species and invertebrates included in the BSS food web.* Retrieved from (Alava et al., 2018).

Functional group	**Latin name**	**Common name**	**MTT (°C)**	**SD**
Char & Dolly Varden	*Salvelinus alpinus*	Arctic Charr	1.51	2.69
Cisco & Whitefish	*Coregonus lavaretus*	Common whitefish	9.96	2.64
Salmonids	*Salmo salar*	Atlantic salmon	8.69	4.27
	Oncorhynchus gorbuscha	Pink salmon	5.36	4.11
	Oncorhynchus keta	Chum salmon	5.57	3.32
	Oncorhynchus nerka	Sockeye salmon	4.79	2.44
	Oncorhynchus tshawytscha	Chinook salmon	6.04	3.61
	Oncorhynchus kisutch	Coho salmon	4.77	2.46
Herring & Smelt	*Clupea pallasii*	Pacific herring	7.43	4.52
	Ammodytes personatus	Pacific sand lance	12.8	4.24
	Osmerus mordax	Atlantic rainbow smelt	2.99	3.05
Arctic & Polar Cod	*Boreogadus saida*	Polar cod	0.85	2.50
Capelin	*Mallotus villosus*	Capelin	3.25	3.37
Flounder & Benthic Cod	*Gadus ogac*	Greenland cod	5.56	5.61
	Eleginus gracilis	Saffron cod	5.04	2.95
	Platichthys stellatus	Starry flounder	4.55	4.22
Small Benthic Marine Fish	*Gobius niger*	Black goby	11.2	7.49
Other Fish	*Gasterosteus aculeatus*	Three-spined stickleback	11.1	8.41
Invertebrates	*Petromyzon marinus*	Sea lamprey	11.1	7.01
	Pandalus borealis	Northern prawn	5.41	4.37
	Chionoecetes opilio	Snow crab/Queen crab	4.04	2.41
	Illex illecebrosus	Northern shortfin squid	14.9	8.13

Table B-2. *GFDL-ESM4 sea surface temperature and sea ice cover projections for SSP1-2.6 and SSP5-8.5 in the study region from 2010-2100.* Retrieved from (Dunne et al., 2020).

	SSP1-2.6		**SSP5-8.5**	
Year	SST °C	SIC %	SST °C	SIC %
2000	-1.239	87.16	-1.239	87.16
2001	-0.537	77.08	-0.537	77.08
2002	-1.155	86.66	-1.155	86.66
2003	-1.321	88.49	-1.321	88.49
2004	-0.917	83.18	-0.917	83.18
2005	-1.413	91.91	-1.413	91.91
2006	-1.148	86.57	-1.148	86.57
2007	-1.295	89.50	-1.295	89.50
2008	-0.987	84.67	-0.987	84.67
2009	-1.408	89.93	-1.408	89.93
2010	-1.067	85.08	-1.067	85.08
2011	-1.011	85.04	-1.011	85.04
2012	-1.079	86.25	-1.079	86.25
2013	-1.025	82.37	-1.025	82.37
2014	-0.730	82.75	-0.730	82.75
2015	-1.028	83.85	-0.805	82.39
2016	-1.170	87.39	-0.966	81.91
2017	-1.139	85.63	-0.717	79.94
2018	-1.546	93.83	-1.011	85.33

2019	-0.695	81.35	-1.015	85.99
2020	-0.863	83.91	-1.398	90.84
2021	-0.615	78.97	-0.983	84.76
2022	-0.921	86.01	-1.208	86.72
2023	-1.286	87.68	-0.898	80.63
2024	-0.542	77.62	-1.317	89.51
2025	-1.167	86.72	-0.969	83.79
2026	-0.863	83.61	-0.881	85.68
2027	-0.883	79.66	-0.944	83.32
2028	-1.098	85.46	-1.251	87.69
2029	-1.047	85.21	-1.108	87.66
2030	-0.921	83.96	-0.623	79.78
2031	-1.036	86.18	-0.387	75.59
2032	-0.892	82.27	-1.045	82.58
2033	-0.981	82.61	-0.599	79.86
2034	-0.529	78.83	-0.583	79.00
2035	-0.852	81.94	-0.754	81.24
2036	-0.829	82.37	-0.894	84.11
2037	-0.331	77.38	-1.088	87.29
2038	-1.001	80.34	-0.952	82.20
2039	-0.444	75.11	-1.022	83.86
2040	-1.090	85.79	-0.842	82.01

2041	-0.642	79.48	-0.493	77.84
2042	-0.511	76.68	-1.087	85.24
2043	-0.967	82.35	-0.782	78.31
2044	-0.738	78.76	-0.339	76.33
2045	-0.748	79.73	-0.402	73.22
2046	-1.248	88.35	-0.550	77.98
2047	-1.093	87.36	-0.916	79.19
2048	-0.828	81.09	-0.769	76.40
2049	-1.050	85.64	-0.602	76.35
2050	-1.032	85.10	-0.364	73.25
2051	-0.408	76.40	-0.148	68.43
2052	-0.221	72.97	-0.657	74.03
2053	-0.650	76.85	-0.633	75.65
2054	-1.031	83.42	-0.435	69.75
2055	-1.051	86.51	0.011	67.03
2056	-1.048	85.64	-1.241	84.71
2057	-0.040	68.93	-0.189	69.85
2058	-1.049	85.29	-0.600	76.94
2059	-1.134	87.48	-0.532	73.39
2060	-1.079	84.69	-0.849	78.87
2061	-0.654	73.37	-0.280	67.59
2062	-0.374	76.61	-0.221	66.74

2063	-0.509	76.45	-1.108	81.72
2064	-0.953	81.33	-0.098	66.56
2065	-1.171	87.90	-0.371	70.09
2066	-0.715	77.71	-0.493	69.15
2067	-0.952	83.00	-0.259	67.20
2068	-0.815	78.50	-0.123	66.36
2069	-0.956	80.75	-0.045	65.55
2070	-0.852	82.76	0.426	59.07
2071	-0.663	80.26	-0.144	62.94
2072	-0.340	74.69	-0.345	68.26
2073	-0.487	74.80	-0.033	64.51
2074	-0.743	80.92	0.191	61.18
2075	-0.803	81.26	0.113	60.55
2076	-0.940	83.60	-0.182	65.87
2077	-1.167	86.91	0.168	61.59
2078	-0.598	77.21	0.240	59.69
2079	-0.170	74.39	0.039	60.32
2080	0.094	71.08	0.214	61.79
2081	-0.854	81.75	-0.296	66.82
2082	-0.443	74.64	0.248	56.85
2083	-0.302	73.06	0.210	56.51
2084	-1.067	83.13	0.518	54.65

2085	-0.836	80.90	0.456	57.03
2086	-1.263	88.06	0.230	58.19
2087	-0.282	72.48	0.094	56.80
2088	-0.658	79.63	0.401	54.93
2089	-0.320	72.66	0.482	52.48
2090	-0.731	78.76	0.339	53.48
2091	-0.698	76.78	1.163	49.00
2092	-0.501	76.84	0.161	52.67
2093	-0.461	73.58	0.209	57.37
2094	-1.120	85.50	0.596	55.50
2095	-0.769	78.16	0.831	51.73
2096	-0.708	80.14	0.446	56.81
2097	-0.657	76.82	-0.010	59.49
2098	-0.174	69.46	0.739	53.44
2099	-0.493	75.85	0.807	47.95
2100	-0.786	79.18	0.494	53.04