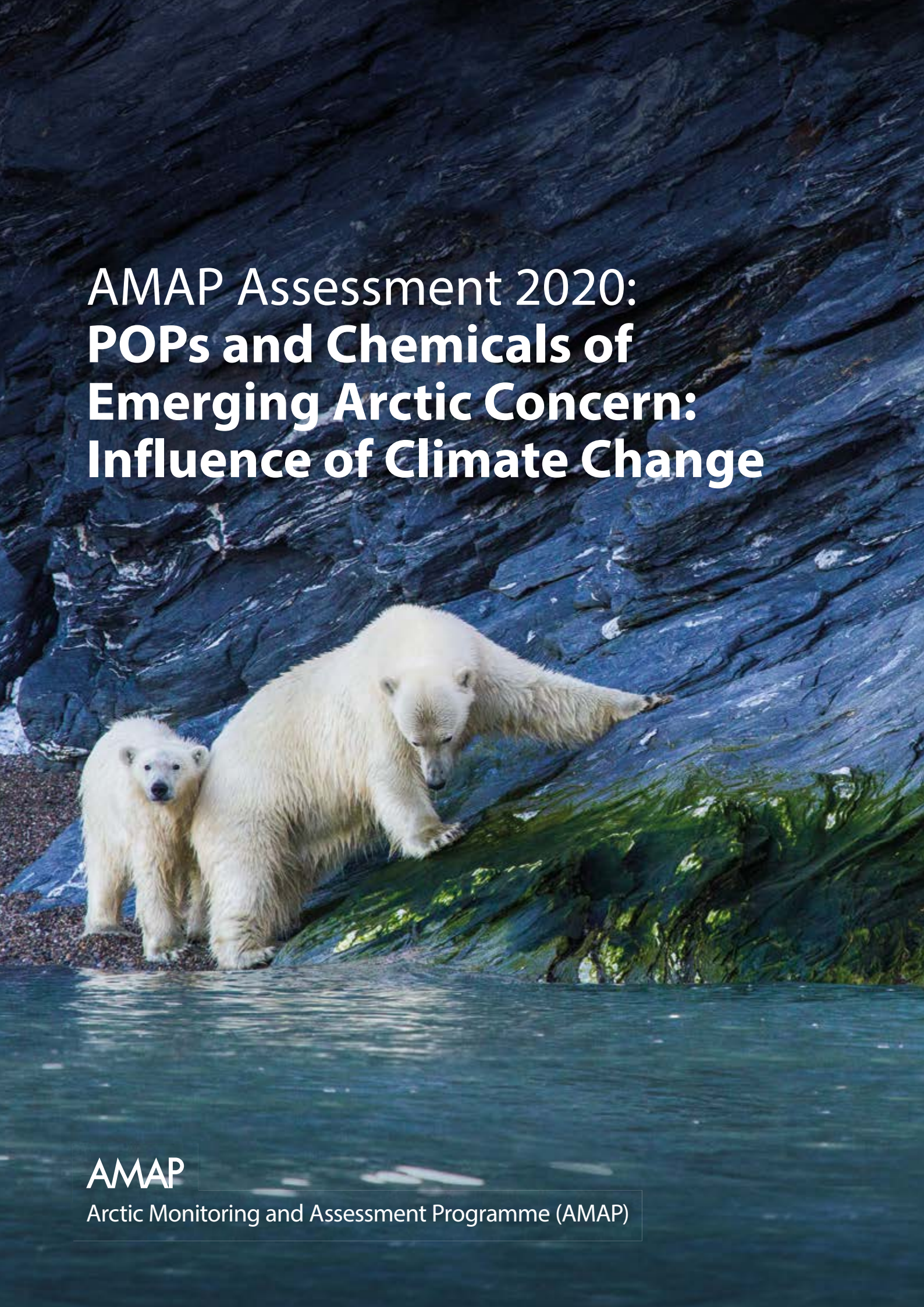

AMAP Assessment 2020:
POPs and Chemicals of Emerging Arctic Concern: Influence of Climate Change

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Arctic Monitoring and Assessment Programme (AMAP)

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Contents

Preface

This assessment report presents the results of the 2020 AMAP Assessment of POPs and Chemicals of Emerging Arctic Concern: Influence of Climate Change. The assessment updates information presented in earlier AMAP assessments delivered in 2002 and 2009, and a joint report prepared with UNEP in 2011.

The Arctic Monitoring and Assessment Programme (AMAP) is a Working Group of the Arctic Council. The Arctic Council Ministers have requested AMAP to:

- produce integrated assessment reports on the status and trends of Arctic ecosystems;
- identify possible causes for the changing conditions;
- detect emerging problems, their possible causes, and the potential risk to Arctic ecosystems including Indigenous Peoples and other Arctic residents;
- recommend actions required to reduce risks to Arctic ecosystems.

This report provides the accessible scientific basis and validation for any statements and recommendations made in related derivative products, including its summary for policy-makers that was delivered to the Arctic Council Ministers at their meeting in 2021.

The present report includes extensive background data and references to scientific literature, and details the sources for graphics reproduced in summary products. Whereas the related summary for policy-makers contains recommendations that focus on policy-relevant actions, the conclusions and recommendations presented in this report also cover issues of a more scientific nature, such as proposals for filling gaps in knowledge, and recommendations relevant to future monitoring and research work.

This assessment of the influence of climate change on POPs and chemicals of emerging Arctic concern (CEACs) was conducted between 2019 and 2020 by an international group of experts. The expert group members and lead authors were appointed following an open nomination process coordinated by AMAP. A similar process was used to select international experts who independently reviewed this report. Information contained in this report is fully referenced and based first and foremost on results of research and monitoring undertaken since 2010. It incorporates some new (unpublished) information from monitoring and research conducted according to well-established and documented national and international standards and quality assurance/ quality control protocols. Care was taken to ensure that any critical probability statements made in this assessment were based only on peer-reviewed materials. Access to reliable and up-to-date information is essential for the development of science-based decision-making regarding ongoing changes in the Arctic and their global implications.

The assessment lead authors have confirmed that both this report and its derivative products accurately and fully reflect their scientific assessment. All AMAP assessment reports are freely available from the AMAP Secretariat and on the AMAP website: www.amap.no, and their use for educational purposes is encouraged.

AMAP would like to express its appreciation to all experts who have contributed their time, efforts and data, in particular the lead authors who coordinated the production of this report. Thanks are also due to Jennifer Balmer for her work as Scientific Secretary to the assessment and the reviewers who contributed to the assessment peer-review process and provided valuable comments that helped to ensure the quality of the report. A list of contributors is included in the acknowledgments at the start of this report and lead authors are identified at the start of each chapter. The acknowledgments list is not comprehensive. Specifically, it does not include the many national institutes, laboratories and organizations, and their staff that have been involved in contaminants-related monitoring and research. Apologies, and no lesser thanks are given to any individuals unintentionally omitted from the list.

The support from the Arctic and non-Arctic countries implementing research and monitoring in the Arctic is vital to the success of AMAP. AMAP work is heavily based on ongoing activities of these countries, and the countries that provide the necessary support for the experts involved in the preparation of AMAP assessments. In particular, AMAP would like to acknowledge Canada, the Kingdom of Denmark and Sweden for their lead role in this assessment, and thank Canada, the Kingdom of Denmark, Norway (Ministry of Foreign Affairs) for their financial support of this assessment work.

The AMAP Working Group is pleased to present its assessment to the Arctic Council and the international science community.

Cynthia de Wit (Assessment co-lead, Sweden)

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Katrin Vorkamp (Assessment co-lead, Denmark)

Ben deAngelo (AMAP Chair)

Rolf Rødven (AMAP Executive Secretary)

Tromsø, July 2021

1. Introduction

Authors: Cynthia de Wit, Derek Muir, Katrin Vorkamp

The Arctic Monitoring and Assessment Programme (AMAP)[1] was established in 1991 as an international program for monitoring and assessing Arctic pollution under the Arctic Environmental Protection Strategy (AEPS, 1991). It is now a Working Group of the Arctic Council[2] and is responsible for monitoring and assessing a range of pollution- and climate-related issues to "*provide reliable and sufficient information on the status of, and threats to, the Arctic environment, and scientific advice on actions to be taken in order to support Arctic governments in their efforts to take remedial and preventive actions relating to contaminants and adverse effects of climate change*" (AMAP, 2010a). In this context, AMAP has produced a series of assessments that address the occurrence of environmental contaminants characterized as 'persistent organic pollutants' (POPs), as well as chemicals and groups of substances that can be summarized as chemicals of emerging Arctic concern (CEACs).

The information contained in the first AMAP assessment report (AMAP, 1998) was instrumental in stimulating bans on selected POPs, including the development of the global United Nations Environment Programme (UNEP) Stockholm Convention on Persistent Organic Pollutants[3] (UNEP, 2001). The Stockholm Convention's POPs Global Monitoring Plan (GMP) tracks the effectiveness of bans and restrictions on listed chemicals using temporal trend data primarily from human tissues and air, but monitoring data for other media are accepted as well. Arctic monitoring data, and especially air and biota temporal trend data sets, have been influential in supporting the further development and implementation of the Stockholm Convention, including the GMP (UNEP, 2013), and the evaluation of chemicals proposed for listing under the Convention.

In the first Stockholm Convention GMP report (UNEP, 2009) it was stressed that climate change could have implications for interpreting POP temporal trend data. In response, a joint UNEP/AMAP (2011) report entitled *Climate Change and POPs: Predicting the Impacts* was produced with the purpose of assessing the potential effects of climate change on POP emission sources, transport processes and pathways, partitioning, degradation and exposure in wildlife and humans. The report recognized climate change as a factor capable of influencing POP levels measured in the environment and humans. Therefore, it is an issue that could potentially interfere with the interpretation of temporal trends used to monitor the effectiveness of the Stockholm Convention. Very few data were available at that time, so the report mainly hypothesized how changes in temperature and other climatic parameters could affect the environmental behavior and fate of POPs based on their physical-chemical properties. The report was global in scope, although Arctic data were used where available, and many of the examples discussed included potential impacts in the Arctic.

The report's general conclusion was that increased temperatures related to climate change would probably be the main factor influencing the fate of POPs. Warmer temperatures were predicted to increase primary emissions of POPs associated with volatilization from in-use products and equipment, waste sites and stockpiles. Due to expected changes in the environmental partitioning of chemicals, warming was also predicted to increase secondary emissions of POPs from environmental stores, including revolatilization from soil, water and particulates, leading to increased air concentrations. Additionally, releases from soil and ice would increase POP levels in aquatic environments. Environmental fate, including the long-range transport of POPs would also be influenced by climate-induced changes in wind speed, precipitation, ocean currents, biotic transport, the frequency of extreme weather events and the melting of polar ice caps and glaciers. Warmer temperatures would likely increase the degradation and transformation of POPs, potentially decreasing environmental concentrations of the emitted compounds, but increasing the proportion of degradation products present in the environment. Thus, changes to the physical environment, both inside and outside the Arctic, would affect the transport of POPs and their behavior in the Arctic. A modeling exercise supported these projections for polychlorinated biphenyls (PCBs) (Lamon et al., 2009; UNEP/AMAP, 2011).

Climate change and climate variability were also predicted to have effects on biodiversity, ecosystem composition and function, and food web structure and dynamics. Climate-induced changes in primary production had previously been noted as a factor that could influence the fate and bioavailability of POPs in the Arctic (AMAP, 2003). The UNEP/AMAP (2011) report found that the available food web modeling for POPs (Borgå et al., 2010) and empirical observations for freshwater fish (Carrie et al., 2010) showed contradictory results, possibly due to the different ecosystems being studied (pelagic marine vs. benthic freshwater), and highlighted the lack of understanding regarding the effects of climate change on primary production and POP cycling. The report also considered the potential effects of higher temperatures on toxicokinetics and the toxicity of POPs to wildlife and humans. Thus, the UNEP/AMAP (2011) report was a significant contribution to the assessment of climate change impacts on the long-range transport and environmental fate of POPs.

The AMAP-coordinated Arctic Health Risks (ArcRisk) project (ArcRisk, 2014; Pacyna et al., 2015; AMAP, 2016a; Carlsson et al., 2018) funded under the European Union's 7th Framework Programme (2009–2014) addressed many of the predicted effects of climate change on POPs with a focus on

1 http://www.amap.no

2 http://www.arctic-council.org

3 http://chm.pops.int

the European Arctic. ArcRisk used modeling tools to study the atmospheric and oceanic transport of POPs to the Arctic and the subsequent bioaccumulation of POPs in the Arctic marine food web under present climate conditions and projected future climate scenarios. Most modeling results projected only modest changes to levels of POPs in air, soils, and water as a result of a warming climate (Wöhrnschimmel et al., 2013; Pacyna et al., 2015). However, the ability to model climate-related impacts on POP bioaccumulation was limited by the lack of understanding regarding the effects of climate change on primary production, species distributions and trophic interactions.

The rapid changes in the Arctic induced by increasing temperatures have been addressed by several organizations (ACIA, 2005; IPCC, 2014) and were recently summarized by AMAP (2018a). The annual average warming in the Arctic continues to be twice that of the global mean, with spatial temperature anomalies that are even higher, especially for October–May (Figure 1.1). Annual precipitation seems to be increasing as well, with less precipitation falling as snow and more as rain in some regions. Sea ice extent is decreasing, and multi-year sea ice is being increasingly replaced by annual ice. Other relevant climate change-related processes include ocean acidification and sea level rise. All these changes have direct and indirect effects on biodiversity and ecosystems (AMAP, 2018a).

The AMAP Working Group thus saw a need for an updated assessment when more data would be available to compare observed climate impacts on POPs to those hypothesized in the UNEP/AMAP report (2011) and predicted by the various modeling scenarios under ArcRisk (ArcRisk, 2014; Pacyna et al., 2015). Recent progress in this area has meant that there is now a basis for a more detailed scientific assessment, including a re-evaluation of knowledge gaps and recommendations for future work.

Recent reviews of climate and POP interactions (McKinney et al., 2015; Ma et al., 2016) provide an additional foundation for this assessment. McKinney et al. (2015) reviewed the literature on climate change-induced ecological changes and alterations in POP exposures. They concluded that dietary changes linked to reduced sea ice were associated with higher contaminant levels in some marine biota, but the influence of changing trophic interactions on POP levels and trends varied widely in both magnitude and direction. Ma et al. (2016) found there was observational evidence indicating that climate variation had an effect on POP levels in biotic and abiotic environments. However, they noted that the statistical power of current Arctic time series for POPs in biota was limited and required more monitoring time points. Both Ma et al. (2016) and McKinney et al. (2015) pointed to the need for more information on dietary composition and other ancillary ecological metrics to help reveal climate change and POP interactions as a result of the complex mechanisms by which these interactions can occur and numerous contributing factors, including changing primary emissions from the manufacture, use, and disposal of chemicals and products.

This assessment does not address the effects of climate change on human exposure to POPs. However, many of the fish and wildlife species referenced in this report, including seabirds, seals, beluga, and polar bears, form part of the diet of Arctic indigenous peoples, and thus provide evidence of the effects

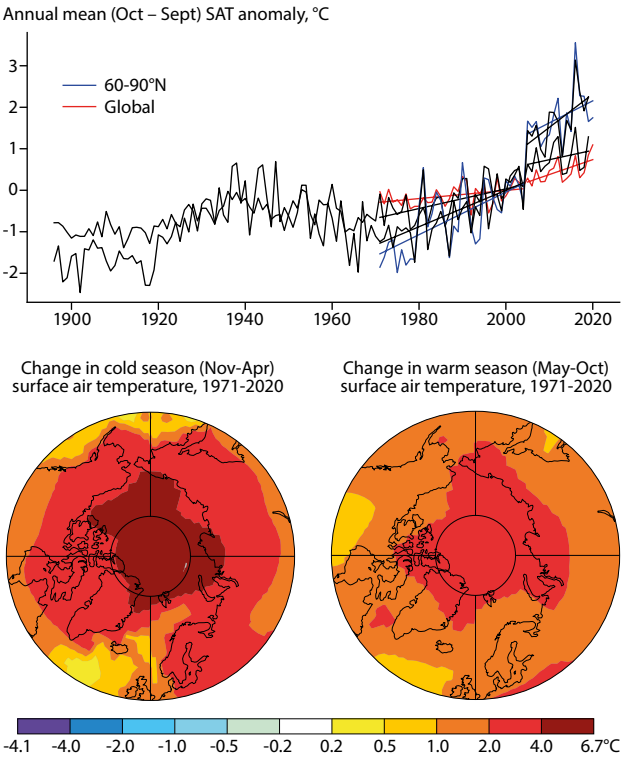


Figure 1.1 Temperature change in the Arctic. Upper panel: Arctic (blue line) and global (red line) mean annual land surface air temperature (SAT) anomalies for the period 1900–2019 relative to the 1981–2010 mean value. Arctic measurements taken from land stations north of 60°N. Source: Overland et al. (2019). Lower panel: Spatial patterns in Arctic surface air temperature changes for the period 1971–2020 in cold (October–May) and warm (June–September) seasons. Source: NASA Goddard Institute for Space Studies, http://data.giss.nasa.gov/gistemp/maps/.

of climate change on POP levels and trends that is valuable for understanding human exposures. The ArcRisk project considered human dietary-exposure scenarios related to the impact of climate change, but noted the complexity involved due to multiple unknowns, including future contaminant trends in fish, potential changes in fishery supply, and nutritional transitions in Arctic communities (ArcRisk, 2014; Pacyna et al., 2015). The AMAP Human Health assessment (AMAP, 2015), while not specifically focusing on exposure to POPs, concluded that the combined effects of climate warming, anthropogenic contaminants and zoonotic diseases represent a significant risk for subsistent food and drinking water supplies of northern communities.

A previous AMAP assessment (AMAP, 2018b) addressed the biological effects of POPs and climate change on fish and wildlife to a limited extent, focusing primarily on high-level predators such as polar bears and killer whales. That report considered some biological effects related to climate change-induced ecosystem changes that could affect wildlife exposure to POPs. However, it did not cover the direct effects of climate change on POP toxicokinetics or toxicity in wildlife.

The current assessment builds on the themes from the UNEP/AMAP (2011) report, ArcRisk (2014) project, and other publications by assessing impacts of climate change on the environmental fate and long-range transport of POPs, as well as climate-related changes to food webs that may impact temporal trends of environmental contaminants. It focuses

Table 1.1 The predominant persistent organic pollutants (POPs) and chemicals of emerging Arctic concern (CEACs) discussed or referenced in this assessment.

Chemical / Chemical Group	Primary Sources			
	Industrial and consumer uses	Agricultural and disease control uses	Unintentional byproducts	Natural products
Legacy POPs				
Polychlorinated biphenyls (PCBs)	X		X	
Hexachlorobenzene (HCB)	X		X	
Polychlorinated dibenzo-*p*-dioxins (PCDDs)	X		X	X
Polychlorinated dibenzofurans (PCDFs)	X		X	X
Dichlorodiphenyltrichloroethane (DDT)		X		
Chlordanes		X		
Heptachlor		X		
Toxaphene		X		
Mirex		X		
New POPs				
Polybrominated diphenyl ethers (PBDEs)	X			
Hexabromocyclododecane (HBCDD)	X			
Perfluorooctane sulfonic acid (PFOS)	X			
Perfluorooctanoic acid (PFOA)	X			
Short-chain chlorinated paraffins (SCCPs)	X			
Pentachlorophenol (PCP)	X	X		
α-, β- and γ-hexachlorocyclohexanes (HCHs)		X		
Endosulfan		X		
Polychlorinated naphthalenes (PCNs)	X		X	
Other Chemicals & Substances				
Perfluorocarboxylic acids (PFCAs)	X			
Perfluorosulfonic acids (PFSAs)	X			
Perfluorohexane sulfonic acid (PFHxS)	X			
Perfluoroalkyl acids (PFAAs)	X			
Organophosphate esters (OPEs)	X			
Dacthal		X		
Chlorpyrifos		X		
Trifluralin		X		
Pentachloronitrobenzene (PCNB)		X		
Polycyclic aromatic hydrocarbons (PAHs)			X	X
Pentachloroanisole (PCA)			X	
Microplastics	X			
Halogenated Natural Products (HNPs)				
Bromoanisoles (BAs)				X
Hydroxylated PBDEs (OH-PBDEs)	X			X
Methoxylated PBDEs (MeO-PBDEs)				X

Legacy POPs: Chemicals included in the original 'dirty dozen' listed under the 2004 Stockholm Convention. New POPs: Chemicals listed under the Stockholm Convention between 2005–2019. PBDEs include tetra-, penta-, hexa-, hepta- and deca-congeners. See Chapter 2.2 for a more detailed list of CEACs (Table 2.2.2) and HNPs (Box 2.2.1).

mainly on legacy or 'initial' POPs (e.g. PCBs; DDTs) due to the large amount of information available. However, data for 'new' POPs (e.g. polybrominated diphenyl ethers (PBDEs) and per- and polyfluoroalkyl substances (PFAS)) are also included where data are available. Thus, when referring to 'POPs' the assessment includes both the initial POPs and new POPs, as defined by the Stockholm Convention.

Selected CEACs (AMAP, 2017a), including halogenated natural products (HNPs), halogenated and organophosphorus flame retardants, and polycyclic aromatic hydrocarbons (PAHs) are also referenced herein. Although not listed under the Stockholm Convention, PAHs are listed by the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (UNECE, 1998) and there are substantial data for pyrogenic-related PAHs in the Arctic (Balmer et al., 2019a). The report also reviews the current knowledge of climate-related effects on microplastic pollution in the Arctic, considering the growing concern for plastics as an emerging pollution issue in the region (AMAP, 2017a). The predominant chemical substances included in this report are listed in Table 1.1.

Box 1.1 Climate oscillation indices

Various parts of this assessment refer to climate oscillation indices, including the Arctic Oscillation (AO), North Atlantic Oscillation (NAO), El Niño Southern Oscillation (ENSO), Pacific/North American Pattern (PNA) and the Pacific Decadal Oscillation (PDO).

These indices reflect cyclical variations in large-scale regional climate patterns. As such, they are associated with different 'climate states' that can be related to contaminant transport pathways and climate-induced changes in environmental contamination, as well as conditions that may represent possible future climate 'norms'.

In particular, the AO and NAO have been associated with changes in environmental contamination in Arctic studies and these are therefore briefly described below. Further details and illustrations can be found on the National Oceanic and Atmospheric Administration (NOAA) website: https://www.ncdc.noaa.gov/teleconnections/.

The AO index is characterized by winds circulating counter-clockwise around the Arctic at around 55°N latitude and is often considered a surface-level expression of the stratospheric polar vortex (Figure 1.2; AMAP, 2003). A positive AO phase (AO+) represents a ring of strong winds circulating around the North Pole confining colder air across polar regions. A negative AO phase (AO-) therefore represents weaker and more distorted winds, which allow an easier southward penetration of colder, Arctic air masses and increased storminess into the mid-latitudes (NOAA, 2019). The NAO index represents the normalized gradient in air pressure at sea level between the Azores/Lisbon, Portugal and Stykkishólmur/Reykjavik, Iceland, as per AMAP (2003). A positive phase of the NAO (NAO+) tends to be associated with above-normal temperatures in the eastern United States and across northern Europe, and below-normal temperatures in Greenland. It is also associated with above-normal precipitation over northern Europe and Scandinavia. A negative NAO phase (NAO-) is associated with the inverse situation (NOAA, 2019).

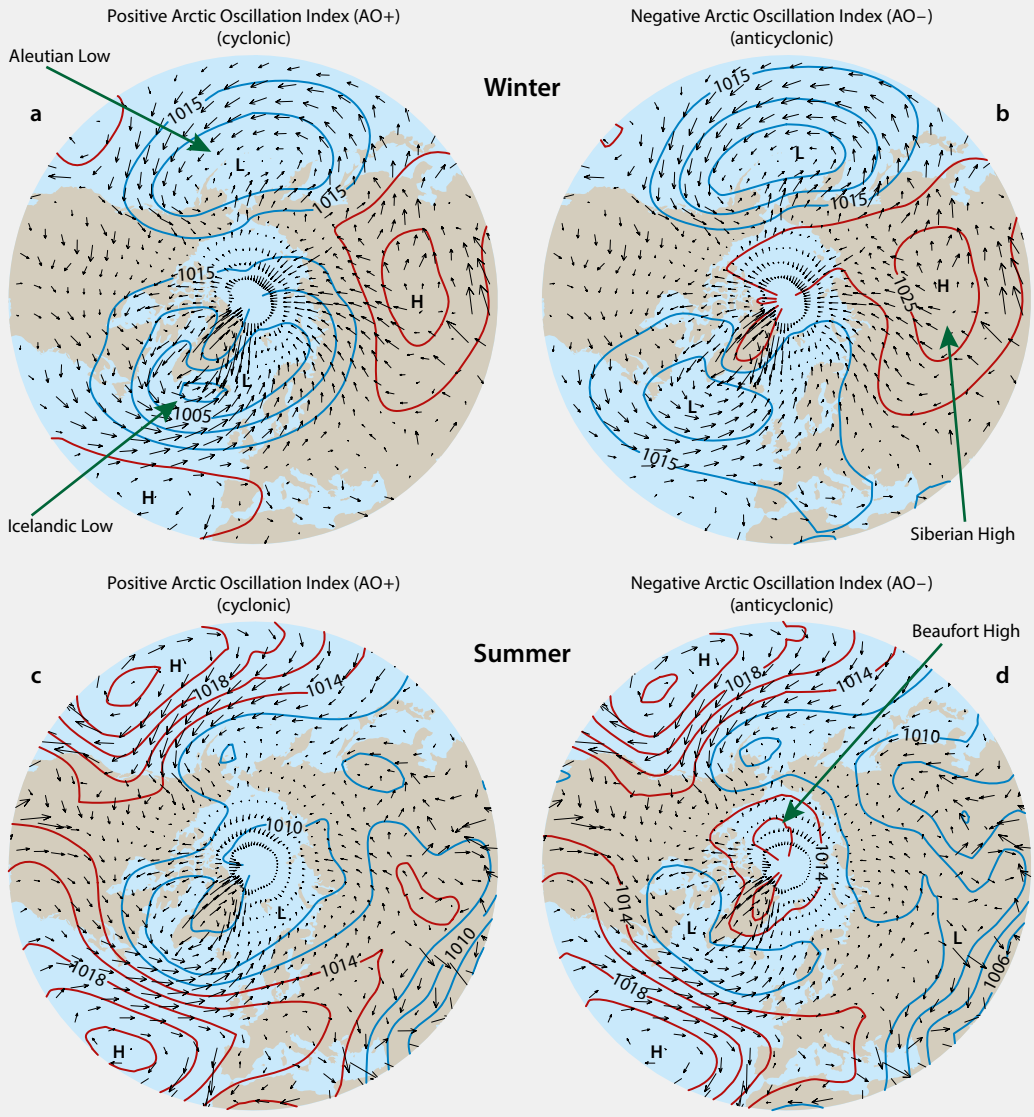


Figure 1.2. Atmospheric pressure fields and wind stream lines in the Northern Hemisphere for (a) strong AO+ conditions in winter; (b) strong AO- conditions in winter; c) strong AO+ conditions in summer; and d) strong AO- conditions in summer. Small arrows show the geostrophic wind field associated with the AO pattern with longer arrows implying stronger winds.

The goal of this assessment is to review the existing information on the effects of climate change on POPs and CEACs in the Arctic and to provide an assessment of current knowledge and recommendations for future work. This assessment attempts to address the following policy-relevant science questions:

- What are the primary sources of POPs, how do POPs reach the Arctic, and how are emissions and source locations of POPs and CEACs affected by climate change?
- Does climate change exacerbate or diminish contaminant transport, accumulation and occurrence in different abiotic media?
- How do local sources contribute to Arctic contamination compared to long-range transport under climate change scenarios?
- How well can we anticipate how POPs and CEACs, as well as microplastics, will impact the Arctic in a changing future climate?
- What are the key climate change-driven physical and/or ecological processes influencing POPs in Arctic wildlife, and how will climate change influence levels of POPs in Arctic biota and food webs?
- Can we link changes in temporal trends of POPs with climate parameters and/or food web changes?
- Do the findings related to temporal trends in POPs in air and biota have implications for the national and international regulation of chemicals?
- How can Indigenous Knowledge (IK) contribute to the discussion of climate-related effects on trends of POPs?

These questions are addressed in Chapter 2. Each subchapter of Chapter 2 summarizes the current status of knowledge regarding climate change-driven effects on POPs and CEACs in the Arctic and presents specific conclusions, recommendations and knowledge gaps. Chapters 2.1–2.4 are based on reviews of the scientific literature. Chapter 2.1 focuses on modeling of emissions and long-range transport of POPs under climate change scenarios. Chapter 2.2 reviews the growing number of studies on levels and trends of POPs and CEACs in abiotic media, such as air and ice cores, and also addresses the potential impacts of increased human activity on contaminant levels in the Arctic. Chapter 2.3 examines the climate-induced changes in ecosystem structure and function that may impact POP exposure, as observed in food webs and migratory species. Chapter 2.4 reviews the growing literature on climate-related effects on temporal trends of POPs in Arctic biota. There are many strong time-series datasets on POPs in Arctic biota (AMAP, 2016b; Rigét et al. 2019), that have not yet been assessed for associations with metrics that reflect climate change and variability, including climate oscillation indices (Box 1.1). Climate oscillation indices are briefly introduced here because they represent a potentially important source of information for some regions of the Arctic and are referred to in all chapters of this report. To strengthen the conclusions drawn from this assessment, Chapters 2.2, 2.3 and 2.4 include some relevant research findings from Antarctica and Tibet, where similar questions on the effects of climate change on POPs in cold climates are being studied. Chapter 2.5 addresses the involvement of Arctic indigenous communities in environmental monitoring and research on POPs. It includes examples of close collaborations between scientific and indigenous communities that have resulted in strong time series for POPs, and describes possible ways that IK of climate-related changes in local physical- and ecological-conditions could be more effectively utilized for interpreting POP trends in wildlife with additional capacity building. Chapter 3 includes general conclusions, key findings and knowledge gaps derived from Chapters 2.1–2.5, as well as policy-related recommendations.

This report constitutes the final part of a multi-component assessment that updates information on temporal trends of POPs in the Arctic (AMAP, 2016b), chemicals of emerging Arctic concern (AMAP, 2017a), the biological effects of POPs and mercury on Arctic wildlife (AMAP, 2018b), and the influence of climate change on POPs and CEACs (this report).

2.1 Modeling emissions and long-range transport of POPs and CEACs under climate change

Authors: Matthew MacLeod and Paul Woods Bartlett

Co-authors: Gerhard Lammel, Jianmin Ma, Alexey Gusev, Li Li, Marilena Muntean

Contributing authors: Ian Cousins, Carey Friedman, Kaj Mantzius Hansen, Yi-Fan Li

2.1.1 Introduction

Models provide a platform to synthesize and evaluate the state of knowledge and understanding of the emissions, distribution, and long-range transport of persistent organic pollutants (POPs) and other environmental contaminants (MacLeod et al., 2010; Di Guardo et al., 2018). Several modeling studies have examined the regional and global fate of POPs and chemicals of emerging Arctic concern (CEACs), including under climate change scenarios. POPs and some CEACs disperse into air, water, soils and sediments, and can be taken up by biota. Many of the processes that determine the environmental fate and transport of these chemicals and their potential for uptake and bioaccumulation in food webs can be influenced by climate change. Improving our understanding of global pollution in a changing climate using models as conceptual platforms requires confronting uncertainties and ignorance in several dimensions, which include: 1) a lack of emission data, not only in temperate regions where use and release of POPs and CEACs is highest, but also in the Arctic, 2) the limited amount of monitoring data appropriate for model validation, especially for emerging POPs and CEACs, 3) uncertainties in chemical degradation pathways and rates, where knowledge is generally limited, but is especially poor for soils, seawater and the cryosphere, and 4) uncertainties in process descriptions and parameterizations, most notably for surface-air exchange processes for all media.

In 2011, a joint report produced by the United Nations Environment Programme (UNEP) and Arctic Monitoring and Assessment Programme (AMAP) entitled *Climate Change and POPs: Predicting the Impacts* summarized what was known at the time regarding the effects of climate change on the primary emissions, fate and transport of POPs to the Arctic (UNEP/AMAP, 2011). Since the drafting of that report, research utilizing models to study the emissions and long-range transport of POPs and CEACs under climate change scenarios has been ongoing. This chapter summarizes the current knowledge of emissions and transport pathways for POPs and CEACs in a changing climate, with a focus on integrated assessments using models. It builds on the 2011 report by addressing four key policy-relevant questions about the pathways for organic pollutants to reach the Arctic. These four questions, addressed in sections 2.1.2 through 2.1.5 are:

What are the primary sources of POPs and how do POPs reach the Arctic?

How are emissions and source locations of POPs and CEACs affected by climate change?

How does climate change affect the transport of POPs and CEACs to the Arctic?

How well can we anticipate how POPs and CEACs will impact the Arctic in a changing future climate?

Here, we address the four questions by summarizing available scientific evidence while identifying key sources of uncertainty and unknowns. Answers are presented with an explicit assessment of the degree of confidence that can be assigned to the information and a discussion of implications, as indicated by colored bars next to each paragraph (Box 2.1.1).

Box 2.1.1

Color key used in this chapter.

- What is known
- What is partially known and/or uncertain
- What is unknown
- Implications

Wherever possible, references to primary scientific literature are provided, but several scientific reviews about modeling the long-range transport of global contaminants under climate change have been published (e.g., Armitage et al., 2011; Gouin et al., 2013; Ma et al., 2016; Wang et al., 2016), and these reviews are cited where interpretations depend on a synthesis of several studies or different lines of evidence.

2.1.2 What are the primary sources of POPs, and how do POPs reach the Arctic?

2.1.2.1 What is known

Releases of POPs into the environment that occur during production, use or disposal, or as a by-product of other activities are referred to as primary emissions, while the remobilization of POPs from environmental reservoirs (often soils or sediments) are considered secondary emissions. Primary emissions of POPs result, for example, from direct application to soils and into air (e.g. pesticides), from volatilization or suspension into air in association with particles (e.g. semi-volatile polychlorinated biphenyls (PCBs) and semi- and low-volatility polybrominated diphenyl ethers (PBDEs)), and by leaching into soil and water during use (e.g. perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and their precursors). Climate change has the potential to influence the location and amount of primary POP emissions by several mechanisms.

The dominant primary emission sources of POPs are determined by the production, use and disposal of relevant chemical products. Historically, perfluorocarboxylic acids (PFCAs) such as PFOA were emitted from industrial discharges during production and use. The manufacture of fluoropolymers contributed the bulk (55–83%) of global PFCA emissions released between 1951 and 2002 (Wang et al., 2014), and the production of PFOS was an important (17%-62%) source of global historical PFOS emissions between 1958 and 2002 (Wang et al., 2017a). For POPs that are pesticides (e.g. dichlorodiphenyltrichloroethane (DDT), technical hexachlorocyclohexanes (HCHs), lindane (gamma-HCH), and dicofol), emission during the use phase (i.e. agricultural application) dominates (Li et al., 2000, 2003, 2015; Hansen et al., 2004, 2008; Becker et al., 2011). For POPs such as these, where primary emissions predominantly originate from specific activities that are responsive to changes in chemical production and/or use, environmental releases can be reduced rapidly and substantially by targeted regulations or voluntary emission reduction programs (Prevedouros et al., 2006; Paul et al., 2009).

In contrast, for POPs that are incorporated into commodities (i.e. equipment or materials), primary emissions can occur throughout the lifecycle of the product. Examples include PCBs used in electrical equipment, sealants, lubricants and heat transfer fluids (Breivik et al., 2007, 2016), and PBDEs (Prevedouros et al., 2004; Schenker et al., 2008a; Earnshaw et al., 2013; Abbasi et al., 2019) and hexabromocyclododecanes (HBCDDs) used as additive flame retardants (Li and Wania, 2018a). Primary emissions of POPs incorporated into commodities are closely linked to the commodity itself, and consequently, the world trade of goods and waste has become a significant vehicle of global-scale distribution of PCBs (Breivik et al., 2016; Li and Wania, 2016) and PBDEs (Breivik et al., 2016).

The relative importance of different lifecycle stages as sources of POPs can also vary. More than 80% of global PBDE emissions have been attributed to the in-service use of PBDE-containing commodities, whereas just 15% originate from industrial processes, and 5% from the disposal of waste in treatment facilities (Abbasi et al., 2019). However, in the case of global HBCDD emissions, industrial sources (i.e. industrial processes and disposal of industrial waste) make almost the same contribution as consumer sources (use phase and disposal of end-of-life waste) to global primary emissions (Li and Wania, 2018a). Moreover, the relative importance of sources at different lifecycle stages varies between geographic regions when regional industrial and consumer activities are different. For example, inappropriate treatment of PBDE-containing waste (e.g. open burning of e-waste) is the second-largest source of PBDEs (after the use-phase) in less industrialized regions in the world, whereas production of the technical PBDE mixtures is the second-largest source of PBDEs in industrialized regions (Abbasi et al., 2019). The relative importance of sources can also change over time, shifting from dominance of industrial processes or in-use stock prior to, or soon after, a ban on production and new use, to dominance of emissions from waste stock long after the ban (Li and Wania, 2018b). For instance, α-HBCDD has become increasingly more abundant than γ-HBCDD in environmental profiles during the past decades, which is indicative of the transition from industrial processes (resulting in a γ-HBCDD-dominant diastereomer profile) to that of the use phase and waste disposal (resulting in an α-HBCDD-dominant diastereomer profile) as the dominant sources (Li and Wania, 2018a).

Commodities with long service lifetimes can retain associated POPs in the use phase of their lifecycle for decades, and since POPs are persistent in landfills, primary emissions of POPs from commodities can continue for decades and potentially even for centuries after a ban on their production and new use (Breivik et al., 2007, 2016; Li et al., 2016; Abbasi et al., 2019). For example, the release of HBCDDs in China is expected to rise again 20 years after the nationwide phase-out pursuant to the Stockholm Convention as HBCDDs embedded in insulating styrene boards are liberated due to the demolition or dismantling of associated products at the end of their lifetimes (Li et al., 2016). In other words, the primary emissions of POPs associated with long-lived commodities is poorly reversible, and these products may act as long-term "time bombs" contributing to environmental releases in the future (Persson et al., 2013; MacLeod et al., 2014).

Other important categories of primary emissions include unintentional emissions of PCBs, hexachlorobenzene (HCB), pentachlorobenzene (PeCB), polycyclic aromatic hydrocarbons (PAHs) and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDDs/PCDFs) from waste incineration and other combustion sources, in particular household wood combustion (Denier van der Gon et al., 2007; Shen, et al., 2013; Yang et. al., 2019), biomass burning from wildfires (Yu et al., 2019), and the formation of POPs from chemical precursors. The oxidative degradation of nonpolymeric fluorotelomer- and perfluorooctanesulfonyl fluoride-based derivatives has been confirmed to contribute to the occurrence of PFOA in the Arctic and global environment (Wania, 2007; Schenker et al., 2008b).

Decades of modeling research have been devoted to studying the long-range transport of organic chemical pollutants in the environment (Wania and Mackay, 1999; Scheringer, 2009). The potential for chemicals to be transported to the Arctic in the environment is determined jointly by 1) how persistent they are in the environment, 2) their mode of emission (i.e. whether primary emissions occur to air, water or soil), 3) the properties of the substance (e.g. vapor pressure, water solubility and octanol/water partition coefficient, which is correlated with the extent of sorption to soil organic matter and lipids for many POPs), and 4) the spatiotemporally variable conditions of the air, seawater, soil, and other phases that make up the Earth system (Figure 2.1.1). For the majority of POPs, the atmosphere is the most important and rapid pathway of transport to the Arctic, but ocean currents may be the dominant transport pathway over decadal time scales for some chemicals (AMAP, 2003).

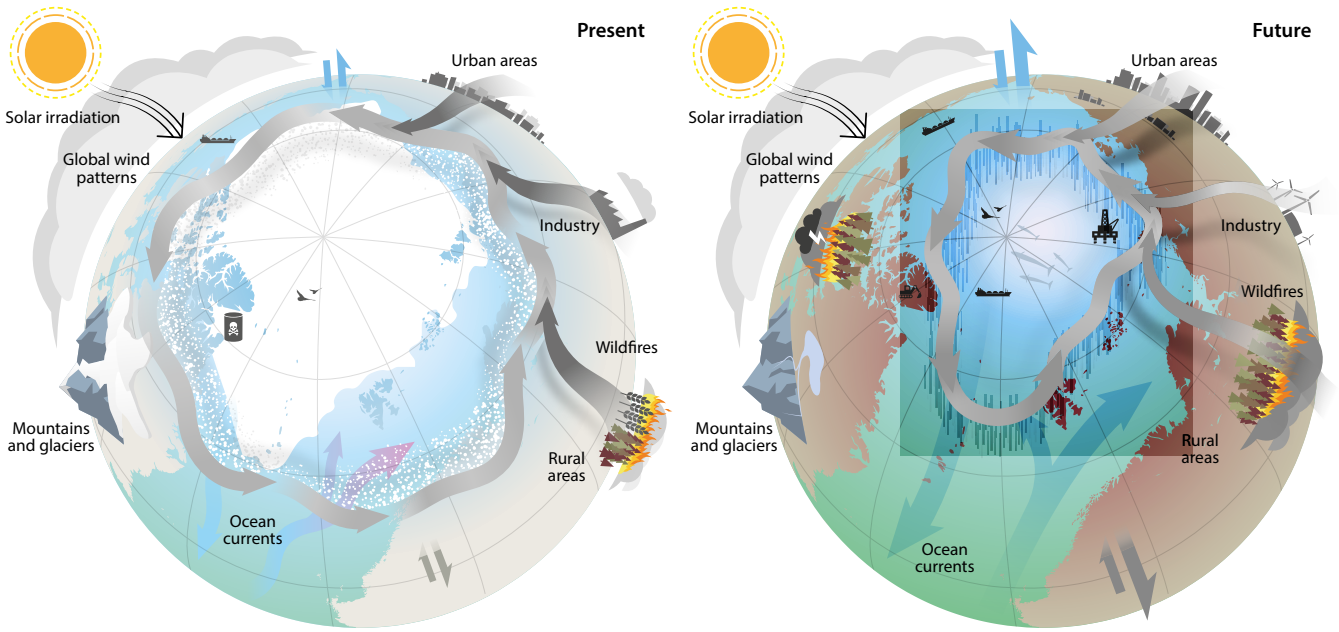


Figure 2.1.1 Pathways for POPs and CEACs to reach the Arctic that could be impacted by climate change.

2.1.2.2 What is partially known and/or uncertain

Primary emission inventories are currently available for a group of POPs that includes: HCHs (Li et al., 2000, 2003), dicofol (Li et al., 2015), PCBs (Breivik et al., 2002a, 2002b, 2007; Cui et. al., 2013), PFCAs (Wang et al., 2014), perfluorosulfonic acids (PFSAs) (Wang et al., 2017a), short-chain chlorinated paraffins (SCCPs)(Glüge et al., 2016), PCDD/Fs (Bartlett et al., 2000; Commoner et al., 2000, 2003; Cohen et al., 2002), PAHs (Shen et al., 2013), PBDEs (Schenker et al. 2008a; Abbasi et al., 2019), and endosulfan (Becker et al., 2011). However, only a few of these emission inventories are spatially- and/or temporally-resolved, and some report only annual global estimated emissions. In the absence of chemical-specific information about the location of primary emissions, generic emission maps that use proxies to allocate emissions spatially have been proposed. For example, the intensity of light emissions to space at night and the intensity of global crop production have been suggested as useful proxies for the location of industrial and agricultural chemical emissions, respectively (von Waldow et al., 2010) and used to drive dynamic simulations using global chemical fate and transport models (Göktaş and MacLeod, 2016). In the absence of information about the temporal evolution of emissions, generic use profiles that represent typical timescales for periods of rising and falling emissions derived from experience with legacy POPs have been used to drive models (e.g. Wöhrnschimmel et al., 2013).

The role of international trade of technical mixtures, finished products and/or waste as a pathway for POPs to remote areas, including the Arctic, is receiving increasing attention. For instance, the transboundary trade of technical PCB mixtures was found to be more efficient than environmental pathways in driving the global distribution of PCBs (Li and Wania, 2016). Global food trade could also provide an efficient and important pathway of POPs between food origins and consumption (Ng and von Goetz, 2017; Jiang et al., 2019). The transboundary trade of POP-containing commodities and waste may explain why certain chemicals are abundant in the Southern Hemisphere where they have never been produced or deliberately used (Breivik et al., 2016; Gilljam, et al., 2016). When products or commodities containing POPs are transported into the Arctic they become local primary sources. Prominent recent examples include the PFCAs, PFSAs and their precursors that are found in aqueous film forming foams. The use of these foams for fire suppression exercises in Arctic airports has resulted in contamination in three Arctic lakes (Meretta, Char, and Resolute) on Cornwallis Island, Canada (Stock et al., 2007).

Many persistent organic chemical pollutants are semi-volatile and may move reversibly from the gas phase to the particulate phase in the atmosphere (Bidleman et al., 1988). In the gas phase, chemicals are more mobile and can travel long distances depending on precipitation and photochemical degradability. In the particulate phase, chemicals can be shielded from oxidant reactions, and their lifetime in air is limited by dry and wet particle deposition. Accurately modeling the gas-particle partitioning of POPs is therefore a key aspect of describing their atmospheric fate and long-range transport. However, atmospheric models often neglect the effect of kinetic limitations on mass transfer on gas-particle partitioning, which becomes significant for higher molecular weight PBDEs (i.e. congeners with log octanol-air partition coefficients (K_{OA}) ≥ 11) (Cetin and Odabasi, 2008; Li et al., 2017a; 2019; Zhao et al., 2021).

2.1.2.3 What is unknown

Emission inventories, especially at a global level, are still absent for many compounds, especially newer POPs and CEACs that are under current discussion as POP candidates. The process whereby POPs volatilize from materials and/or surfaces of consumer commodities, and how volatilization responds to environmental conditions (e.g. ambient temperature and

wind speed) require more study. In addition, information about the release of POPs from waste treatment, disposal, and incineration facilities (e.g. landfills, dumpsites, and wastewater treatment plants) remains lacking. For some POPs, the extent of primary emissions arising from the degradation of precursors, and the impact of that process on Arctic contamination, remains poorly constrained. For example, the degradation of fluorotelomer-based polymers was recently proposed as a possible mechanism for the observed rebound of fluorotelomer alcohol (FTOH) concentrations in the global environment and as a considerable source of PFCAs in the future (Wang et al., 2014; Li et al., 2017b).

Measured values of physical-chemical properties needed to estimate emissions and model environmental fate and transport to the Arctic are lacking for many new chemicals and CEACs. AMAP initiated a crowd sourcing effort to collect physical chemical properties of CEACs, but most of this data is derived from structure-property relationship models (AMAP, 2017a).

2.1.2.4 Implications

Models link emission inventories to measurements and can be used to demonstrate a quantitative understanding of source-to-receptor concentration relationships over distance and time. Both emissions and environmental levels are uncertain, but emissions are often more uncertain, and in extreme cases, the emission estimates needed to drive models may be lacking entirely for CEACs and newer POPs.

There is clearly an incentive to apply models to analyze scenarios within the range of uncertainty for emissions, chemical properties and process parameterizations to generate testable hypotheses that can guide targeted measurements. The process of applying models in an iterative fashion with evolving information about emissions, environmental levels and properties has been called the "integrative approach" to understanding chemical pollution (Shatalov et al., 2010; Gusev et. al., 2012; Dutchak and Zuber, 2013).

Improving primary emission inventories for POPs and CEACs should be a high priority. Studies that use "bottom-up" approaches, particularly those with integrative, lifecycle perspectives, to estimate emissions from information about the production, use and disposal of chemicals should be encouraged. However, given the difficulties associated with carrying out "bottom-up" studies, research using "top-down" approaches where models are first fitted to measurement data to estimate emissions, and then those emission estimates are evaluated in model scenarios that can be validated against independent measurement data, should also be encouraged (e.g. Gasic et al., 2009; Moeckel et al., 2010; Buser et al., 2013; Bogdal et al., 2014; Csiszar et al., 2014).

The "top-down" approach can be used to constrain the locations and strength of emissions in inventories by minimizing the errors between measured and predicted air concentrations. However, the application of a "top-down" approach in the construction of POP emission inventories must be done in a manner that is appropriate for the amount of data available. The "top-down" method must be based on sufficiently abundant sampling data to constrain the number of degrees of freedom reflected by fittable parameters in the optimization. "Top-down" emission estimates made with too few observations relative to the number of degrees of freedom can lead to erroneous estimates of emissions rates (Daley, 1991).

Ultimately, the burden of chemicals present in the environment (and the exposure of biota and humans to chemicals via the environment) is determined by primary emissions, so success in mitigating exposures should be measured by the reduction of emissions (Hung et al., 2013).

2.1.3 How are emissions and source locations of POPs and CEACs affected by climate change?

2.1.3.1 What is known

Climate change is expected to drive movements of people and changes in economic activities that in turn affect the rate and location of primary emissions of POPs and CEACs from the production, use and disposal of chemicals and associated commodities. If economic activity and population growth are enhanced in northern latitudes, primary emissions of POPs will be brought closer, or even into, the Arctic. For example, shipping and oil exploration activities are expected to increase in the Arctic (Peters et al., 2011), and scenarios based on that expectation have been used as a basis to estimate changes in PAH emissions in the Arctic region (Friedman et al., 2014).

Higher temperatures will lead to higher rates of primary and secondary emissions of POPs to the atmosphere through volatilization. Increased volatilization emissions driven by higher temperatures were identified as one of the main impacts of climate change in global model scenarios for PCBs (Lamon et al., 2009; Friedman and Selin, 2016) and for non-ionizing chemicals that span the entire plausible range of air, water and soil partitioning properties (Wöhrnschimmel et al., 2013).

A fraction of POPs that enter the environment accumulate in natural reservoirs, such as glaciers, permafrost, soil, and sediment, from which they can be re-mobilized when climate change drives a step-change in environmental conditions. For example, Bogdal et al. (2009) demonstrated that rapid glacial melt driven by a warming climate could increase levels of POPs in Alpine lakes due to the release of pollutants that were deposited to the glaciers during periods of high primary emissions.

The impact of climate change on emissions may also vary between different POPs and CEACs due to differences in the geographical location(s) of their emission sources. This is because primary emissions can be globally-distributed in patterns corresponding to locations of chemical production, use or disposal. For example, many of the pesticides banned as POPs under international regulations were released in the tropics and subtropics, which corresponds to the geographic distribution of the main crops and disease vectors that were targeted for application. Changes in the location of agricultural production due to climate change would lead to corresponding changes in the location of emissions of future

POPs (Wöhrnschimmel et al., 2013). Industrial chemicals and chemicals used in consumer products were mostly produced and used in northern temperate latitudes, thus emissions tend to correspond to the geographic distribution of populations in major developed and developing nations.

2.1.3.2 What is partially known and/or uncertain

An increase in wildfires resulting from climate change can be expected to increase the emission and redistribution of POPs and CEACs sequestered in trees, plants, permafrost and soils (e.g. Eckhardt et al., 2007; Luo, et al., 2020), but this has yet to be quantified on a large scale.

Higher temperatures under climate change scenarios could promote faster degradation of chemicals in waste stocks such as landfills and dumpsites, and thus reduce primary emissions relative to a baseline climate scenario (Breivik et al., 2002b). However, the extent that enhanced degradation could alter the emission rates of POPs from waste stocks has not been fully quantified. Furthermore, since the dominant sources of POPs vary between lifecycle stages and between regions, the geographic variability of climate change will impact individual chemicals in different and uncertain ways.

Mitigation of climate forcing by replacing fossil fuels with renewable energy sources may decrease emissions of PAHs and POPs from combustion sources. However, an increase of waste-to-energy incineration could result in larger emissions of a range of combustion-generated chemicals.

Climate changes that cause a shift in the geographic range and/or abundance of insects that are disease vectors or threats to agricultural productivity could plausibly result in the re-introduction or expansion of the use of POP pesticides, such as DDT (Chevrier, 2016).

Generic scenarios that illustrate possible future shifts in the location of emissions of POPs and CEACs have been proposed for industrial and agricultural chemicals and used to drive a global chemical fate and transport model (Wöhrnschimmel et al., 2013). The two scenarios assume enhanced chemical use in northerly latitudes based on forecasts of the future development of the Arctic economy and changing fertility of agricultural lands. Changes in the rate and location of emissions in the modeled scenarios increased concentrations of POPs in the Arctic released from global industrial and agricultural sources by up to factors of 2.1 and 1.6, respectively.

An increased incidence of extreme weather events will affect the mobilization of POPs from primary and secondary sources (Ma et al., 2005; Umlauf et al., 2005; Smit et al., 2008; Santiago and Rivas, 2012; Chi et al., 2013; Zuo et al., 2013; Lu et al., 2016). However, the net effect of mobilization of POPs due to extreme weather events has not so far been quantified in global-scale assessments.

Models suggest that secondary sources of POPs to the atmosphere can be important (Gouin and Wania, 2007; Armitage et al., 2011; Lammel and Stemmler, 2012; Wöhrnschimmel et al., 2012a), and the hypothesis that semi-volatile POPs move through the global environment in a series of "hops" analogous to chemicals in a gas chromatograph has been highly influential (Wania and Mackay, 1996). However, empirical evidence of secondary sources impacting the Arctic remains elusive (Roberts, 2012; Kong et al., 2014; Ubl et al., 2017). The relative influence of primary and secondary sources on Arctic pollution now and in the future thus remains unclear. However, as discussed above, primary sources are known to be important, and for many POPs are expected to continue for years or decades, even with aggressive abatement measures.

2.1.3.3 What is unknown

It is impossible to fully anticipate how production, use and emissions of POPs and CEACs will respond to pressures from climate change. Generic emission scenarios that have been proposed and analyzed with models do not reflect all possible future paths for all chemicals. Thus, the changes in exposure of the Arctic to specific POPs and CEACs is unknown.

2.1.3.4 Implications

High uncertainty and missing information about POP and CEAC emissions limit our understanding of the fate and transport of global pollutants. Research that combines emission estimates, models and measurements in an integrated approach to understanding chemicals in the global environment should be encouraged since it can help identify knowledge gaps and provide the basis for formulating testable hypotheses about key drivers of chemical fate and transport. Such studies also provide the basis for scenario analyses that can forecast levels and trends of global pollutants that can be expected under alternative climate and chemical use scenarios, as addressed in more detail in Section 2.1.4 below.

2.1.4 How does climate change affect the transport of pollutants to the Arctic?

2.1.4.1 What is known

Modeling studies have addressed how climate change influences the global fate and transport of chemicals (e.g. Ma et al., 2005; MacLeod et al., 2005; Lamon et al., 2009; Gasic et al., 2010; Wöhrnschimmel et al., 2012a; Armitage and Wania, 2013; Hansen et al., 2015). Since models are simplifications of reality, modeling studies can only examine a limited subset of climate conditions that might affect chemical fate and transport to the Arctic. Aspects of climate change that have been examined in modeling studies include: (1) enhanced primary and secondary emissions driven by temperature, (2) changes in transport efficiency and patterns due to changing wind directions, wind speeds and changes in the frequency and strength of patterns of natural climate variability, (3) effects of precipitation changes, (4) changing ocean currents, (5) melting of ice in polar caps, glaciers and permafrost, (6) effects of extreme weather events on chemical transport at the regional scale, (7) scenario analysis for possible changes in degradation and transformation of pollutants, (8) environmental partitioning between gas and particle phases, (9) biotic transport, and (10) changes to organic particulate matter dynamics in the Arctic Ocean (Armitage and Wania, 2013).

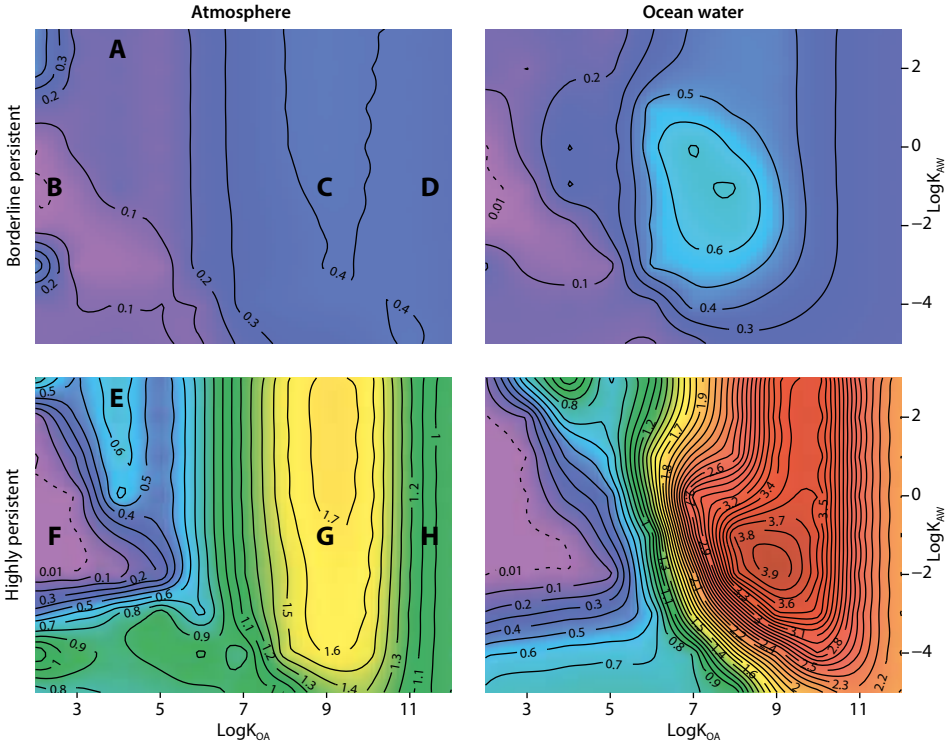
One hundred year forecasts generated by regional, hemispheric and global-scale models in the ArcRisk EU project estimated climate change would affect modeled PCB concentrations by a factor of 2 to 5 (Carlsson et al., 2018). The effect of climate change on modeled concentrations of pollutants with a wide range of partitioning properties and degradability in Arctic air and water using one of these models was similarly determined to be between a factor of 1.7 and 4 (Wöhrnschimmel et al., 2013). The results from model simulations suggest that the impact of climate change on chemical concentrations in the Arctic is small compared to the uncertainties inherent in modeling POP concentrations in the global environment.

2.1.4.2 What is partially known and/or uncertain

Large-scale climate variability shapes the atmospheric transport of pollutants, and may shift under climate change as the frequencies and intensities of climate patterns change (e.g. Gillett and Fyfe, 2013). Under present climate conditions, net import fluxes into the Arctic are declining for legacy POPs (Hansen et al., 2015; Wöhrnschimmel et al., 2016; Carlsson et al., 2018), but a rebounding of the net import flux of DDT is forecast under future climate conditions (Schenker et al., 2008c; Octaviani et al., 2015).

The global fate and transport of PCB-153 and α-HCH were modeled to compare a historical 20th century climate scenario (20C3M) with the Special Report on Emissions Scenarios A2 (SRES-A2) climate scenario (Wöhrnschimmel et al., 2013). Concentrations of PCB-153 in Arctic air were higher by a factor of 2 in the year 2100 under the SRES-A2 scenario compared to the 20th century scenario, with the effect of temperature on primary and secondary emissions accounting for most of the increase. For PCB-153, reduced ice cover in the Arctic Ocean under the SRES-A2 scenario led to slightly lower concentrations in air, and higher concentrations in the Arctic Ocean relative to the 20th century scenario because of enhanced air-ocean deposition. In contrast, for α-HCH, concentrations in the Arctic atmosphere in the year 2100 under the two scenarios were similar, but concentrations in the Arctic Ocean were forecast to be lower by a factor of 3 due to a higher net volatilization facilitated by a lack of ice coverage and changing ocean circulation patterns. In these scenarios for PCB-153 and α-HCH, climate change-induced differences in air flow and precipitation patterns did not strongly affect modeled concentrations in Arctic air or water.

The same model was used to compare modeled concentrations of non-ionizable organic chemicals spanning the range of plausible combinations of solubilities in air, water and octanol in Arctic air and ocean water under the 20C3M and SRES-A2 scenarios (Figure 2.1.2) (Wöhrnschimmel et al., 2013). Concentrations of chemicals with degradation half-lives set at the criteria values for persistence established under the Stockholm Convention and the Convention on Long-range Transboundary Air Pollution (CLRTAP) were uniformly



Chemical classification (according to Gouin and Wania)		Dominant fate process in climate change
A/E	volatile flyer	temperature ↑ ⇒ degradation ↑
B/F	volatile water-soluble multiple-hopper	sea ice ↓ ⇒ volatilization from Arctic Ocean ↑
C	borderline-persistent multimedia multiple-hopper	temperature ↑ ⇒ degradation ↑
D	borderline-persistent single hopper	temperature ↑ ⇒ degradation ↑
G	highly-persistent multimedia multiple-hopper	temperature ↑ ⇒ volatilization from surface and particles ↑ ⇒ atmospheric transport to the Arctic ↑ sea ice ↓ ⇒ deposition into Arctic Ocean ↑
H	highly-persistent single hopper	sea ice ↓ ⇒ deposition into Arctic Ocean ↑

Figure 2.1.2 Ratio of modeled average pollutant concentrations in the Arctic atmosphere (left column) and ocean water (right column) for the period 2080–2100 under the SRES-A2 climate change scenario and an industrial emission scenario that includes enhanced economic activity in the Arctic, compared to 20th century climate and baseline emissions. The top panels are chemicals with half-lives set at the persistence cut-off values defined in the Convention on Long-Range Transboundary Air Pollution (CLRTAP) and the bottom panels are chemicals with half-lives set at values typical of highly persistent POPs. Warm colors (yellow and red) indicate higher concentrations in the Arctic under the climate change scenario, and cool colors (blue and purple) indicate lower concentrations. Dominant processes that give rise to the differences between the climate change scenario and the baseline scenario for the selected chemicals A–H are summarized in the table. Adapted from Wöhrnschimmel et al., 2013.

lower under the SRES-A2 climate change scenario due to enhanced degradation at higher temperatures. For highly persistent chemicals with half-lives selected to represent highly persistent POPs, higher concentrations by up to a factor of 1.7 in the atmosphere and up to factor of 4 in the Arctic Ocean were forecast under SRES-A2. The dominant fate processes that account for the differences in the SRES-A2 climate change scenario relative to the baseline 20C3M scenario are indicated in the table in Figure 2.1.2.

2.1.4.1 What is unknown

All models are simplifications of real systems. POP models in particular usually lack the capacity to account for potential non-linear feedbacks (positive and negative) between POPs and other climate change effects. As time series of monitoring data for POPs in the Arctic and globally are extended and expanded, data-analysis techniques such as blind signal separation, independent component analysis and machine learning (e.g. Ameri et al., 2002) might be applicable to the extraction of climate change signals from POP time series data. Data-focused approaches such at these could potentially generate new hypotheses about mechanistic links between POPs and climate change that could be incorporated in the next generation of POP models.

There is ongoing interest in a potential climate "tipping point" in the Arctic connected to an abrupt increase in the amplitude of seasonal variability of sea ice area in the mid-2000s (Livina and Lenton, 2013). This abrupt change, and others occurring over the past two decades, may be linked to "step changes" in POP concentrations in the Arctic (Zhao et al., 2015), but need to be further verified by extended time series of *in situ* measurement data.

2.1.4.4 Implications

Our understanding of current POPs and the environmental changes forecast to result from climate forcing indicate that transport pathways of POPs to the Arctic will be affected by climate change, but that the effect is small relative to uncertainties in the models and small relative to the effect of changes in emissions that could occur over the same time period. There are some cases where actions on climate change will also help mitigate chemical transport to the Arctic (Friedman et al., 2014), but in general, chemical exposure should be managed independently of consideration of climate effects.

2.1.5 How well can we anticipate how POPs and CEACs will impact the Arctic in a changing future climate?

2.1.5.1 What is known

Current models indicate that emissions are the dominant factor determining exposure of the Arctic environment, biota and human populations to POPs. Modeled scenarios for PCB-153 and α-HCH (Wöhrnschimmel et al., 2013) forecast that emission reductions would drive concentration declines of these POPs in Arctic air and ocean water by factors between one thousand and one million by the year 2100. The estimated reductions in POP concentrations from lowered emissions are much larger than the concentration changes of less than a factor of four that are attributable to changing climate.

2.1.5.2 What is partially known and/or uncertain

There are inherent uncertainties in modeling POPs and CEACs in the environment. With regards to POPs, models of PCBs, α-HCH and DDT are the most constrained in terms of knowledge of their emissions, physical-chemical properties and fate processes. However, other POPs and CEACs with combinations of properties that have not been well studied, are subject to high uncertainties (e.g. per- and polyfluoroalkyl substances (PFAS) and organophosphate ester (OPE) flame retardants and plasticizers).

2.1.5.3 What is unknown

Our expectations about how climate change might affect the transport of chemicals to the Arctic is built upon experience with well-studied legacy POPs and model scenarios constrained by forecasts of climate conditions under possible greenhouse gas emissions scenarios. New POPs with environmental fate profiles that cannot be extrapolated from current knowledge will not conform to these model scenarios. Similarly, the potential effects of regime shifts in climate conditions that result from transgressing "tipping points" in the climate system are not considered in current models.

2.1.5.4 Implications

For persistent substances that cause poorly-reversible global exposure, a precautionary approach to chemical management is prudent, especially given that these substances can pose a range of hazards that are difficult to anticipate (Persson et al., 2013; MacLeod et al., 2014; Cousins et al., 2016, 2019), and could be compounded by deficiencies in our understanding of how climate change and chemical stresses might interact.

2.1.6 Conclusions

The modeled impacts of climate change on concentrations of chemicals in the Arctic are small when compared to the uncertainties inherent in modeling concentrations of POPs in the global environment, and much smaller than the effects that can be achieved by reducing emissions over comparable timescales. Our current understanding of POPs and other global pollutants, and the environmental changes related to climate forcing, indicate that although transport pathways of POPs to the Arctic will be affected by climate change, it is the amount and location of emissions in combination with the properties of a chemical that will predominantly determine its potential to pollute the Arctic. Although there are some potential co-benefits from actions on climate change that can also help mitigate chemical transport to the Arctic, in general, chemicals with the capacity to undergo global-scale transport should be managed independently of consideration of climate effects. A precautionary approach to the management of

persistent chemicals with the potential to be global-scale pollutants is prudent because the hazards associated with these chemicals are difficult to anticipate and they cause poorly-reversible global exposure, and thus potentially also poorly-reversible effects.

Environmental exposure to chemicals and the resultant exposure of biota and humans to chemicals through the environment, are driven by primary emissions, so success in mitigating exposure should be measured by the reduction of emissions. Highly uncertain and missing information about POP and CEAC emissions is one of the key factors limiting capabilities to model exposure levels and trends in the Arctic.

Models link emission inventories to measurements and can be used to demonstrate a quantitative understanding of source-to-receptor concentration relationships over distance and time. Models combined with emission estimates and measurements in an integrated approach are a powerful framework for understanding global chemical pollution. Emissions, environmental levels and models all have associated uncertainties, but often emissions are the most uncertain component, and the emission estimates needed to drive models may be lacking entirely for newer POPs and CEACs. Applying models to analyze scenarios that are within the range of uncertainty for emissions, chemical properties and process parameterizations can generate testable hypotheses to guide targeted measurements of POPs in the environment and efforts to refine emission inventories. Integrative approaches that require collaboration across disciplines and agencies have proven to be an important tool in effectiveness evaluations of the Stockholm Convention (Harner et al., 2015).

2.1.7 Knowledge gaps and recommendations

Research that combines emission estimates, models and measurements in an integrated approach towards understanding chemicals in the global environment should be encouraged since it can provide the basis for formulating testable hypotheses about key drivers of chemical fate and transport and supports effectiveness evaluations of the Stockholm Convention. Such studies also provide the basis for scenario analyses that can forecast the levels and trends of global pollutants expected under alternative scenarios for climate and chemical use and emissions.

Improving primary emission inventories for POPs and CEACs should be a high priority because of their capacity to unlock the potential of integrated approaches. Studies that use "bottom-up" approaches, particularly those with integrative, lifecycle perspectives to estimate emissions from information about the production, use and disposal of chemicals, should be encouraged. However, given the difficulties associated with carrying out "bottom-up" studies, research that uses a "top-down" approach should also be employed. In "top-down" approaches, models are first fitted to measurement data to estimate emissions, then those emission estimates are evaluated in model scenarios that can be validated against independent measurement data.

Modeling approaches could more directly and effectively contribute to understanding the influence of climate change on pathways of pollutant transport to the Arctic through future efforts to:

- Model Arctic pollution using scenarios and emission projections that reflect sources likely to be strongly affected by climate change, such as emissions from thermal sources, shipping, and other local sources in the Arctic.
- Distinguish between, and estimate the relative proportion of Arctic pollution from long range, regional and local sources (i.e. relative source attribution).
- Evaluate trends in POP and CEAC deposition to the Arctic, including the contribution of major emission sources to the region (e.g. Task Force on Hemispheric Transport of Air Pollution (TF HTAP) source-receptor modeling exercises).
- Reduce uncertainties in physical-chemical properties of POPs and CEACs, particularly when they impede estimating emissions and contribute to high uncertainties in modeling.
- Conduct process-oriented research studies of POPs and CEACs to support improved model performance for key processes, including gas-particle partitioning, degradation and surface-air exchange.
- Use sensitivity model simulations to evaluate the complex influences of climate-related changes in atmospheric composition and meteorological conditions on contaminant transport and fate, including non-linear effects, feedbacks, extreme weather events, and tipping points at local, regional, and global scales.
- Sponsor and organize emission and LRT modeling capacity-building workshops for scientists and educators.

2.2 Physical environment: levels and trends

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2.2.1 Introduction

How can the effects of climate change on contaminants in the Arctic act as early warning signals for impacts on contaminants globally?

The Arctic environment is vulnerable to global environmental changes. The Arctic cryosphere (marine and terrestrial) is rapidly declining with observable and documented impacts on Arctic ecosystems (Comiso, 2012; Comiso and Hall, 2014; Gremillet et al., 2015; AMAP, 2016a; Biskaborn et al., 2019; Yumashev et al., 2019). According to the most recent assessment by the Intergovernmental Panel on Climate Change (IPCC, 2019), warming occurs faster and with greater magnitude in the Arctic compared to other parts of the world. Climate change in the Arctic is resulting in extreme sea ice retreat, melting glaciers, thawing permafrosts and a greater frequency of extreme weather events (Cohen et al., 2014). Such changes also affect the environmental distribution and fate of contaminants such as persistent organic pollutants (POPs), Figure 2.2.1. Effects on contaminants occur not only because of changes in Arctic surface conditions (e.g. increased open water area, loss of glaciers, perturbation of snow deposition patterns) and physical processes (e.g. air and water circulation patterns, precipitation rates), but also because the most important drivers of POP transport, partitioning, and transformation are their physical-chemical properties, many of which are temperature-dependent. Temperature-dependent properties of POPs include their vapor pressure, Henry's Law constant, partitioning coefficients, and rate of degradation or transformation (including photolysis, hydrolysis, etc.) (Ma et al., 2016). Lamon et al. (2009) estimated that a 1°C increase as a result of climate change would increase the volatility of POPs such as polychlorinated biphenyls (PCBs) by 10 to 15%, and thus, increase their mobility. Other biogeochemical processes

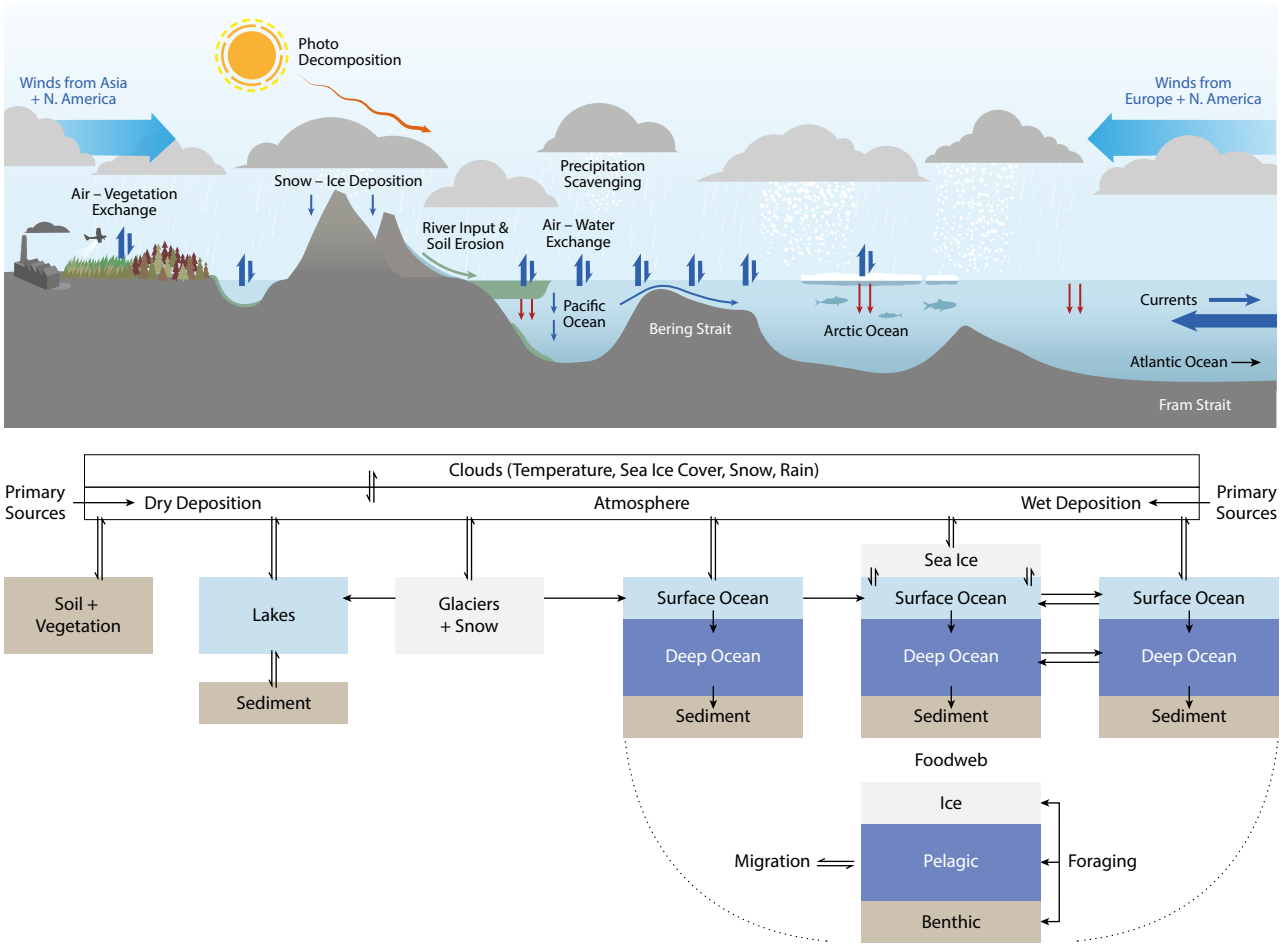


Figure 2.2.1 Simplified summary of climate change influences on transport and transformation processes of POPs in the abiotic environment of the northern hemisphere. See also Figure 2.1.1. Modified from Ma et al. (2016).

Figure 2.2.2 Locations of key studies on the Arctic physical environment referenced in this chapter.

affecting POPs, such as the 'biological pump' and degradation in marine waters (Galbán-Malagón et al., 2012, 2013), also show climate sensitivity.

As a result of climate teleconnections between the Arctic and other parts of the world, the amplified effects of climate change in the Arctic will contribute to higher sea levels and more intense and frequent extreme climate events (e.g. heavy rainfalls, droughts, severe storms) globally (Cohen et al., 2014; Kusangaya et al., 2014; McBean and Ajibade, 2014). Together with generally increasing global surface temperatures and decreasing ice and snow cover, these climate-related changes will also serve to mobilize and redistribute POPs in the environment of more southerly latitudes. Thus, the amplified effects of climate change on contaminants in the Arctic can provide early warning signals for similar effects globally.

Many of the processes responsible for the distribution and transformation of POPs can be influenced by climate change. Figure 2.2.1 summarizes how a changing climate may influence the environmental distribution of POPs in the northern hemisphere, with a focus on abiotic processes. The impacts of climate change on biogeochemical processes can often have opposing effects on POPs. For example, ambient temperature increases favor the volatilization of chemicals, thus enhancing their mobility in the atmosphere and potential for long-range atmospheric transport (LRAT). On the other hand, increases in precipitation enhance the scavenging, deposition, and removal of chemicals from the atmosphere.

Climate change occurs over long periods of time (i.e. decades), therefore its effects on POPs need to be evaluated over similar time scales. Here we attempt to make use of available temporal and spatial measurements of POPs in air, water, oceanic- and lake- sediment cores, and ice- and snow- cores to provide insight into whether the cumulative impacts of climate change within the Arctic exacerbate or diminish contaminant transport and accumulation patterns in different abiotic media. Figure 2.2.2 shows the locations of key studies on the physical environment reviewed in this chapter. In addition to POPs, we will also explore the effects of climate change on chemicals of emerging Arctic concern (CEACs), identified by the Arctic Monitoring and Assessment Programme (AMAP) as chemicals newly detected in the Arctic environment (AMAP, 2017a). Given the relatively recent discovery of CEACs, less is known about their sources and environmental fate; however, it is expected that climate change will influence the presence and fate of CEACs similarly as for POPs.

2.2.2 Direct and indirect effects of climate change on contaminants in the Arctic

Does climate change within the Arctic exacerbate or diminish contaminant transport, accumulation, and occurrence in different abiotic media (i.e. physical environmental compartments)?

Changes in ecosystem processes and structure (e.g. food webs, deposition pathways, cryosphere, etc.), as well as

Table 2.2.1 Direct and indirect influences of climate change on the fate and presence of pollutants in the Arctic.

Influence of climate change	Contaminant-related processes potentially affected
Direct influences	
Increased ambient temperatures (sea, land, atmosphere)	• Pollutant re-mobilization • Long-range transport • Transformation pathways
Ocean acidification	• Transformation conditions
Changing weather patterns (e.g. precipitation, seasonal characteristics, frequency of extreme events)	• Long-range atmospheric transport • Deposition and precipitation event frequency
Sea level rise	• Transfer between terrestrial and marine environments • Marine transport pathways
Loss of cryosphere	• Translocation, re-mobilization, and redistribution of contaminants
Different radiation characteristics (e.g. black carbon, cloud condensation nuclei)	• Photochemistry and transformation pathways
Altered carbon cycling and sequestration	• Biotic and abiotic transformation
Water mass transport changes (e.g. changing ocean currents)	• Long-range oceanic transport
Increased dust aerosol loadings	• Additional advective transport and particle-mediated transport
Indirect influences	
Food web composition changes (e.g. invading species)	• Bioaccumulation and transformation
Re-mobilization of pollutants	• Re-emission from sediment, ice surfaces, and soils
Land degradation (e.g. increased erosion)	• Re-mobilization and re-emission • Transfer between terrestrial and marine environments
Biodiversity loss	• Bioaccumulation and transformation
Behavioral pattern changes (e.g. animal migration)	• Contaminant exposure • Bioaccumulation • Biovector-based transport of pollutants
Human socio-economic development	• New pollutant sources • New exposure routes
New economic opportunities in the Arctic	• New pollutant sources • Re-mobilization
Increased agricultural diseases and pests	• Pesticide and pest control agent use
Increased frequency of boreal forest fires	• Pollutant re-mobilization and redistribution
Effects on the oceanic biological pump	• Ocean sequestration of pollutants

thermodynamically-driven interactions between chemicals and their surrounding environment, will inevitably influence the presence, lifetime, and mobility of chemicals in Arctic ecosystems – either directly or indirectly. Climate change directly affects contaminants by altering the physical environment, such as increasing ambient air and water temperatures or changing physical processes in the Arctic (e.g. retreating ice, melting permafrost, increasing precipitation, erratic warming events, changing river flows, etc.), but also indirectly influences contaminants through the secondary changes and consequences that result from an altered physical environment. Examples of indirect effects include new or different pollutant sources associated with changes or shifts in anthropogenic activities within the Arctic (e.g. shipping, mineral exploitation), or variations in biological processes (e.g. changes in the magnitude and extent of the oceanic biological pump driving contaminants to deeper waters). Direct and indirect effects of climate change relevant to the presence and distribution of contaminants in the Arctic are summarized in Table 2.2.1.

The direct effects of climate change on the presence and profiles of various organic (and inorganic) pollutants have been previously documented (Dalla Valle et al., 2007; Rajkumar et al., 2013; Vione and Scozzaro, 2019; Wilson et al., 2019). However, under the current, most accepted climate change scenarios forecast for the Arctic, it is expected that the indirect effects of climate change will have a significantly stronger effect on the environmental fate of contaminants compared to direct influences (AMAP, 2011a; Kallenborn et al., 2012; Pacyna et al., 2015; AMAP, 2016a; Ma et al., 2016; Carlsson et al., 2018). Compound-specific environmental factors like environmental stability, transformation, bioavailability, and environmental mobility can be monitored in future studies aimed at characterizing and summarizing the effect of climate change on the presence and behavior of chemical contaminants in the Arctic, bearing in mind that such factors may vary geographically across the region (AMAP 2011a, 2016; Kallenborn et al., 2018; Carlsson et al., 2018).

Table 2.2.2 Relevant groups of chemicals of emerging Arctic concern (CEACs) identified in AMAP (2017), many of which have little information existing on their occurrence or transport pathways within and outside the Arctic. Some chemicals originally identified as CEACs have since been listed as POPs under the Stockholm Convention. Refer to Table 1.1 for updated designations.

CEAC group	Abbreviation	Characteristic compounds	Main pathways to the Arctic		
			Long-range transport	Local sources	Notes
Per- and polyfluoroalkyl substances	PFAS	perfluoroalkyl acids (PFAA) including perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexane sulfonic acid (PFHxS); volatile neutral PFAA precursors including fluorotelomer alcohol (FTOH) and perfluoroalkylsulfonamide-based substances	X	X	Local communities and airports may act as local sources
Brominated flame retardants	BFRs	decabromodiphenylether (PBDE-209)	X	X	Dump sites may act as local sources
Chlorinated flame retardants	CFRs	dechlorane plus	X	X	Dump sites may act as local sources
Organophosphate ester-based flame retardants and plasticizers	OPEs	chlorinated OPE such as tris(2-chloroethyl) phosphate (TCEP); alkylated OPE such as tri-*n*-butyl phosphate (TnBP), tris meta-(cresyl) phosphate (TmCP); aryl-OPE such as 2-ethylhexyl diphenyl phosphate (EHDPP)	X	X	Airports may act as local sources
Phthalates		diethylphthalate	X	X	
Short-chain chlorinated paraffins	SCCPs	$C_{10}H_{17}Cl_5$	X	X	
Siloxanes		hexamethyldisiloxane (HMDS), decamethylcyclopentasiloxane (D_5)	X	X	Atmospheric transport is a major source to the Arctic, however, local sources (e.g. personal care product use) also exist
Pharmaceuticals and personal care products	PPCPs	ibuprofen, caffeine		X	Sewage outflows are primary local sources
Polychlorinated naphthalenes	PCNs	halowax	X	X	
Hexachlorobutadiene	HCBD	HCBD	X		
Current-use pesticides	CUPs	chlorpyrifos, chlorothalonil, dacthal	X	X	Agricultural applications are primary sources
Pentachlorophenol and pentachloroanisole	PCP and PCA	PCP and PCA	X	X	
Organotins		R_nSnX_{4-n}, where R represents an alkyl or aryl group and X is represented by an anion such as chloride, oxide, hydroxide, acetate, or other functional group, e.g. tributyltin (TBT).		X	Use associated with population and shipping densities. Harbors are major local sources
Polycyclic aromatic hydrocarbons	PAHs	napthalene, anthracene	X	X	Summer wildfires in sub-Arctic regions act as episodic sources
Unintentionally produced PCBs	uPCBs	PCB-11	X	X	
Halogenated natural products	HNPs	brominated phenols, mixed halogenated compounds	X	X	Algae blooms are primary sources
Marine plastics and microplastics	-	polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), including monomers and additives	X	X	

As many CEACs are directly associated with human activities, they have the potential to originate from within the Arctic region itself. AMAP (2017) identified 17 CEAC groups, of which approximately 11 may originate from local sources in addition to long-range transport (LRT) (Table 2.2.2). Additionally, Röhler et al. (2020) developed a sample clean-up and analytical method to screen air samples for new chemicals of concern in Arctic air. In air collected from the Zeppelin station on Svalbard (Figure 2.2.2) they found 700+ and 1200+ chemicals of interest in the particle and gas phase fractions, respectively, including 73 new potential CEACs (i.e. compounds previously not reported in Arctic environments). There is very little information about the occurrence and transport pathways of many CEACs; thus, it is difficult to determine how climate change will affect their distributions in the Arctic environment. Better knowledge of the sources, environmental fate, and impacts of climate change on CEACs is required to coordinate pollutant regulations on a circum-Arctic scale (AMAP, 2017a).

2.2.3 Within-Arctic redistribution of POPs and CEACs

What are the most sensitive transfer processes that affect the movement of contaminants between polar reservoirs?

Given the semi-volatile nature of POPs and many CEACs, transfer of contaminants between environmental compartments (e.g. air to surfaces, water to sediments, ice to water, etc.) will be affected by changes in ambient temperature and Arctic processes, such as snow, rainfall, and the changing cryosphere. Re-mobilization of legacy POPs present in Arctic surface seawater back to the atmosphere due to the decline in sea ice cover was examined by Ma et al. (2011). By combining empirical monitoring data and model-based scenario assessments, this study showed that the physical-chemical properties of the respective substances combined with the diminishing cryosphere is resulting in the re-emission of contaminants from Arctic surface compartments (i.e. 'reservoirs' such as snow, ice, soil, sea water) and influencing longer-term time trends of selected POPs in the Arctic atmosphere.

The Arctic marine environment (e.g. marine sediments) is considered an important depositional sink for legacy POPs in the Arctic, largely due to the settling of organic matter-bound POPs through the biological pump operating in oceanic surface waters (Galbán-Malagón et al., 2012). For most of the substances addressed in this report, the Arctic marine environment and associated food webs are the largest environmental reservoirs due to the areal extent of the Arctic Ocean and coastal seas relative to terrestrial areas. Therefore, the loss of the terrestrial cryosphere is not expected to significantly contribute to the diffusive re-mobilization of legacy POPs on an Arctic-wide basis (Rigét et al., 2019). Wöhrnschimmel et al. (2013) examined the temporal profiles of PCBs and their responses to Arctic environmental changes. This comprehensive study combined a chemical fate model, scenario assessments, and empirical data evaluations in the context of a changing Arctic. Based on these evaluations, the authors concluded that long-range atmospheric transport from potential primary sources (e.g. fresh usage) and secondary sources (e.g. volatilization from soil), rather than secondary re-emissions from Arctic compartments, will continue to be the major inputs of PCBs and other POPs into the Arctic environment in the future.

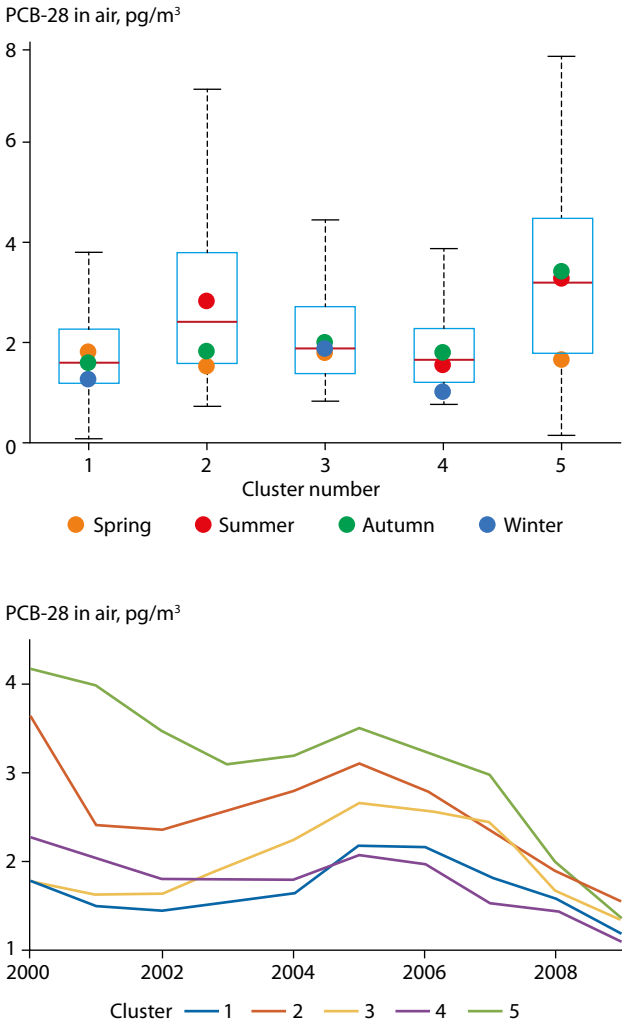


Figure 2.2.3 PCB-28 concentrations in air from Zeppelin station on Svalbard classified by origin of air masses (land-derived: clusters 1,3, and 4; ocean-derived: clusters 2 and 5). (upper) Box plot of PCB-28 concentrations in air. Boxes show interquartile range with whiskers that extend to the most-extreme values; colored dots indicate median concentrations per season. Concentration variability and seasonal variation were highest for ocean-derived transport regimes compared to land-derived transport regimes originating from continents. PCB-28 concentrations in cluster 2 were highest during the summer (red dot). (lower) Running median (4 years) of PCB-28 concentrations by cluster between 2000 and 2009. Highest concentrations of PCB-28 were observed when air masses originated from oceans. Modified from Ubl et al. (2017).

More recently, Ubl et al. (2017) used a cluster analysis to study the relationship between atmospheric transport regimes and PCB concentrations in air measured at Zeppelin station on Svalbard. They found that the concentrations of less-volatile PCBs (e.g. PCB-101) were higher during the winter with land-derived air masses originating from Europe. However, air concentrations of lighter PCBs (represented by PCB-28) were higher during the summer, and were associated with air masses of an oceanic origin and longer residence times closer to the station (Figure 2.2.3). They attributed this observation to the increased emissions from local PCB sources (e.g. PCBs in paints, mines, and heavily contaminated soil) during the warm season, which masked the effect of LRT. In a warming

Arctic, emissions from local environmental reservoirs will likely become more important for more volatile chemicals (e.g. PCB-28).

In the light of a rapidly changing environment, internal Arctic redistribution and transformation processes will be more prominent in shaping the environmental fate of legacy POPs, such as PCBs, through the following processes:

1. Wet deposition will occur more often in the form of rain instead of snow, which will have effects on the scavenging effectiveness and deposition profile of POPs present in the atmosphere.
2. Microbial transformation will play a more prominent role in the degradation of POPs. Climate change will modify the structure of microbial communities and the rate of microbial degradation.
3. Transarctic ice transport will be less important for the marine redistribution processes of POPs, particularly after riverine release of POPs.
4. Changes in food web composition will contribute to changes in environmental partitioning and accumulation of POPs, as well settling fluxes of organic matter-bound chemicals.
5. Increasing ablation and melt of ice caps and glaciers will continue to be a source for POP re-mobilization in the Arctic environment (Steinlin et al., 2014; Wang et al., 2019). Legacy POPs with physical-chemical properties similar to PCBs may be expected to exhibit similar fate characteristics.

Increased re-mobilization of legacy POPs from marine reservoirs due to accelerating Arctic environmental change and marine cryosphere loss, in combination with complex inner-Arctic redistribution processes, may add to the overall POP levels and potential exposure risk for marine food webs and indigenous populations.

2.2.3.1 Effects of warming and biogeochemical change on air-water-sediment exchange

The decrease in sea ice coverage appears to be enhancing primary production in the Arctic Ocean due to the increased light entering the sea, longer growing season, and increased nutrient availability from marginal sources. Climate change will also modify the seasonal timing of phytoplankton blooms in the Arctic (Wassmann, 2011). Massive phytoplankton blooms underneath regions fully covered by sea ice have been observed, suggesting blooms may be initiated whenever light and nutrients are sufficient for photosynthesis, independent of the timing of ice retreat (Arrigo et al., 2012; Lowry et al., 2014). Since many POPs and CEACs are hydrophobic with a tendency to associate with organic matter, after entering surface seawater these chemicals sorb to particles and are transported to the deep ocean via vertical sinking, resulting in increased sedimentation of chemical contaminants in a process driven mainly by the biological pump operating in near-surface waters (Galbán-Malagón et al., 2012). This process also draws dissolved chemicals out of the seawater, inducing an air-water fugacity gradient that enhances their flux from air to water (Jurado and Dachs, 2008; Kuzyk et al., 2010; Berrojalbiz et al., 2011; Galbán-Malagón et al., 2012). In addition, the ocean sequestration of atmospheric POPs also modifies or can modulate the transport of POPs to the Arctic. Under a climate change scenario, the activity of the biological pump will change in magnitude and geographic extent. High-productivity regions and their large settling fluxes of organic matter are expected to shift northward to Arctic regions currently covered by sea ice (Wassmann, 2011), thus leading to a larger sequestration of POPs in deep Arctic seawater and sediments. Liu et al. (2021) reported the full-depth profiles of polycyclic aromatic hydrocarbon (PAH) concentrations in sea water from the Pacific to the Arctic and Atlantic Oceans at 44 sites measured on-board the RV Xuelong (Snow Dragon) research expedition from July to September 2012. PAHs in the water columns generally showed a "surface enrichment and depth-depletion" pattern. Using particle-reactive radioisotopes, Liu et al. (2021) showed high scavenging efficiency, with PAH sinking fluxes of 2773±1259 ng/m^2/d. Lateral transport, as traced by transient tracers, did not show a significant contribution to the PAH inventories in the Arctic surface ocean. Their findings indicate the importance of biogeochemical-modulated contaminant transport, which would be enhanced due to climate change.

Another important biogeochemical process influencing contaminant exchange is the degradation of organic pollutants by microorganisms. For example, microbial degradation of hexachlorocyclohexanes (HCHs) has been reported for Arctic waters (Harner et al., 2000; Galbán-Malagón et al., 2013), and occurs at a higher magnitude during bloom events that is sufficient to induce a depletion of dissolved phase HCH concentrations and drive atmospheric deposition of HCHs. Other organic pollutants besides HCHs may be microbially-degraded in the Arctic Ocean, and this process could be favored under a climate change scenario with higher temperatures. On the other hand, organic pollutants exert an influence on microbial community structures and functions that could impact degradation. For example, Cerro-Gálvez (2019) has shown that inputs of organic pollutant mixtures to coastal waters from Svalbard increase growth of the microbial biosphere and induce a transcriptomic response related to the degradation of organic pollutants and other detoxification processes.

2.2.3.2 Gas-particle partitioning and climate change

Volatilization rates as well as gas-particle partitioning are two of the key factors that determine the environmental fate of semi-volatile organic chemicals (SVOCs), including POPs and many CEACs. Gas-particle partitioning and volatilization rates of SVOCs can be estimated from their octanol-air partitioning coefficients (K_{OA}) and vapor pressures (Bidleman, 1988; Pankow, 1998), both of which are temperature-dependent, and therefore likely to be impacted by climate change.

Wei et al. (2016) reported SVOCs with phase-change enthalpies between 50 KJ/mol and 100 KJ/mol exhibited an exponential increase in gas-phase concentrations in response to temperature increases. The majority of PCBs, polybrominated diphenyl ethers (PBDEs), organophosphate ester-based flame retardants

and plasticizers (OPEs), PAHs, dioxins, and furans fall in this range (Acree and Chickos, 2010). Gas-phase concentrations of these SVOCs decreased by an average of 7% with a 1°C reduction in temperature but increased by an average of 50% with a 1°C increase of temperature (Wei et al., 2016).

In addition to phase-change enthalpies, particle concentrations in air seem to be an important driver for partitioning of organic contaminants onto particles (Bidleman, 1988; Sühring et al., 2016a; Kondo et al., 2018). Elevated coarse particle loads at lower temperatures, e.g. in the Arctic (Götz et al., 2008) were found to lead to higher OPE fractions on particles (Sühring et al., 2016a). Consequently, the higher average temperatures due to climate change could result in a higher fraction of OPEs being found in the gas-phase.

It is not clear whether an increased gas-phase concentration of OPEs (and other SVOCs) would lead to an increase or decrease in atmospheric transport compared to particle-associated long-range transport. An increase could occur, because larger particles (and the contaminants sorbed to them) are scavenged by wet and dry deposition, which reduces the overall travel distance of the particle and any sorbed contaminants (Thuens et al., 2014). Higher gas-phase concentrations could therefore reduce the removal of contaminants from the atmosphere, allowing them to travel further. This hypothesis has been supported by model predictions of PAH concentrations in the Arctic under future climate change scenarios. Using a chemical transport model (GEOS-Chem), Friedman et al. (2014) predicted increasing concentrations of volatile PAHs and decreasing concentrations of particle-bound PAHs under climate change conditions. For soil/snow-air partitioning, Casal et al. (2018a) demonstrated that PCB and HCH concentrations show temperature-dependent variation; however, such seasonality is not observed for high molecular weight PAHs. On the other hand, particle adsorption can reduce degradation of contaminants (Zhou et al., 2012). Therefore, higher gas-phase concentrations could also lead to increased degradation (e.g. photochemical oxidation) and, thereby reduce the potential for long-range atmospheric transport, which has been discussed for decabromodiphenylether (PBDE-209) (Möller et al., 2011).

2.2.3.3 Sea-spray aerosols and their influence on contaminant transport

Sea spray aerosols (SSAs) are droplets of sea water that are ejected into the atmosphere by the bursting of bubbles on the sea surface. The bubbles, mostly formed by breaking waves, fragment into small "film droplets" (SSAs < 1 µm) and/or burst, releasing larger "jet droplets" (SSAs > 1 µm) (Spiel, 1994; Wang et al., 2017b). The formation of SSAs is a complex process that is highly dependent on environmental parameters such as air temperature, water temperature, and wind speed (Lewis and Schwartz, 2004). The bubbles that ultimately lead to the formation of SSAs have been shown to enrich organic matter, and with it, persistent and mobile chemicals such as per- and polyfluoroalkyl substances (PFAS), OPEs, and other organic compounds that are present in the water column (McMurdo et al., 2008).

Johansson et al. (2019) reported a 62000-fold enrichment of PFAS concentrations in SSAs compared to bulk water in a sea-spray chamber experiment. They concluded that SSAs were a significant source of PFAS to the atmosphere. Using the Norwegian Earth System Model (NorESM), Johansson et al. (2019) estimated the global emissions and transport of PFAS due to SSAs. They concluded that PFAS emissions through SSAs would be highest in latitudes between 45°N and 60°N, and 45°S and 60°S, respectively (Figure 2.2.4). Casal et al. (2017) have shown that SSAs can be an important source of PFAS to Antarctic waters when scavenged during snow deposition events. Such a process could be important for the Arctic Ocean as well.

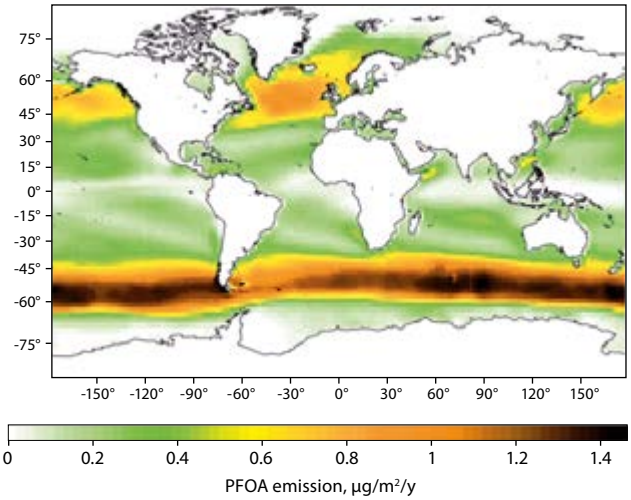


Figure 2.2.4 Spatial depiction of total predicted yearly emissions of perfluorooctanoic acid (PFOA) via sea spray aerosols. Source: Johansson et al. (2019).

Since the formation and size of SSAs are highly dependent on environmental factors, climate change could have a substantial impact on their role as vectors for PFAS and other organic chemicals to the atmosphere. However, the complex interactions of water temperature, air temperature, wind speed, and salinity in SSA formation mean it is not possible to draw global conclusions regarding the impact of climate change on SSAs and their role in contaminant transport.

The reduction of sea ice due to climate change is expected to lead to a general increase in SSAs within the Arctic (Struthers et al., 2011). However, laboratory studies by Mårtensson et al. (2003) and in situ measurements by Nilsson et al. (2007) showed that increasing water temperatures could lead to a decrease of nano-sized SSAs, which have been shown to have the highest enrichment potential for organic matter (Keene et al., 2007), and by extension, organic contaminants potentially as well. For larger SSAs (> 100 nm), the same experiments showed an increase of the number of SSAs with increasing seawater temperature (Mårtensson et al., 2003; Nilsson et al., 2007).

Model predictions for Europe by Soares et al. (2016) suggest an increase of sea salt emissions into the air north of Iceland and around the United Kingdom under climate change scenarios. These findings are congruent with the regions predicted to have elevated PFAS emissions associated with SSAs (Johansson et al., 2019) (Figure 2.2.4), suggesting that SSA-related emissions of organic chemicals, such as PFAS, could further increase in the Arctic due to climate change.

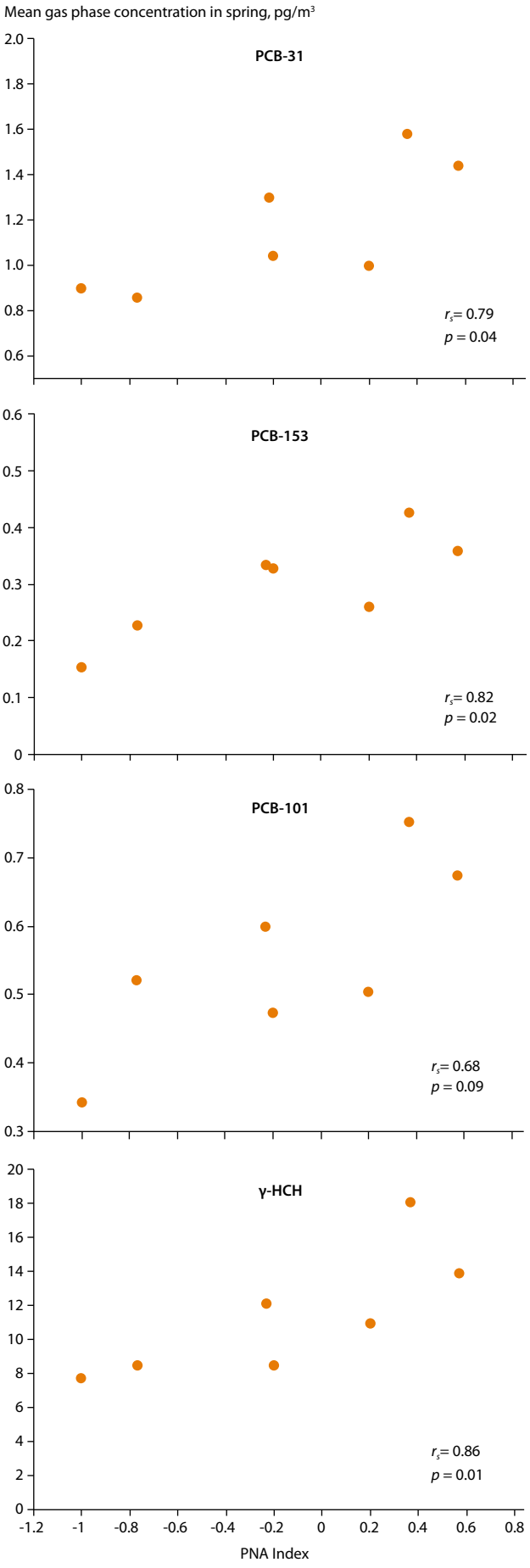


Figure 2.2.5 Correlations between the Pacific North American (PNA) climate index and spring air concentrations of PCBs and γ-HCH measured at Alert, Canada between 1993-1999. Source: Hung et al. (2005).

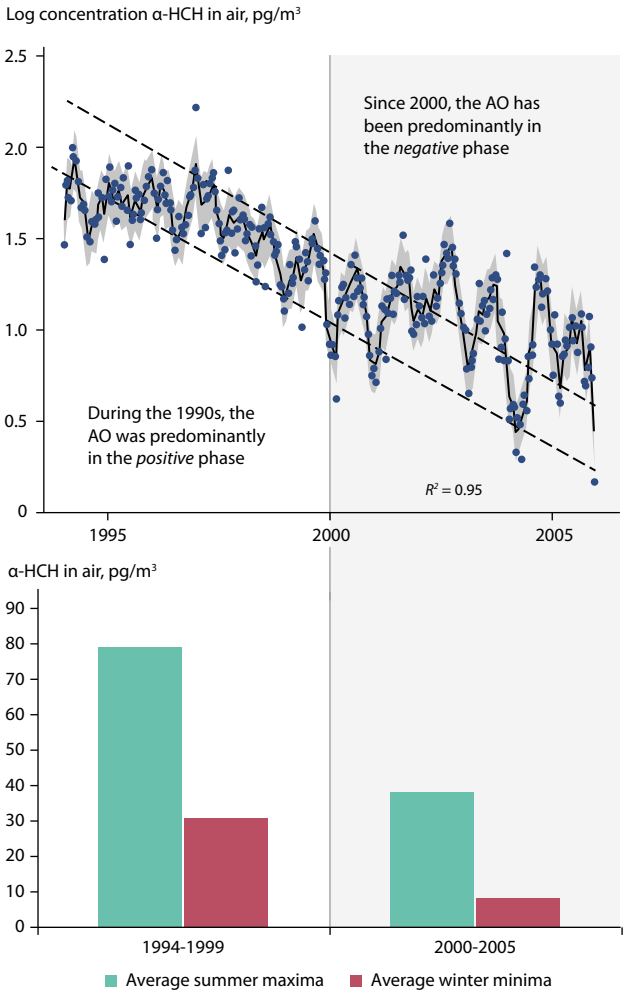


Figure 2.2.6 Arctic oscillation (AO) influence on α-HCH air concentrations measured at Zeppelin Mountain, Svalbard. (upper) Bi-weekly measurements of α-HCH in air between 1994-2006. Dashed lines indicate expected differences between summer and winter concentrations, assuming constant decline. (lower) Difference in average summer- and winter- α-HCH air concentrations pre- and post-2000. Adopted from Becker et al. (2008).

2.2.4 Influence of climate patterns on the behavior of POPs

How do large-scale climate variation patterns influence POP transport to, and distribution within, the Arctic?

Climate variation patterns, such as the Arctic Oscillation (AO), North Atlantic Oscillation (NAO) and El Niño Southern Oscillation (ENSO), not only affect volatilization of POPs from secondary sources, they can also influence the atmospheric circulation patterns which distribute chemicals around the globe (Figure 2.2.1). Although episodic atmospheric transport events of POPs appear to occur randomly, LRAT follows patterns defined by the physics of atmospheric circulation and physical-chemical properties of the chemicals (Ma et al., 2016). For instance, the trans-Pacific transport of POPs follows west to east storm tracks across the Pacific Ocean (Bailey et al., 2000; Zhang et al., 2008). Storm tracks are climatological winds that occur repeatedly, but their positions and strength may change with climate variabilities. Hung et al. (2005) reported that the gas-phase concentrations of PCB-31, PCB-101, PCB-153 and γ-HCH measured in air at the Canadian Arctic station of Alert (1993-1999) during

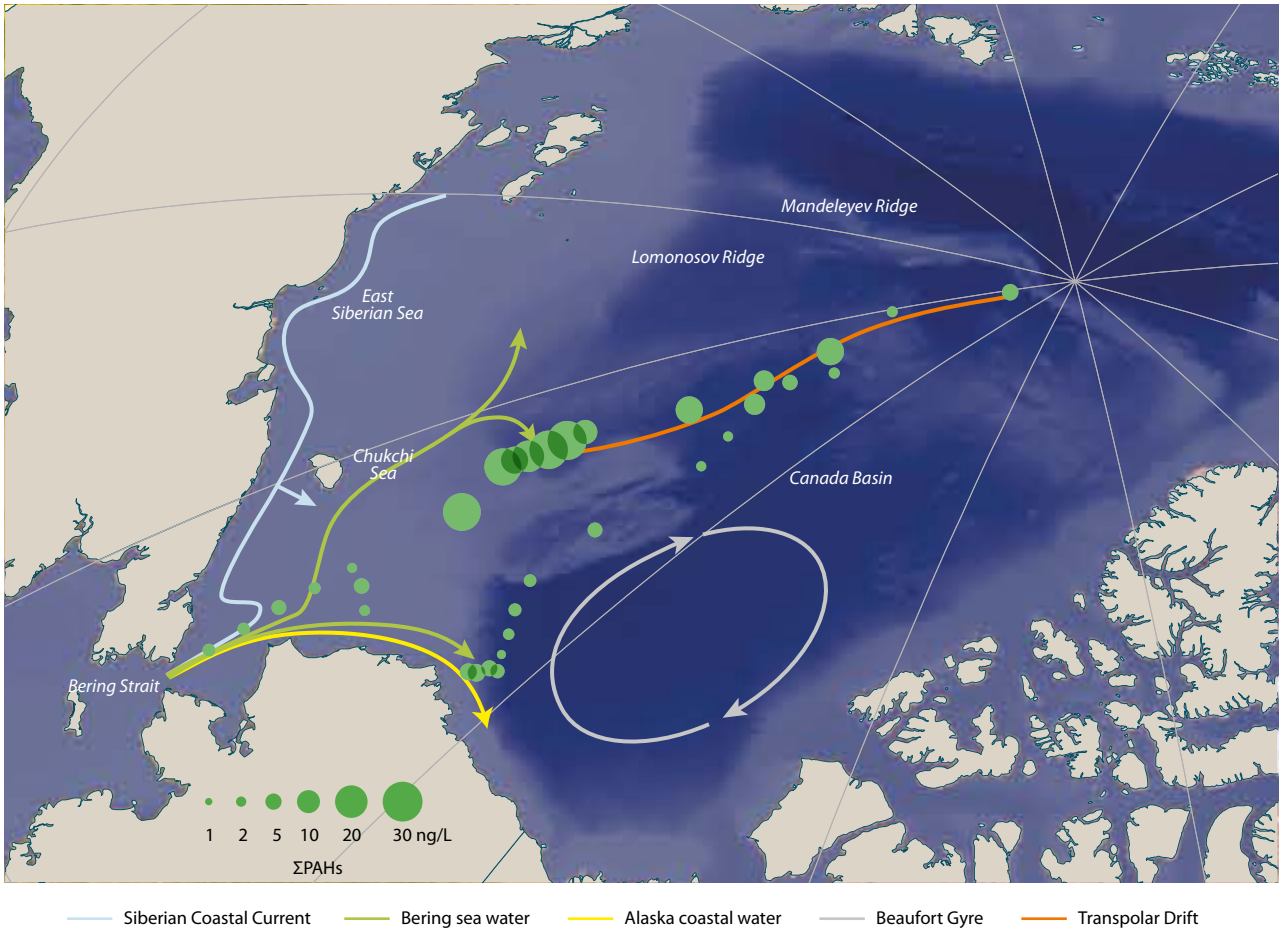


Figure 2.2.7 Concentrations of total dissolved PAHs (ng/L) in surface waters of the Arctic Ocean. Modified from Lin et al. (submitted).

the spring months showed statistically significant ($p<0.1$) correlations with the Pacific North American (PNA) index (Figure 2.2.5). In addition, concentrations of PCB-31, PCB-44 and PCB-138 correlated with the NAO index. Although the underlying mechanisms responsible for these correlations are unknown, such relationships suggest climate variation patterns are capable of influencing the distribution and levels of POPs spatially and temporally within the Arctic. Temperature anomalies associated with climate variation patterns can enhance the volatilization of POPs and affect gas-particle partitioning of atmospheric contaminants, which in turn, affects their transport mechanisms to the Arctic. Climate variation patterns, such as the NAO and AO, can also alter moisture fluxes (Macdonald et al., 2005a), which will affect upstream scavenging of POPs, influencing their transport potential.

Becker et al. (2008) studied twelve years of α-HCH and γ-HCH measurements in air collected from Alert (Canada) and Zeppelin Mountain (Svalbard). It was found that AO fluctuations influenced the α-HCH time-series at Zeppelin, but not at Alert, and not for γ-HCH at either location (Figure 2.2.6, top panel). During the 1990s, the AO was predominantly in a positive phase; after 2000, the AO was mainly in a negative phase. A change in trends after 2000 is apparent for α-HCH measured at Zeppelin, which was supported by a statistically significant relationship ($p<0.05$) between the summer AO index (June-August) and α-HCH concentrations (Figure 2.2.6). The authors have also found that there were much greater variations in concentrations between summer and winter months when AO was in the negative phase (post-2000) (Figure 2.2.6, bottom panel).

Climatic oscillations can also influence the transport of contaminants in Arctic Ocean and coastal seas. During the July to September 2010 R/V Xuelong (Snow Dragon) ship-based expedition, Lin et al. (submitted) found that the PAH concentrations in surface seawater from the Eurasian margin of the Chukchi Plateau were nearly an order of magnitude higher than those in the North American margin (Figure 2.2.7). By examining sea ice back trajectories and stable oxygen isotope data, the authors concluded that the observed concentration difference was driven by the Transpolar Drift and Beaufort Gyre under an enhanced AO, pointing towards significant PAH input from river runoff and ice-melt originating from the Eurasian margin under a warming Arctic. A mass balance model showed that 83% of the PAH input to the Chukchi Plateau was from marginal sources (i.e. river discharge, shelf input, and sediment-laden meltwater), with nearly 100% of PAHs associated with river discharge coming from Eurasia. The model also suggested that 64% of PAHs would be removed by volatilization, indicating the Arctic Ocean has shifted from a receptor to a strong secondary source for PAHs to air. This study demonstrated that a warming Arctic will likely remobilize PAHs from environmental sinks in marginal areas to surface seawater and the atmosphere, thus influencing the global distribution of these compounds.

2.2.5 Changing long-range transport, secondary emissions, human activities, and local sources

How do contaminant pathways into- and within- the Arctic change due to climate change, and how do local sources compare to long-range transport?

Long-range atmospheric transport continues to be a source of POPs and other contaminants (e.g. CEACs) to the Arctic. Oceanic transport is also relevant for more water-soluble contaminants (e.g. perfluoroalkyl acids (PFAAs)) released into coastal seas in temperate regions and then transported northwards. The declining concentrations observed for many POPs in the Arctic atmosphere over the last 15 years or so, is to some extent, being affected by re-mobilization (e.g. revolatilization) of previously deposited stocks of certain chemicals present in Arctic seawater, meltwater, and other compartments. In addition, climate change effects on the Arctic and sub-Arctic, such as an increased frequency and extent of forest fires, changing permafrost, and local pollution sources influencing CEAC levels, in particular, have the potential to influence contaminant levels in the region, and are addressed in the following sections.

2.2.5.1 Changes in long-range transport and secondary emissions – air

Climate change has brought about an ablation of Arctic sea ice, permanent ice, and glaciers. Reductions in the areal extent of sea ice, or earlier seasonal break-up and thaw of ice floes, can affect the water to air transfer of chemicals that have accumulated in surface seawater. For instance, an abrupt increase in air concentrations of α-HCH in the Canadian Arctic coincided with ice breakup in the central Canadian Archipelago (Jantunen et al., 2008). Hung et al. (2016) reported increasing air concentrations of PCB-52 and PCB-101 at the coastal site of Stórhöfði, Iceland in close proximity to retreating sea ice and the deglaciating, retreating ice caps of Mýrdalsjökull and Eyjafjallajökull (Figure 2.2.8, top and second panels). After consistently declining trends from the 1990s to the early 2000s, β-HCH measured in air at Alert, Canada and Stórhöfði, Iceland show complex trends (Figure 2.2.8, third and fourth panels) (Hung et al., 2016). At Alert, β-HCH concentrations increased from 2003 to 2007, and declined thereafter to 2012. In 2007, episodes marked by high concentrations of β-HCH at Alert corresponded with a record low in Arctic sea ice extent. This observation is consistent with the hypotheses of Li et al. (2002) that the dominant transport pathway for β-HCH is via the ocean, and re-emissions from open waters sustain levels in Arctic air (Wöhrnschimmel et al., 2012b). In contrast, at Stórhöfði the seasonal summer maxima was consistently apparent in β-HCH concentration variability (Figure 2.2.8, fourth panel). The different β-HCH concentration patterns observed at Alert and Stórhöfði could be explained by differences in their surrounding environments. In the early 2000s, Alert was mostly ice-bound throughout the year, whereas Stórhöfði is close to open ocean year-round. Therefore, the high concentration episodes observed at Alert were probably due to LRT, while the consistent β-HCH summer peaks at Stórhöfði were probably due to increased volatilization from nearby open waters.

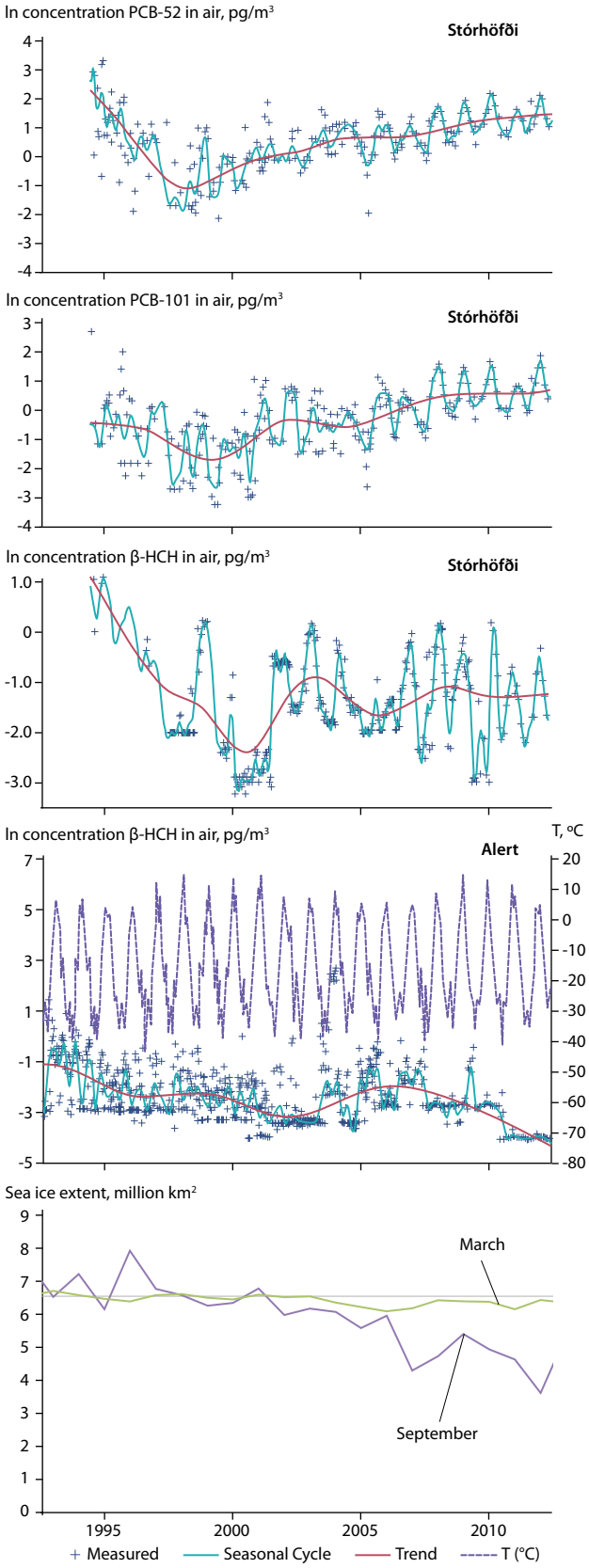


Figure 2.2.8 Temporal trends of contaminant concentrations in air at Alert, Canada and Stórhöfði, Iceland in relation to sea ice extent. Temporal trends of PCB-52 (top panel) and PCB-101 (second panel) concentrations in air at Stórhöfði, Iceland. Temporal trends of β-HCH at Stórhöfði (third panel) and Alert (fourth panel). Air temperatures at Alert are shown in purple dashed line in panel (fourth panel). Average sea ice extent (fifth panel) in March and September (source: National Snow and Ice Data Center). September is usually the month when sea ice reaches its annual minimum extent. For comparison, the 1981 to 2010 average is 6.52 million km². (Adapted from Hung et al., 2016).

Anttila et al. (2016) examined the potential influence of secondary emissions of PAHs and some POPs on future trends by comparing air measurements and time series at the remote Arctic site of Pallas to a site in southern Scandinavia located closer to primary sources between 1994 and 2011. Assuming that future emissions of legacy POPs remain similar to the time periods during which the atmospheric trends were developed (1994-2011), their projections indicated PCBs will be depleted sooner, and chlordanes later, at the Arctic site of Pallas compared to southern Scandinavia, while the most long-lived chemicals will remain in the atmosphere for another couple of decades.

POPs may also be directly released into the sea as a result of melting snow and ice (Geisz et al., 2008). Pućko et al. (2010a, 2011, 2012a,b, 2013, 2015) have shown that sea ice brines can concentrate POPs relative to sea ice and deliver them efficiently to lower trophic levels in the food chain (e.g. phytoplankton, zooplankton). Gioia et al. (2008) showed that air concentrations of PCBs in the Atlantic sector of the Arctic Ocean were higher in the sea ice marginal zone, probably related to their volatilization from melting ice. Recently, at coastal Antarctica, it has been shown that snow melting drives coastal seawater levels of POPs and a net volatilization of PCBs and other POPs, regardless of their physical-chemical properties (Casal et al., 2019).

It is predicted that climate change will increase the frequency and intensity of wildfires which release contaminants, particularly PAHs, but also previously deposited PCBs, into the atmosphere. Yu et al. (2019) found elevated air concentrations of phenanthrene, pyrene, and retene at the Canadian High Arctic station of Alert from 2001-2005 and in 2015 which coincided with more frequent summer forest fire events in Canada, Alaska, and Greenland in these years. In this study, a global 3-D transport model also predicted that warming would result in higher air concentrations of lighter PAHs due to revolatilization from environmental sinks, while particle-bound PAHs (e.g. benzo[*a*]pyrene), were less affected by temperature. In an earlier study, using a combination of statistical fingerprinting techniques and Moderate Resolution Imaging Spectroradiometer (MODIS) satellite images, Sofowote et al. (2011) attributed significant contributions of retene in air at the Canadian sub-Arctic site of Little Fox Lake in autumn 2008 to wildfires occurring in British Columbia, the western United States, and north-eastern Asia during this time. Lin et al. (2020) used an integrated source apportionment technique to reveal five potential sources of PAHs found in 34 surface sediments from the northern Bering-Chukchi margin. The northeast Chukchi Sea exclusively had PAH profiles indicative of contributions from softwood combustion (characterized by retene), a likely result of the increasing wildfires in Alaska related to climate change. Eckhardt et al. (2007) reported extremely high concentrations of PCBs measured at Zeppelin, Svalbard, in spring 2006 and July 2004 as a result of biomass burning in Eastern Europe and boreal forest fires in North America, respectively. Together with warming, an increase in wildfires within and outside the Arctic would release POPs and CEACs (such as PAHs) to the atmosphere and enhance their mobility in the environment.

2.2.5.2 Changes in long-range transport and secondary emissions – ice/snow, surface water and land

POPs that were previously deposited from the atmosphere and subsequently incorporated into ice 'reservoirs' such as snow and glaciers (including ice sheets, ice fields, and ice caps), are being released and remobilized in meltwater due to climate change-induced glacier ablation and melting. These phenomena have been studied outside of the Arctic in alpine systems such as the European Alps (Bizzotto et al., 2009; Bogdal et al., 2009, 2010; Miner et al., 2017) and in Tibetan Plateau glaciers. For example, dichlorodiphenyltrichloroethane (DDT) peaked in the sediment of glacier-fed Lake Oberaar, Switzerland, around 1970 then re-increased in the 1990s, coinciding with the accelerated melting of the adjacent glacier (Bogdal et al., 2010). In lake sediment from the central Tibetan Plateau (Figure 2.2.9), two concentration peaks of DDTs and HCHs were observed; one in the 1970s, corresponding

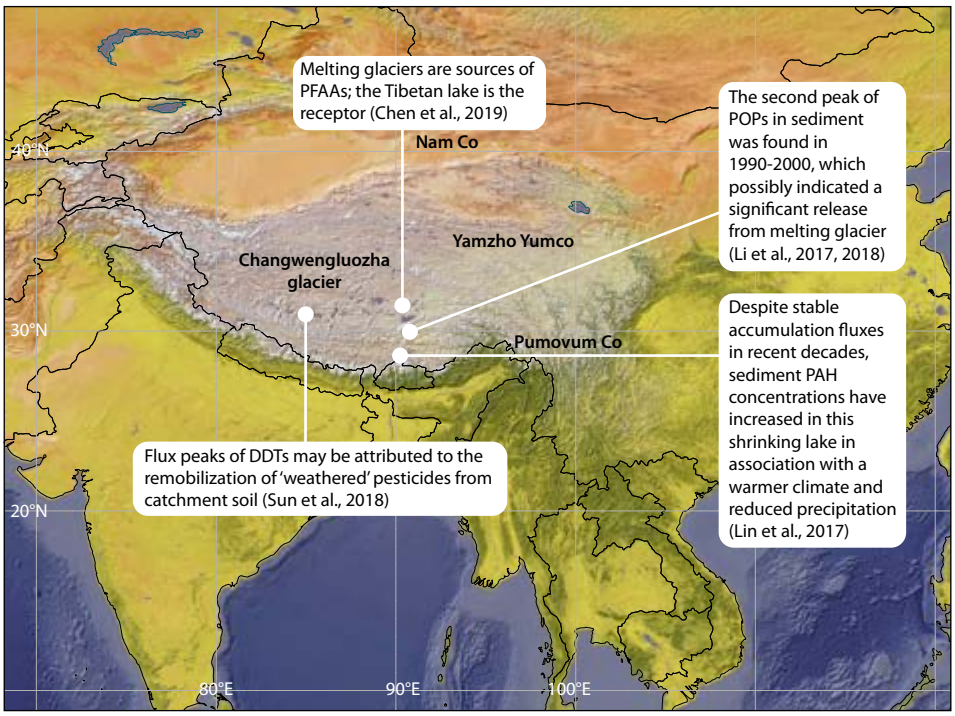


Figure 2.2.9 Climate change-related effects on the distribution of POPs, PAHs and PFAAs observed on the Tibetan Plateau.

to their heavy usage, and the second one between 1990-2000, possibly indicating a significant release from melting glaciers, as the production and usage of DDTs and HCHs has declined significantly since the 1970s (Li et al., 2017c; Sun et al., 2018). This phenomenon is broadly consistent with observations in the European Alps. In addition, flood waters from melting glaciers have influenced POP profiles in receiving lake sediments. Sun et al. (2018) found that there was no difference in the (DDD+DDE)/ΣDDT ratio measured throughout the depth of sediment cores collected from a Tibetian Plateau lake receiving glacial meltwater. In other words, the glacial meltwater retained the DDT signature emitted and deposited in the glacier during the peak usage period in the 1970s. The environment of the Tibetan Plateau, being subject to climatic conditions similar to polar regions, exhibits weak microbiological activity due to low temperatures, thus limiting the transformation of DDT to DDD and DDE. Similarly, a constant β-HCH/ΣHCHs ratio was also observed throughout the lake sediment core depth, however, the ratio was much higher than that of the technical mixture range. The authors attribute this observation to the weathering of HCHs during the process of long-range atmospheric transport, and thus the unchanging HCH signature reaffirms that there is limited degradation of HCHs in the Tibetan Plateau environment after deposition. These studies provide some insights into the influence of melting glaciers on the fate of POPs and confirm that melting glaciers can modify, and even amplify, the organochlorine pesticide (OCP) burden of the contemporary Tibetan environment.

The situation in Tibet is complicated however, because lakes in the Tibetan Plateau are experiencing different environmental changes. In addition to potentially receiving glacial meltwater, some lakes are experiencing greater precipitation, while others are shrinking as a result of the warming climate and reduced precipitation. In the shrinking lake Pumoyum Co located in the southern Tibetan Plateau (Figure 2.2.9), sediment PAH concentrations increased over the past decade, but accumulation fluxes remained stable (Lin et al., 2017), as opposed to the increasing fluxes of OCPs observed in lakes in the northern Tibetan Plateau (Li et al., 2017c; Sun et al., 2018). This observation is attributable to the recent low precipitation rates and lower catchment erosion within the watershed of Pumoyum Co (Lin et al., 2017). This indicates that the impact of climate change and warming is complex, whereby pollutant migration can be influenced significantly by catchment erosion (via sedimentation rates), which is dependent on the meteorological conditions (dry or wet) of the lake's catchment.

Chen et al. (2019) studied the release of water soluble PFAS from a Tibetan glacier. The PFAS composition in Lake Nam Co (Figure 2.2.9) was similar to that observed in glacier ice. Additionally, during the melt season, release fluxes of PFAAs were strongly related to glacial melt intensity (i.e. river flow rate). Moreover, at the end of the melt season, when melt intensity is very low, PFAA release fluxes were found to be positively correlated with PFAA concentrations in glacier ice. This evidence suggests that melting glaciers are sources of PFAAs, while the Tibetan lakes serve as the receptors. Such effects will increase the risks of emerging pollutants to freshwater sources and is of great concern.

Similar to these studies, it may be possible to relate temporal contaminant deposition in Arctic systems receiving enhanced

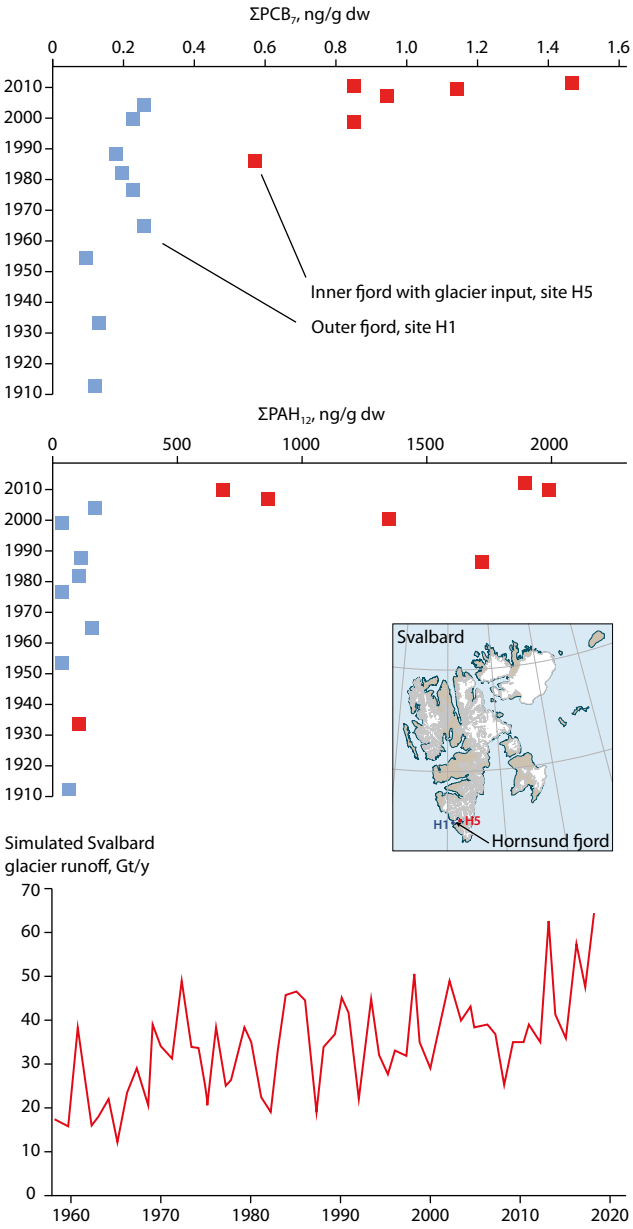


Figure 2.2.10 Sediment contaminant concentrations and glacial run-off on Svalbard. (top and middle) Concentrations of ΣPCB_7 and ΣPAH_{12}, respectively, in dated sediment cores from two sites located in the outer fjord (H1), and inner glacial bay (H5), in Hornsund fjord, southern Svalbard. Data source: Pouch et al. (2017). (bottom panel) Simulated glacier runoff (Gt/y) for all of Svalbard from 1957 to 2018. Data source: van Pelt et al. (2019).

contaminant loads due to climate-induced melting glaciers. Pouch et al. (2017, 2018) reported contaminant deposition in sediment cores collected in 2013 along a transect between the inner and outer areas of two fjords, Hornsund and Kongsfjorden, on the Spitsbergen coast of Svalbard. Approximately 67% and 77% of the drainage basins are covered by glaciers in the Hornsund and Kongsjorden fiords, respectively. Inner parts of the fjords receive enhanced inputs of glacier meltwater (Husum et al., 2019; Błaszczyk et al., 2019) and these inner parts (i.e. glacial bays) are characterized by high sediment accumulation rates (0.23-0.39 cm/year) and low organic matter content (Pouch et al. 2017, 2018). Pouch et al. observed sediment cores sampled from the innermost glacial bays of the fjords contained greater concentrations of ΣPCB_7 and ΣPAH_{12} (Figure 2.2.10, top and middle panels), and hexachlorobenzene (HCB). In the glacial bays of Hornsund

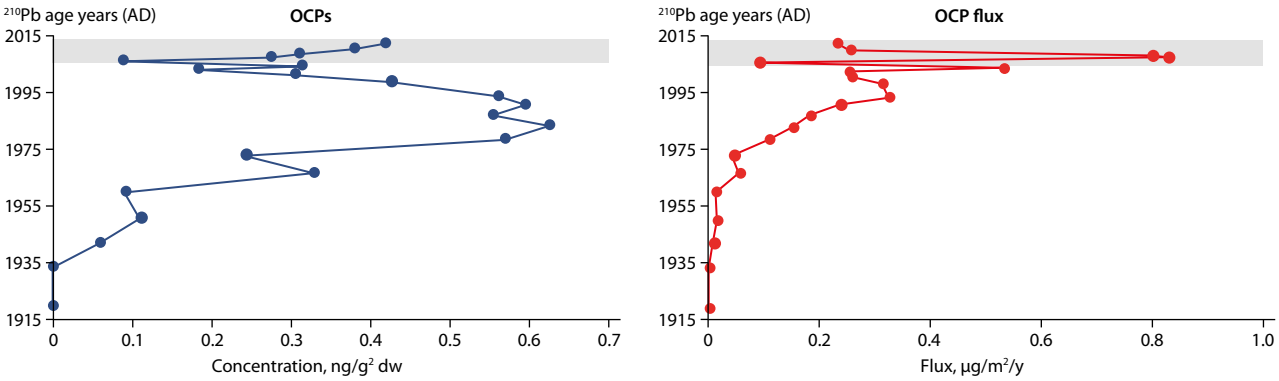


Figure 2.2.11 Concentrations and fluxes of total organochlorine pesticides (OCPs), including DDT, HCH, dieldrin, endrin, and chlordane-related compounds in a dated sediment core from Lake Hazen, Canada. Grey area denotes period of elevated glacial runoff and higher sedimentation rates. Redrawn from Lehnherr et al. (2018).

and Kongsfjorden, ΣPCB_7 were as high as 1.47 and 1.41 ng/g dw, respectively, compared to the outer fjord maximas of 0.25 and 0.62 ng/g dw. Similarly, the Hornsund glacial bay sediment in site H5 contained maximas of 1974 ng/g dw ΣPAH_{12} and 144 pg/g dw HCB compared to 209 ng/g dw ΣPAH_{12} and 53.6 pg/g dw HCB from the corresponding outer fjord area in H1. The congener profile in the inner fjord areas was dominated by light chlorinated PCBs (PCB-28 and PCB-52), which tend to prevail in atmospheric samples, providing further evidence that they originate from direct inputs of glacier meltwater. The exponentially higher rate of deposition of contaminants in the glacier bays of Svalbard post-1990 were attributed to the corollary increase in glacier runoff due to climate change. Van Pelt et al. (2019) applied a multilayer subsurface model combined with a surface energy balance to estimate glacier runoff for all of Svalbard using high resolution climate data (i.e. air temperature, precipitation, relative humidity, cloud cover, and air pressure), glacier coverage, elevation parameters, and snow/firn conditions. The multi-decadal runoff simulation (Figure 2.2.10, bottom panel) indicates a gradual increase in annual run off from 11 gigatonnes per year (Gt/y) to 34 Gt/y, primarily driven by projected temperature increases during the summer melt period.

Fluxes of POPs and CEACs have also been shown to increase in Lake Hazen, the largest lake by volume north of the Arctic Circle, based on a study using dated sediment cores (Lehnherr et al., 2018). Surface temperatures of glacier-covered regions of the Lake Hazen watershed experienced a 2.6°C warming over the period 2000–2012, with the greatest change occurring from 2007 to 2012. The increasing air temperatures and decreasing surface albedo, resulted in major increases in glacial runoff inputs to the lake. Glacier runoff increased lake outflow by 370% for the period 2007–2012 relative to 1996–2006. Sediment accumulation rates post-2007 were on average eight times higher relative to the pre-1948 baseline period. Legacy OCPs were determined in a sediment core from a deep point (267 m) in the lake. OCP fluxes peaked in the 1970-80s, consistent with the known uses of DDT and other OCPs, and showed rising concentrations post–2000 (Figure 2.2.11) reflecting re-mobilization of OCPs previously deposited and stored in glaciers. Previous studies based on a core collected in 1990 at the same location had shown low fluxes of OCPs (Muir et al., 1995).

In the post-2007 glacial-mass loss era in Lake Hazen, in which the runoff rate reached 1-1.8 Gt/y (Figure 2.2.12) and sediment accumulation rates increased by a factor of eight, analyses of

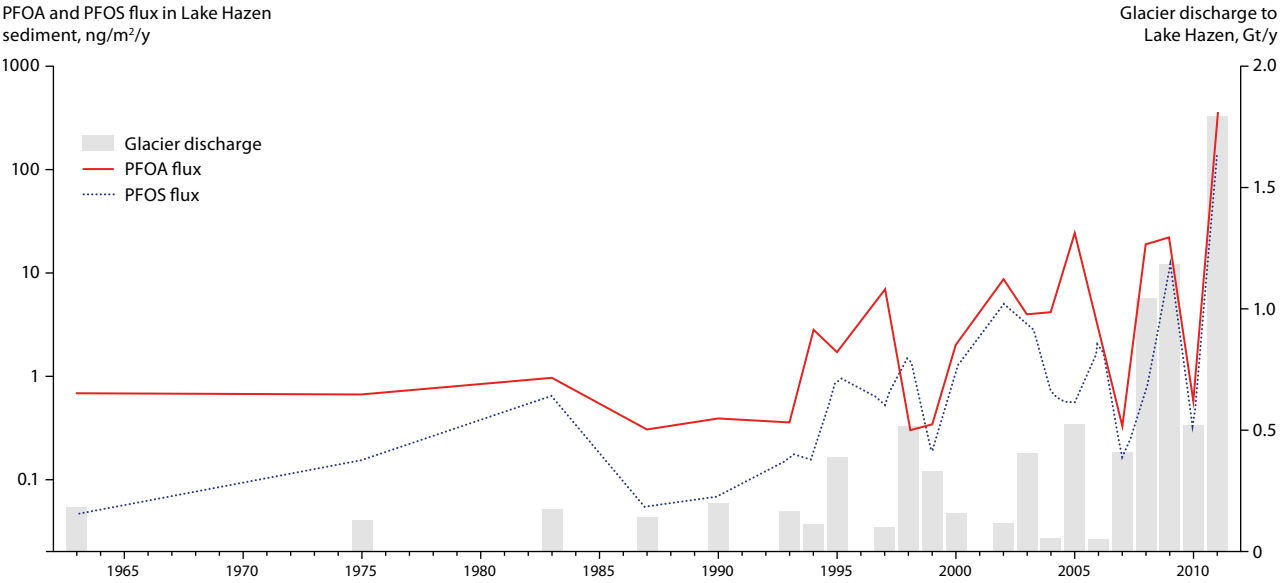


Figure 2.2.12 Deposition of PFOA and PFOS into Lake Hazen sediments in relation to climate-driven variability in glacier discharge rates. Data sources: MacInnis et al. (2019a) and Lehnherr et al. (2018).

dated sediment cores indicated enhanced delivery of PFAS to the lake (MacInnis et al., 2019a). The peak PFAS sedimentary fluxes in Lake Hazen are coincident with the glacier discharge volumes (Figure 2.2.12).

Climate change has been linked to increasing meltwater discharges, shorter periods of ice coverage, increased particle loads, and generally higher flow rates in rivers discharging into the Arctic Ocean (Burkow and Kallenborn, 2000; St. George, 2006). The increased discharge volume and flow velocity from rivers is expected to bring with it an increased contaminant load of both dissolved and particle-bound contaminants (Teran et al., 2012). The increased contaminant load has been hypothesized to include remobilized legacy POPs retained in sediments (Teran et al., 2012), PAHs from sediment-laden ice and coal deposits eroded by deglaciation (Lin et al., 2020), as well as POPs and CEACs with sources close to settlements (Burkow and Kallenborn, 2000; Environment Canada Fisheries and Oceans Canada and Northern Affairs Canada, 2008; Teran et al., 2012; Rodgers et al., 2018).

Taken together, these studies reporting increased contaminant deposition via glacier melt in alpine and Arctic regions provide insights into the influence of melting glaciers on the fate of POPs, and confirm that melting glaciers can amplify the contaminant burden of receiving waters globally. As such, further characterization of contaminants in the cryosphere is warranted to estimate potential releases through meltwaters into Arctic rivers, lakes, and ultimately the ocean. This is particularly germane for those substances known to biomagnify in Arctic marine food webs.

Some CEACs, such as PFAS, especially the anionic PFAAs and halogenated OPEs, have appreciable water solubility, low Henry's Law constants, and slow degradation rates, which presents concerns regarding their susceptibility to long-range oceanic and riverine transport (Reemtsma et al., 2016; Neumann and Schliebner, 2017; Rodgers et al., 2018). In both of these substance groups, concentrations similar to, or exceeding, those of traditional POPs have been reported in Arctic environmental media, including air, snow, water, and biota (AMAP, 2017a; McDonough et al., 2018; MacInnis et al., 2019b; Muir et al., 2019; Sun et al., 2020).

Investigating potential LRT pathways of selected CEACs, Sühring et al. (2016b), McDonough et al. (2018), and Schmidt et al. (2019) found strong indications for oceanic- and riverine-based LRT of chlorinated organophosphate esters (Cl-OPEs) and PFAS. Spatial analysis of Cl-OPEs in the Arctic indicated higher concentrations in near-shore areas, suggesting input from snow melt runoff and riverine sources (Sühring et al., 2016b; McDonough et al., 2018). Moreover, model estimates further supported the hypothesis that oceanic (including riverine) transport of Cl-OPEs could explain some of the high concentrations reported in the Arctic, despite the lack of predicted atmospheric LRT for most OPEs (Sühring et al., 2020). However, sampling of glacier-fed rivers and lake water in the Canadian Lake Hazen watershed has revealed the presence of Cl-OPEs, implying atmospheric transport and subsequent deposition and accumulation in snow and glacier ice (Sun et al., 2020). Cl-OPEs were the most prevalent OPEs in six glacier-fed tributaries sampled in 2015 and 2018. The estimated loading into Lake Hazen by glacier-fed rivers in 2015 was 2.62±1.3 kg Cl-OPEs, and 7.04±3.24 kg total OPEs (ΣOPE_{14} comprised of Cl-OPEs, alkyl-OPEs, and aryl-OPEs). Sun et al. (2020) also reported relatively lower OPE concentrations in non-glacier impacted Arctic lakes. ΣOPE_{14} concentrations in East and West Lakes on Melville Island, Canada corresponded to 4.7–5.3 ng/L, and in Small Lake on Cornwallis Island, Canada, 6.5 ng/L. These levels are approximately half the concentration of Lake Hazen (10-15 ng/L ΣOPE_{14}) despite Lake Hazen being further north and more remote from anthropogenic activity. These results highlight the delivery of OPEs to Arctic lakes via annual atmospheric deposition, with release of accumulated OPEs in glaciers acting as an additional significant source in specific watersheds.

PFAAs can also be deposited from the atmosphere via snowfall. Recently, MacInnis et al. (2019b) investigated the delivery of PFAS from overlying snow and ice into Lake Hazen during the short and pronounced summer-melt period. ΣPFAA were found to be well mixed throughout the 250 m water column during the ice-covered period (September to May, ΣPFAA <0.5 ng/L), elevated in the top 5 m during the melting period (late May, ΣPFAA up to about 3.1 ng/L), and were well-mixed during the ice-free open water period (July-August, ΣPFAA <0.5 ng/L). Many Arctic lakes, like Lake Hazen, are covered by ice and snow for many months of the year resulting in a sudden surface pulse of contaminants during spring melt. Depth-concentration profiling of contaminants in Arctic lake waters during melting events can provide pertinent data for understanding the contaminant-specific flux into underlying waters, as well as transfer further downstream with exiting river or meltwater flow.

2.2.5.3 Indirect effects of a changing Arctic on contaminant sources

It is well known that PCBs from domestic (Sandanger et al., 2003; Jartun et al., 2009; Dudarev et al., 2019a), industrial (Jartun et al., 2009; Garmash et al., 2013) and military sources (Kuzyk et al., 2005; Kalinovich et al., 2008, 2012; Luttmer et al., 2013; Brown et al., 2013, 2015) contribute to local Arctic pollution. PCBs are also leaking from building materials and electrical installations in major settlements in the Arctic, e.g. Longyearbyen and Barentsburg on Svalbard (Pedersen et al., 2011). Many of these sources were registered and, where possible, treated, sealed or removed (Pedersen et al., 2011). Unfortunately, emissions from diffusive sources continue even 40 years after the global ban of PCBs. However, less is known about the emissions of CEACs associated with local domestic and/or industrial sources (AMAP, 2017a; Kallenborn et al., 2018), which are expected to increase as the indirect result of increasing population and human activities due to Arctic warming. For example, many CEACs are released into the environment through sewage. Many sewage systems are inadequate, deteriorating (Daley et al., 2014; Sarkar et al., 2015; Jensen et al., 2018) and difficult to upgrade in the Arctic (Warren et al., 2005; Prowse et al., 2009). Untreated sewage is either directly released into estuarine, coastal, and freshwater systems, or stored for long periods in sewage lagoons. In a warming Arctic, the permafrost underneath such lagoons is melting, and can no longer prevent contaminants from seeping

into the adjacent soil and aquifers (Gunnarsdottir et al., 2013; Jensen et al., 2018).

Sühring et al. (2016b) reported tri-n-butyl phosphate (TnBP), an OPE which is a major component in aircraft fire-resistant hydraulic fluid, Skydrol 500B-4 (Healy et al., 1992), in air samples near a military airport at Resolute Bay in the Canadian Arctic. Consistent with these results, Sun et al. (2020), reported TnBP in two lakes, Resolute Lake and Meretta Lake, at concentrations ranging from 20-40 ng/L. These lakes are in closer proximity to the airport and waste disposal sites in the Resolute Bay community, compared to four other lakes that had TnBP at much lower concentrations, 1-3 ng/L (Sun et al., 2020). High local concentrations of TnBP have previously been linked to emissions from airports (Marklund et al., 2005). Additional OPEs with apparent local sources (i.e. high concentrations and detection frequencies at land-based stations, and low detection rates from ship-based sampling) include 2-ethylhexyl diphenyl phosphate (EHDPP) (Salamova et al., 2014; Sühring et al., 2016b) and tris meta-(cresyl) phosphate (TmCP) (Sühring et al., 2016b). EHDPP contamination has been linked to the use of hydraulic fluids in fishing boat motors from the higher concentrations detected close to the coast or around harbors compared to those measured in open ocean waters (Schmidt et al., 2019). An increase in fishing activities in the Arctic due to reduced sea ice, as well as an increase of industrial activities, military activities, shipping, and tourism (increasing frequency of flights) are likely to result in an increase of these types of contaminants (e.g. TnBP and EHDPP) in local environments.

Furthermore, the retreat of land-fast ice (glaciers) and permafrost is expected to allow the exploitation of previously unavailable resources (minerals, petroleum, gas etc.). For mining and refining, new settlements and infrastructure will be established or expanded, leading to an increased potential for local pollution (Forbes et al., 2004; Prowse et al., 2009; Clark et al., 2016; Crepin et al., 2017). Pollution can also be related to water discharges from offshore installations, as well as from unplanned discharges. Unplanned discharges include leaks of petrochemicals, but also discharge of PFAS-containing fire-fighting foams in the case of incidents, training, or testing of firefighting equipment as reported for Longyearbyen and Ny-Ålesund (Svalbard) (Skaar et al., 2019). Sühring et al. (2017) reported the potential significance of offshore oil and gas installations as sources for marine PFAS contamination based on a model simulation using use and discharge data of offshore fire-fighting foams. Lescord et al. (2015) also noted highly elevated perfluorooctane sulfonic acid (PFOS) concentrations (30-40 ng/L) in water from lakes downstream of the Resolute Bay airport, and also reported levels of other PFAS substances associated with fire-fighting activity, including fluorotelomer sulfonic acid and other perfluoroalkyl sulfonic acids.

The development of economically-feasible technology for fossil fuel production from shale gas (i.e. fracking technologies) and oil sands in the North American Arctic has led to increased land-based exploitation of petroleum resources (Hester and Harrison, 2015; Robertson and Chilingar, 2017; Jacobs and Testa, 2019). Such production technologies are associated with considerable environmental consequences, including large pollutant burdens and destruction of habitats in oil sands regions (Lindstrom and Braddock, 2002; Jautzy et al., 2013; Bauer et al., 2019; Zhao et al., 2019). Recent reports confirm an increase in local contamination of PAHs and POPs in the context of oil sands and shale gas production sites (Noah et al., 2014; Graney et al., 2017; Trellu et al., 2017; Landis et al., 2019). Furthermore, the reclamation of previously degraded sites is currently progressing very slowly in Canada and the US, leaving large areas without the expected conservational or recreational value (RRTAC, 1983; Rooney et al., 2012; Kovalenko et al., 2013; Noah et al., 2014, 2015).

The increased probability of oil spill incidents associated with oil and gas production and refinement are expected to add to the pollutant loads in the Arctic. Experiences from earlier oil spill incidents in the Arctic (both on land and offshore) revealed that such events have severe consequences both for humans and wildlife in the region (Jernelov, 2010; Aune et al., 2018; Carroll et al., 2018; Vergeynst et al., 2019). Furthermore, as a consequence of reduced summer sea ice, new shipping routes along the northeast and the northwestern Arctic coasts are currently being explored as potential international shipping routes (Harrould-Kolieb, 2008) that will serve as viable alternatives for the major Asia-Europe/America shipping routes (Wilson and Rodriguez, 2012; Tedsen et al., 2014; Lasserre and Faury, 2020). Such Arctic shipping routes will shorten transportation times considerably, and thus are economically beneficial (Smith and Stephenson, 2013). However, Arctic transit routes will inevitably lead to local pollution in the region. Four major environment threats need to be considered as a result of increases in Arctic shipping in the regulatory context: accidents resulting in related oil/chemical spills, emissions from combustion engines, release of wastes into Arctic waters (e.g. bilge release and ballast tank maintenance), and emissions of chemicals used as surface coatings on ship hulls (Caddell and Thomas, 2013).

2.2.6 Changing Arctic cryosphere (snow, permafrost, sea ice and glacial melt) – contaminant amplification

How does cryospheric change affect the fate of contaminants in the Arctic?

While climate change is driving a strong declining trend of areal sea ice cover during the Arctic summer, the nature of sea ice is also changing. The Arctic Ocean is now dominated by first-year sea ice (FYI) with a substantial decline in older, multi-year ice (MYI); in other words, there has been a shift from a perennial icescape to an annual one (NSIDC, 2019). Climate warming is resulting in the thawing of permafrost in lake catchments and shorelines with a subsequent alteration of lake and river water chemistry. The thawing of ice-rich permafrost ground is resulting in the formation of thermokarst, the detached layers of land, shallow lakes, and wetlands created from land subsidence. In turn, this can influence watershed hydrology and delivery of dissolved organic carbon (DOC) and particulate organic carbon (POC) to lakes and to coastal seas (Kokelj et al., 2005; Bring et al., 2016). Furthermore, sea-level air temperatures are increasing (Overland and Wang, 2016), and as an apparent consequence, precipitation type (snow vs. rain) and amounts are changing, and possibly increasing across large parts of

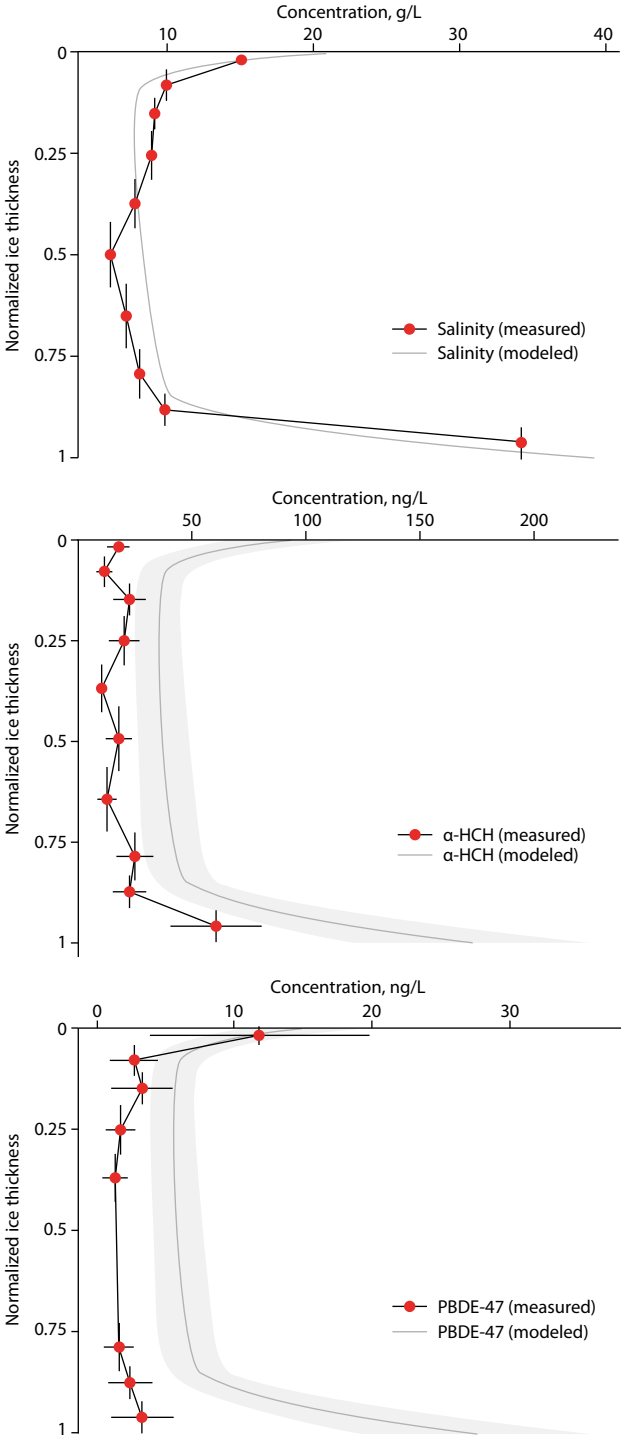


Figure 2.2.13. Modeled and measured concentration profiles for salinity, α-HCH, and PBDE-47 in artificial sea ice grown at -18°C to a depth of 26 cm in a specialized sea ice facility. Vertical bars indicate ice layer thickness. Variability (± 2SD) is indicated for both measured data (horizontal bars) and results of a one-dimensional sea ice brine dynamics model (grey shade). Source: Garnett et al., (2019).

the Arctic (AMAP, 2017b). The influence of these cryosphere changes on contaminant behavior is likely to be pronounced and is explored further in the following sections.

2.2.6.1 Contaminant fate in changing sea ice

MYI used to cover ~60% of the Arctic Ocean, but over the last few decades has declined to <30%, with the oldest ice (4+ years old) comprising only 3-5% of the ice cover in the Arctic (NSIDC, 2019). With regards to the entry and fate of organic contaminants, sea ice has previously been viewed simply as a seasonal barrier limiting the transfer of contaminants between the atmosphere and the surface ocean (Pućko et al., 2015). However, field studies have demonstrated that the sea ice snowpack is a notable repository of atmospherically-derived chemical pollutants, including PFAS (Cai et al., 2012) and OCPs (Pućko et al., 2011). In addition, sea ice itself also contains these chemicals, with α- and γ-HCH present in brine-rich FYI at concentrations much higher than the underlying sea water and other Arctic media (Pućko et al., 2010a). This implies that single-season ice or FYI can accumulate organic contaminants to such an extent that concentrations of these chemicals are amplified relative to the underlying sea water or surface meltwater (Pućko et al., 2015). This process appears to be controlled by brine, the saline water present in young ice. As sea ice forms, most of the salts present in the freezing seawater are rejected into the underlying ocean, leaving only a small amount trapped within a network of highly saline brine pockets. As ice continues to grow, more salts are expelled, and seasonal meltwater at the surface 'flushes' the sea ice, reducing its bulk salinity further. Seasonal or FYI has a higher bulk salinity (Notz and Worster, 2009) and contains more brine per unit volume than MYI, and this brine appears to influence the behavior and fate of organic micropollutants present in the sea ice system (Pućko et al., 2010a; Garnett et al., 2019).

Garnett et al. (2019) conducted experiments in a controlled sea ice facility to understand the uptake and accumulation of a range of POPs in young, growing sea ice. Sea ice formation was shown to result in the entrainment of the chemicals from seawater. The subsequent contaminant concentration profiles in bulk ice showed the highest levels in both the upper (ice-atmosphere interface) and lower (ice-ocean interface) layers, indicating that the incorporation and distribution of POPs is strongly influenced by brine advection within young ice. Figure 2.2.13 shows the similarity in the vertical profiles of contaminants and NaCl in ice, with both displaying a characteristic 'C'-shape driven by brine dynamics in the ice. Application of a one-dimensional sea ice brine dynamics model was able to accurately replicate the organic contaminant profile in the ice. The concentrations of seven POPs (α- HCH, γ-HCH, PCB-28, PCB-52, chlorpyrifos, PBDE-47, PBDE-99) were higher in the ice brine than the bulk ice (i.e. frozen water, trapped air, brine pockets). The concentrations in ice brine were similar to, or higher than, the concentrations in the beneath-ice seawater, indicating an enrichment of semi-volatile contaminants. Enrichment factors (EF), the ratio of the chemical concentration in ice brine to that in the underlying seawater, exceeded one for some of the chemicals in the study (e.g. α-HCH, chlorpyrifos, and PCB congeners).

Table 2.2.3 highlights EFs calculated from the experimental ice chamber work of Garnett et al. (2019), as well as those derived from several Arctic sea ice studies. The highest EFs were observed in the saltiest brine located in the upper ice sections (closest to the atmosphere) similar to what was also observed in FYI in the Amundsen Gulf of the Canadian Arctic. EFs up to four were observed for α- and γ-HCH (Pućko et al., 2010a, 2011), with concentrations that exceeded any other abiotic media in the Arctic. A common feature on the surface of new

Table 2.2.3 Summary of enrichment factors (EF±1 SD) reported for NaCl and chemical contaminants in various sea ice compartments, where EF is the ratio of the chemical concentration in the ice compartment to that in the underlying seawater or uppermost sea ice layer. Modified from Garnett et al. (2019).

Enrichment factor	Bulk ice depth, cm	Chemical								Data Source
		NaCl	α-HCH	γ-HCH	Chlorpyrifos	PCB-28	PCB-52	PBDE-47	PBDE-99	
$EF_{BULK\ ICE\ /\ SEAWATER}$	17±1	0.4±<0.1	0.1±<0.1	0.1±0.1	0.2±0.1	0.2±0.1	0.2±<0.1	0.1±0.1	0.4±0.2	(1)
	26±1	0.3±<0.1	0.2± 0.1	0.3±0.2	0.3±0.2	0.2±0.1	0.1±<0.1	0.2±0.1	0.4±0.2	(1)
	30	0.4	0.4	0.5	nm	nm	nm	nm	nm	(2)
	90	0.2	0.3	0.3	nm	nm	nm	nm	nm	(3)
	5	0.3	0.3	0.4	nm	nm	nm	nm	nm	(3)
$EF_{BRINE\ /\ SEAWATER}$	26±1	1.4±<0.1	0.6± 0.2	1.0±0.8	1.2±0.7	1.3±0.5	1.2±0.3	0.7±0.5	0.9±0.6	(1)
	90	4.4	3.9	4	nm	nm	nm	nm	nm	(2)
$EF_{FROST\ FLOWER\ /\ SEAWATER}$	na	2.4±<0.1	0.2± 0.1	0.2±0.2	0.3±0.2	0.2±0.1	0.4±0.1	6.6±4.4	24±15	(1)
$EF_{FROST\ FLOWER\ /\ SEA\ ICE\ LAYER}$	na	5.0±<0.1	1.5± 0.5	2.0±1.5	2.4±1.4	2.5±0.9	3.0±0.7	30±20	50±31	(1)

EF: enrichment factor, na: not applicable, nm: not measured
Data Sources: (1) Garnett et al., 2019 (2) Pućko et al., 2010a (3) Pućko et al., 2010b

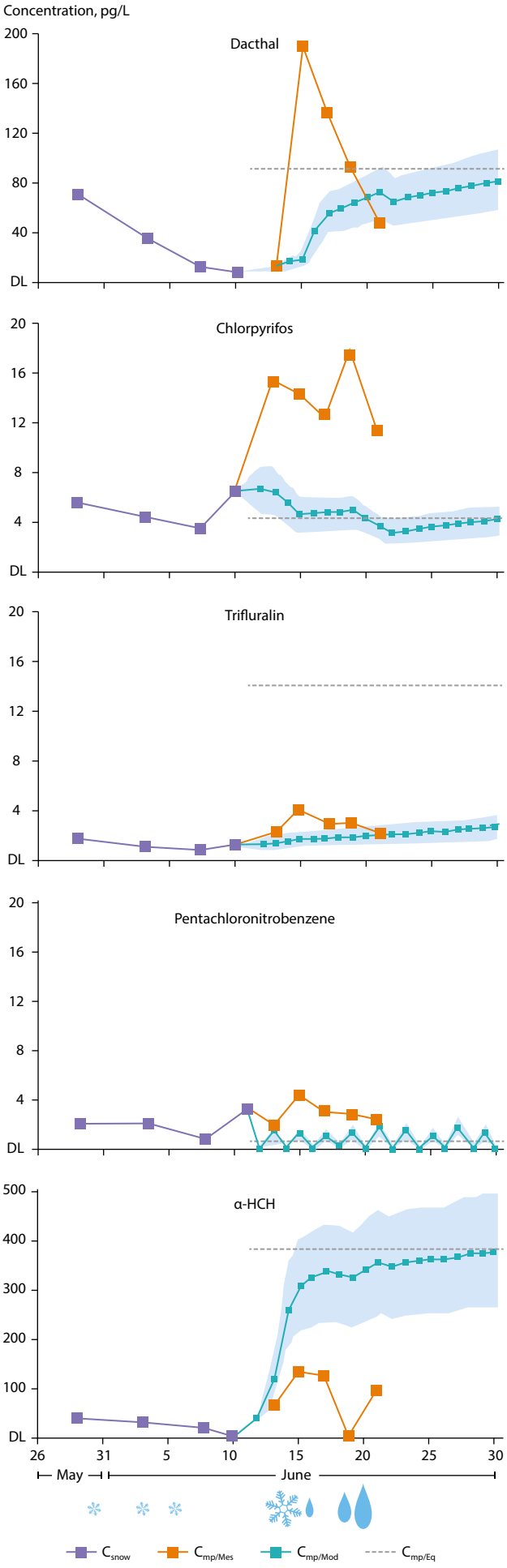
and young ice is frost flowers. These are clusters of dendritic ice crystals that have a very high salt content and form at the interface between a warm ice surface and a cold atmosphere, particularly on new ice which possesses a thin liquid brine layer on the surface (see Barber et al., 2014). Contaminant enrichment in frost flowers is significant, with EFs >10 for PBDE-47 and PBDE-99 observed in the experimental sea ice facility (Garnett et al., 2019). Importantly, this phenomenon was also observed in frost flowers sampled on coastal sea ice near Barrow, Alaska (Douglas et al., 2012), with EFs approaching 40 for an array of organic contaminants. In a warmer Arctic, the frequency of frost flower occurrence is likely to increase over much wider areas of the Arctic Ocean. The implications of this enrichment of contaminants in frost flowers is unclear at present, but is likely to result in enhanced surface to air transfer, and 're-cycling' of contaminants in the Arctic marine environment over a large area.

2.2.6.2 Role of the marine snowpack and seasonal thaw

The transfer of organic contaminants between the ice-rafted snowpack and sea ice is facilitated by the ingress of ice brine into the base of the snowpack during the winter. Observations conducted on FYI in the Amundsen Gulf in the Canadian Arctic estimated that upward percolating sea ice brine provides as much as 50% of the α- and γ-HCH burden present in the overlying snowpack (Pućko et al., 2011). This process is clearly facilitated by the movement of brine (i.e. 'brine engine') present in young and FYI and is greatly diminished or absent in MYI (Pućko et al., 2015). Snow meltwater can percolate into ice during the onset of thaw and was observed to result in an increase of 2 to 32% of the α- and γ-HCH burden in the sea ice.

As the melt season progresses, the thawing snowpack gives rise to melt ponds which are a dominant feature on ice floes during the early summer (Fetterer and Untersteiner, 1998). Organic contaminants present in the melting snowpack may either volatilize back to the atmosphere, depending on their volatility and aqueous solubility, and/or be supplied to melt-ponds. Gas exchange between the atmosphere and the melt-pond surface may serve to increase levels in the pond water, particularly for those contaminants initially found at very low levels in either the snowpack or melt-pond. For example, current use pesticides (CUPs), advected into the Arctic through LRAT during the early summer will undergo air-surface exchange with net loading into melt-pond water. In addition, wet deposition through late season snowfall and rainfall events can significantly enhance contaminant loading to melt ponds (Pućko et al., 2017). Figure 2.2.14 illustrates a time-series of CUP concentrations in the snowpack and melt ponds over the May/June melt season on landfast FYI between Griffith and Cornwallis Island in the Canadian Arctic Archipelago. Of the five CUPs investigated, chlorpyrifos (insecticide) and dacthal (herbicide) showed substantially higher concentrations in the melt-pond water compared to predicted concentrations based solely on gas exchange with the atmosphere. This indicates the important role of the melting snowpack in releasing contaminant residues, while also pointing to the significance of precipitation events which serve to add these chemicals to the melt pond directly. Precipitation in the form of snowfall, and importantly, rain events in late winter, are predicted to increase over large parts of the Arctic Ocean (AMAP, 2011b), enhancing the transfer of atmospheric contaminants to sea ice surfaces and melt ponds. Melt-pond enrichment factors (MEFs), the ratio of contaminant concentrations in the melt pond to those in the underlying seawater, of ~2–10 were calculated for the pesticides endosulfan I, chlorothalonil, and chlorpyrifos. This was based only on net loading through gaseous transfer from the atmosphere using concentrations measured in air during the Arctic summer period. MEFs for dacthal, based on direct measurements in melt-pond water, were found to be much higher, at ~10-16. As a consequence of melt pond enrichment, the exposure of ice algae and phytoplankton to contaminants could significantly increase following late-season melt-pond drainage, either via water percolating through the remaining ice, or drainage directly into beneath-ice seawater.

Contaminant enrichment in sea ice is driven mainly by brine processes in young FYI. As FYI is prevalent over a much wider area of the Arctic Ocean following the decline in the extent of MYI, then seasonal enrichment and subsequent release of



contaminants from melting ice is likely to provide a significant route of contaminant exposure to the lower trophic levels of the marine food web. Importantly, many organisms situated at the base of the pelagic food web are abundant in sea ice and inhabit the network of brine inclusions and channels located towards the base of the ice. The exposure of sympagic biota in particular, such as ice algae and the associated zooplankton present in brine channels, will be significant over a much wider area in a warming Arctic Ocean. There is some uncertainty over the release of organic contaminants during ice melting, with less soluble contaminants retained in the ice matrix until final melt (Garnett et al., 2019). Furthermore, the various stages of melt pond evolution, and subsequent meltwater drainage, could give rise to meltwater percolating into the ice pack appreciably. Alternatively, and in addition to percolation, meltwater may drain abruptly into the underlying seawater depending on ice breakup and the nature of the melt season (e.g. rapid thaw vs. slow thaw) (Pućko et al., 2017). This will have implications for the delivery and exposure of organic contaminants to ice-associated biota. For example, the earlier onset of seasonal thaw, formation of melt ponds, and final drainage following ice floe breakup, could coincide with the widespread under-ice phytoplankton spring bloom (Arrigo et al., 2012). However, this will vary spatially across the Arctic Ocean and coastal seas, as well as temporally, depending on the ice type and the nature of seasonal warming each year.

2.2.6.3 Permafrost degradation and mobilization of POPs

Climate warming is resulting in the thawing of permafrost in lake catchments and shorelines with resulting alterations in lake and river water chemistry. The thawing of ice-rich permafrost ground causes the formation of thermokarst, detached layers of land, shallow lakes, and wetlands formed from land subsidence, that can influence watershed hydrology and delivery of DOC and POC to lakes and the ocean (Bring et al., 2016; Kokelj et al., 2005).

Permafrost thawing and thermokarst formation have implications for the partitioning and transport of POPs that may be deposited to terrestrial environments via atmospheric deposition and then mobilized with DOC/POC runoff. Eickmeyer et al. (2016) found that slump-affected lakes contained higher total organic carbon (TOC)-normalized concentrations of ΣPCB, HCB and ΣDDT than nearby reference lakes that were unaffected by thaw slumps (Figure 2.2.15). ΣPCB, HCB and ΣDDT concentrations were positively correlated with mean total sedimentation rate for each lake (Figure 2.2.15). The higher TOC-normalized concentrations in slump-affected lakes were explained by the reduced availability of organic matter for adsorption in the water column, so that the POPs were associated with a smaller pool of organic carbon. Slump-affected lakes are generally observed with lower DOC

Figure 2.2.14 Concentrations of dacthal, chlorpyrifos, trifluralin, pentachloronitrobenzene, and α-HCH in snow (C_{snow}) and in melt ponds as measured ($C_{mp/Mes}$), modeled ($C_{mp/Mod}$) and calculated at equilibrium partitioning with atmosphere ($C_{mp/Eq}$), along with the relative types and amounts of precipitation (denoted as size and type of symbols underneath graphs) as a function of season progression. Light blue shadows show uncertainty range for $C_{mp/Mod}$; concentration scales not uniform between graphs. Measurements were conducted on melting landfast ice in the Canadian Archipelago. Source: Pućko et al., 2017.

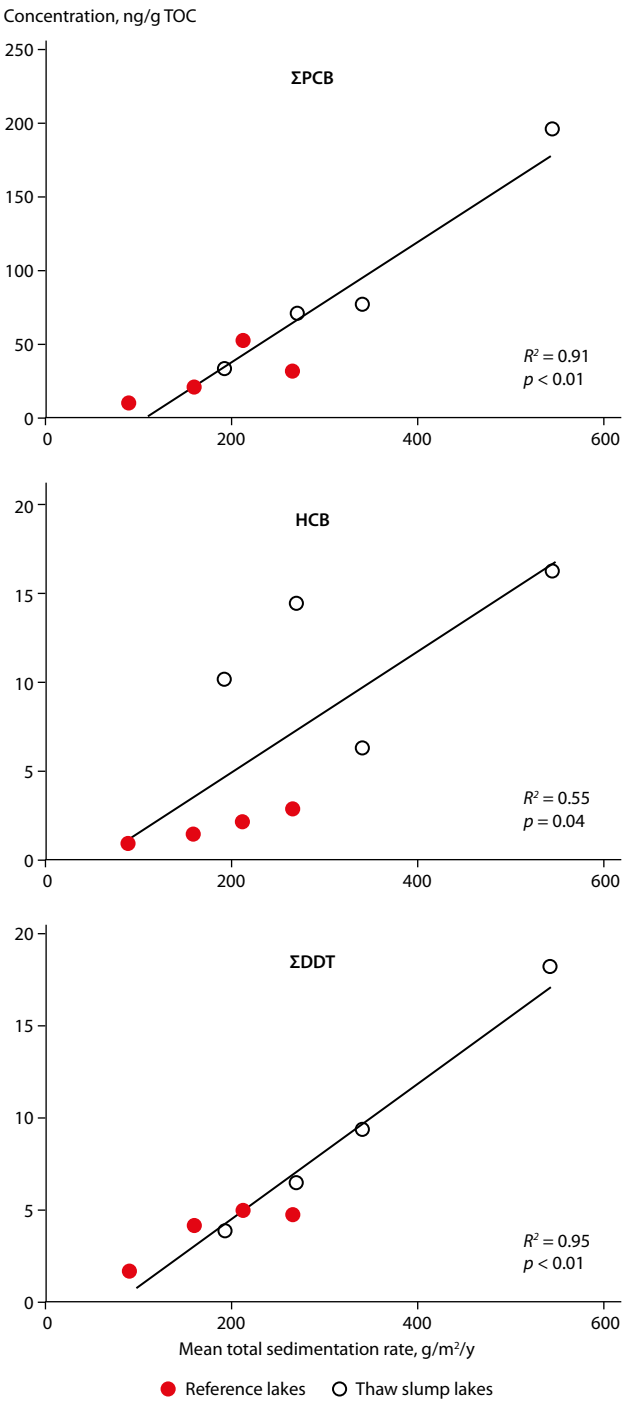


Figure 2.2.15 Concentrations (ng/g TOC) of ΣPCBs, HCB and ΣDDT in sediments from eight study lakes, including reference lakes (red points) and lakes affected by thaw slumps (white points) in tundra uplands adjacent to the Mackenzie Delta, Canada, plotted versus the mean total sedimentation rate (g/m²/y) in each lake. Redrawn from Eickmeyer et al. (2016).

concentrations than unaffected lakes, which is thought to be related to the low DOC in runoff from recently disturbed areas of exposed mineral soils (Kokelj et al., 2005; Thompson et al., 2012).

Fluxes of POPs (concentrations × sedimentation rates) were generally higher and more variable in slump-affected lakes than the reference lakes in the tundra uplands adjacent to the Mackenzie Delta (Eickmeyer et al., 2016). Although temporal resolution was limited due to low sedimentation rates, the historical profiles generally had subsurface maxima in both reference and impacted lakes. Subsurface maxima have generally been observed in arctic lake sediment cores

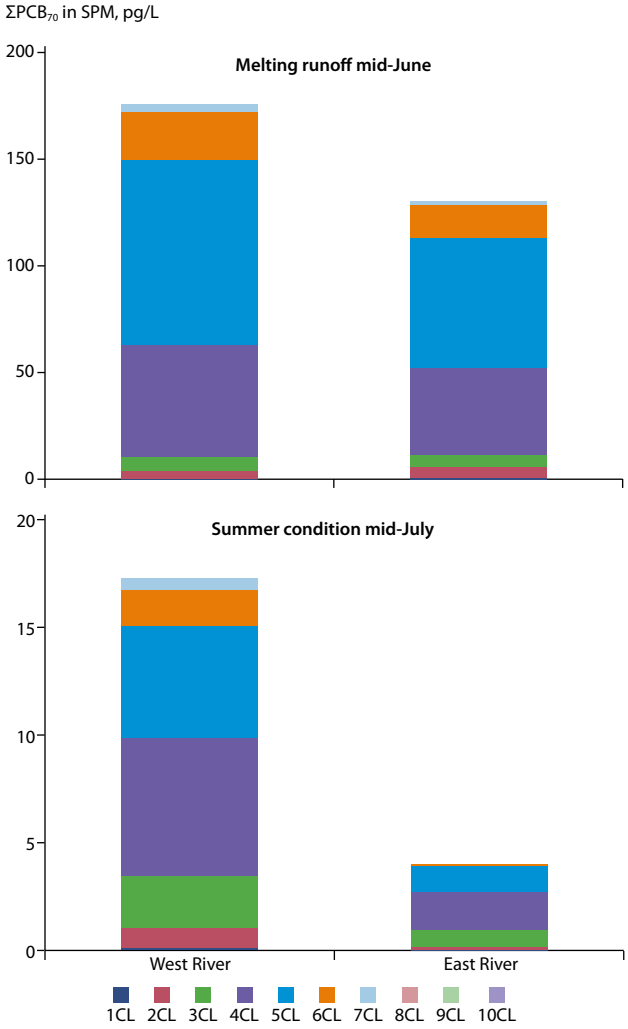


Figure 2.2.16 Concentrations of ΣPCBs$_{70}$ and homolog groups based on degree of chlorination (CL) in suspended particulate matter (SPM) during melt runoff (mid-June) and ice-free (mid-July) conditions in West and East Rivers on Melville Island, Canada in 2016. Redrawn from Cabrerizo et al. (2019a).

because of the greater emissions of POPs in previous decades (Muir et al., 1996; Stern et al., 2005). Thus, it appears that current permafrost degradation and slumping at these sites increased inputs of POPs but not sufficiently to cause a major change in inputs relative to past deposition in the 1960-80s. Fluxes of POPs have also been shown to increase in Lake Hazen in the Canadian Arctic based on studies using dated sediment cores (see Section 2.2.5.2).

Cabrerizo et al. (2019a) compared concentrations of PCBs and OCPs in river water from the West Lake catchment with the nearby East Lake river on Melville Island in the Canadian Arctic archipelago. The West Lake catchment experienced impacts from permafrost degradation associated with significant warming during the period 2007 to 2012, while the catchment of East Lake has undergone little change (Lamoureux and Lafrenière, 2014). PCB concentrations and homolog profiles were similar in both rivers during the snowmelt period in mid-June (Figure 2.2.16), suggesting a similar source such as snow melt runoff. However, in the brief snow-free summer period during July, concentrations and PCB homolog profiles differed between the two rivers. Much higher concentrations of dissolved and particulate PCBs and OCPs were observed in the West River, and lower proportions of di-, octa- and nona-chlorinated

PCB congeners were seen in the East River, suggesting differing sources. It should be noted that atmospheric deposition is the only source of POPs to these very remote catchments.

Higher concentrations of PCBs on suspended particulate matter (SPM) were also detected in West Lake (ΣPCB_{70} = 75 pg/L) in comparison to East Lake (ΣPCB_{70} =1 pg/L). West Lake has been impacted by several subaqueous slumps since 2007 which may be related to permafrost degradation, although this is unconfirmed (Roberts et al., 2017). West Lake is very turbid, with 50- to 100-fold greater turbidity compared to East Lake. Thus, the source of higher PCBs in West Lake is likely a combination of greater catchment inputs and higher SPM content. West Lake's elevated levels of SPM-associated PCBs are also reflected in the higher PCB levels detected in its biota, including zooplankton, fish stomach contents, and landlocked char (Cabrerizo et al., 2018a; Cabrerizo et al., 2019a). Whether elevated SPM-associated POPs in lake water is also observed in other turbid lakes or thermokarst-impacted lakes is not known at present. Using satellite imagery, one study found that 288 lakes on Banks Island in the western Canadian Arctic archipelago had been impacted by retrogressive thaw slumps, with the majority of the changes occurring between 1999 and 2013, a period of significant warming (Lewkowicz and Way, 2019).

Figure 2.2.17 Four glacial sites on Svalbard where ice core and/or surface snow have been analyzed for POP deposition.

2.2.6.4 Deposition of POPs to Svalbard glaciers and ice caps in a changing climate

The deposition of various contaminants, particularly POPs, has been identified in ice cores, surface snow, or both from four glaciers located on Svalbard (Hermanson et al., 2005; Ruggirello et al., 2010; Hermanson et al., 2010; Garmash et al., 2013; Kwok et al., 2013; Hermanson et al., 2020). The four glacier sites are shown in Figure 2.2.17. One goal of these studies was to identify potential POP source regions, which are generally understood to be the northern areas of Europe and Russia. Being that the glaciers and ice caps sampled in these studies are >500 meters above sea level (MASL), they are likely receptors of pollution from LRAT, although it is recognized that the coastal communities in the southern part of Spitzbergen, the main island of Svalbard, are also a potential source of POPs (Lundkvist et al., 2008).

Contaminants, including PCBs, brominated flame retardants (BFRs), and pesticides (both legacy and current-use) are well characterized in these Svalbard glaciers and/or the surface snow at the sites. Historical data from ice cores indicates that in some cases, the discontinued use of pesticides has resulted in lower inputs to the glaciers. However, for PCBs, inputs appear to vary across the years, indicating significant recycling in the global environment despite the cessation of new production. Cross-Svalbard results are available for some pesticides (Ruggirello et al., 2010) and show that the eastern site at Austfonna receives inputs of a larger number of pesticides and greater amounts of some, than Holtedahlfonna, suggesting that the dominance of southeast winds on Svalbard favors deposition to Austfonna. This was also observed in elemental carbon deposition work by Forsström et al. (2009). More recently, a cross-Svalbard investigation of PCB deposition to surface snow at the four sites in Figure 2.2.17 shows greater inputs to the western Kongsvegen site than the eastern sites at Lomonosovfonna and Austfonna (Hermanson et al., 2020). It was concluded that the Kongsvegen site was affected by local sources of PCB from the western communities on Svalbard, in addition to an apparent overlay of a commercial PCB mixture from Europe observed at all sites. The differences in the elevations of the four sites are key to this. Kongsvegen (700 MASL) and Austfonna (740 MASL) are within the tropospheric boundary layer (TBL) during the winter, therefore, both may be affected by local sources, although Austfonna is more remotely located from the western settlements. Holtedahlfonna (1150 MASL) and Lomonosovfonna (1202 MASL) are above the TBL all year (Esau et al., 2012) and as a result, local emissions are unlikely to accumulate there.

Air temperatures on Svalbard are increasing, and as an apparent consequence, precipitation amounts are also increasing. The influence of these changes on POP deposition could be significant because higher air temperatures will result in higher vapor pressures that retain highly volatile contaminants in the gas phase, but also shift traditionally less-volatile contaminants towards the gas phase.

Characterization of the effects of climate change on transport and accumulation of POPs in the Svalbard cryosphere and other glaciated areas in the Arctic requires four types of reliable information: (1) LRAT sources; (2) the climate change forcing or warming at those sources; (3) present and historical deposition of contaminants to ice caps and glaciers; (4) the current climate-change influences on those ice caps and glaciers. Of these four, the only reliable existing data for Svalbard glaciers are for contaminant deposition (3). Investigators have hypotheses about source regions (1), as noted earlier as being in northern Europe and Russia (i.e. northern Eurasia), however the exact source locations within these areas are not known. Additionally,

there are some data suggesting positive temperature anomalies in these regions that may change the emissions of POPs (2). The most significant missing information regards the current climate change impact on Svalbard glaciers (4).

The effect of temperature increases on the glaciers themselves is particularly important because on glaciers, contaminants interact only with air and the snow surface, as there is no canopy, soil or meltwater (Daly and Wania, 2004). The temperature at glacier surfaces is therefore critical. It is known that air temperatures will affect surface snow temperatures, however these can vary widely from year to year independent of the effects of climate change. For example, Erath (2005) found that temperatures in the upper 0.5 m of seasonal snow at Kongsvegen, Svalbard varied from -18°C in April 2000 to -5°C in April 2001. Although both temperatures are well below freezing, the striking difference shows that the unusually cold, late winter in 2000 influenced these snow temperatures.

Current conditions of air temperatures and precipitation amounts on Svalbard glaciers and ice caps are less well understood than those at sea level because of the absence of long-term data characteristic of glaciers throughout the Arctic (Claremar et al., 2012). Due to this data gap, the effects of climate change on glaciers are typically described on large spatial scales, while the contaminant results are reported on smaller spatial scales, such as single sampling sites. For example, Claremar et al. (2012) mention that the glacial mass on all of Svalbard has been falling for a century as a result of warming following the end of the Little Ice Age in about 1870.

Glacial recession is another large-scale measurement, and Erath (2005) shows that the Kongsvegen glacier has receded several km since 1960. Aas et al. (2015) mention using the placement of stakes to measure net snow accumulation during the previous winter on several Svalbard glaciers. However, these stake measurements are read only once each year, which loses finer-scale seasonal information. While each of these approaches is an indication of the effects of temperature and precipitation changes on glaciers over time, they do not describe the more localized or short-term effects, which may vary among sites because of differences in elevation. A potential solution to account for elevation differences is to use existing long-term data from one elevation to estimate the temperature at another elevation using the lapse rate (i.e. the rate at which temperature falls with altitude). However, Erath (2005) notes that the lapse rates at Kongsvegen differ from typical averages and are different between seasons. Automatic weather stations (AWSs) offer another solution. AWSs can operate unattended for extended periods of time, but with varying levels of success (Erath, 2005). On Svalbard, there are numerous AWSs, but only two AWSs have been installed and working on glaciers since the 2000s. Continuous data from AWSs would better clarify warming since the 1990s caused by increasing greenhouse gas forcing (Claremar et al., 2012).

The two operating AWS systems on Svalbard are at Kongsvegen site 6 (used by Erath, 2005) at 537 MASL, and the other at Austfonna (370 MASL) (Aas et al., 2015). Both of these AWSs measure air temperature and precipitation, two vital variables for understanding how the climate at these sites may be changing. However, both of these are well below the peak altitudes on both glaciers where samples for POPs measurement have been collected (700 MASL at Kongsvegen and 740 MASL at Austfonna), and there are no AWSs at Holtedahlfonna and Lomonosovfonna. Both existing AWSs have provided data to verify results of climate models (Aas et al., 2015), in this case the Weather Research & Forecasting (WRF) model combined with the Glacier Climatic Mass Balance (CMB) model. The results of a 10-year study by Aas et al. (2015) show that the model overestimated the measured temperature at Kongsvegen by 0.2°C and underestimated the temperature at Austfonna by 1.9°C, but that the variances between measured and modeled temperatures were consistent. Erath (2005) showed that the AWS at Kongsvegen provided some useful snow accumulation data, but AWS maintenance problems caused a loss of data from time to time. In the WRF-CMB model results, snow accumulation data tended to be variable, and it was concluded that the model was insufficient to resolve a number of small-scale processes (i.e. those on the order of less than one km because of the topography on Svalbard), which could include processes that might affect contaminant deposition.

While these model results are encouraging, the indication is that more AWSs would help understand the conditions at higher elevation glaciers where contaminant samples are collected. Without more AWS data, the understanding of how glacier climatic processes are changing will be very uncertain. Continued investigation of POPs deposition may need to rely on other information or estimates to describe climate-related changes over time.

The deposition of contaminants, such as BFRs, CUPs, and PFAS, have been previously reported in the Devon Island ice cap, Canada, located at 2000 MASL (Meyer et al., 2012; Zhang et al., 2013; MacInnis et al., 2017; Pickard et al., 2018). In these studies, temporal trends in depositional fluxes were related to changes in emission inventories. Climate change was not explored as a significant influence in contaminant deposition in these studies. However, the utility of ice cores has long been demonstrated as a proxy for paleoclimate records, with much of that research focusing on climate reconstruction dating thousands of years ago. Thus, there is untapped potential for using ice core analyses to relate contaminant trends to more recent climate data in order to understand the influence of climate change on contaminant dynamics. New tracers for environmental processes are emerging that could be used to examine sources and mechanisms of contaminant transport (i.e. advective vs. diffusive transport) to the Arctic. For example, a recent study presented analysis of four aromatic acids in ice cores that have potential to serve as tracers for biomass burning events and biogenic aerosols (Wan et al., 2019) and another report developed a method for using short chain fatty acids in ice cores as a proxy for sea surface microlayer aerosol transport (Pokhrel et al., 2015).

2.2.7 Halogenated natural products (HNPs) and the influence of climate change

Halogenated natural products (HNPs) are organic compounds containing bromine, chlorine, iodine, and sometimes fluorine, that are mostly biosynthesized by a variety of marine and terrestrial organisms. HNPs are considered CEACs as many

Box 2.2.1 Halogenated natural products (HNPs)

Halogenated natural products (HNPs) are CEACs (AMAP, 2017a; Bidleman et al., 2017a) that are biosynthesized in marine (mainly) and terrestrial environments. HNPs are produced through biosynthesis by marine bacteria, phytoplankton, macroalgae and some invertebrate animals. Hydrogen peroxide, released during photosynthesis and photorespiration, oxidizes seawater halides under catalysis by vanadium peroxidase. Oxidized halogen species then react with organic substrates within the species or with dissolved organic matter (Lim et al., 2017; Wever et al., 2018; Bidleman et al., 2017, 2019a, 2020; Keng et al., 2020). Two subclasses of HNPs exist:

- *Volatile low molecular weight natural halocarbons (nHCs)* are mainly methanes and ethanes containing chlorine, bromine and/or iodine; mixed substitutions are common. They play a key role in regulating tropospheric and stratospheric ozone. A large database exists for nHCs in the atmosphere and ocean surface water (Ziska et al., 2013).
- *Higher molecular weight halogenated natural products (hHNPs)* have similar physicochemical properties (e.g. partitioning coefficients such as K_{OW}, K_{AW}, K_{OA}) as many POPs and CEACs (Bidleman et al., 2017, 2019a), therefore food web bioaccumulation and toxic properties should be similar, recognizing potential differences in metabolism (Vetter and Gribble, 2007). Far less information exists for hHNPs in abiotic media, though the database is larger for fish, birds, and marine mammals (Alonso et al., 2014; Bidleman et al., 2019a). Bromoanisoles (BAs), which originate from bromophenols (BPs), are the most frequently reported hHNPs in air and water, however, there are a number of other hHNPs measured in the environment (Table 2.2.4). See Bidleman et al. (2017a, 2019a, 2019b, 2020) for more information.

Table 2.2.4 Groups of higher molecular weight halogenated natural products (hHNPs).

hHNP group	Abbreviation
bromoanisoles	BAs
bromophenols	BPs
hydroxylated polybrominated diphenyl ethers	OH-PBDEs
methoxylated polybrominated diphenyl ethers	MeO-PBDEs
mixed halogen compound (sesquiterpene)	MHC-1
polybrominated dibenzo-p-dioxins	PBDDs
polybrominated hexahydroxanthene derivatives	PBHDs
polyhalogenated carbazoles	PHCs
polyhalogenated 1,1'-dimethyl-2,2'-dimethylbipyrroles	PDBPs
polyhalogenated 1'-methyl-1,2'-bipyrroles	PMBPs
polyhalogenated N-methyl indoles	PMIs
polyhalogenated N-methylpyrroles	PMPs

have persistent, bioaccumulative, and toxic properties similar to those of anthropogenic contaminants, and in some cases, have been found at levels in the environment that equal or exceed those of POPs (AMAP, 2017a). This section distinguishes between two groups of HNPs: light halomethanes and haloethanes, referred to as natural halocarbons (nHCs), and higher molecular weight HNPs (hHNPs). See Box 2.2.1 for more information on these two subclasses.

2.2.7.1 Natural halocarbons (nHCs)

Halocarbons, both natural and anthropogenic, have received much attention as regulators of atmospheric ozone. The nHCs research community is thus well advanced in making measurements, modeling transport pathways, evaluating impacts on tropospheric and stratospheric ozone, and forecasting trends in a changing climate (Bidleman et al., 2017a, 2020). Climatic change is expected to affect nHCs in the ocean through shifts in species distributions, changes in ocean chemistry (e.g. pH, salinity, nutrient availability) and changes in physical factors (e.g. temperature, UV radiation, oceanic/atmospheric circulation, glacial melt, precipitation, coastal runoff). nHC emissions and injection into the stratosphere are sensitive to these factors as well as others associated with sea-air exchange (i.e. air and water concentrations, temperature, winds, ice cover) and atmospheric processes (i.e. vertical convection, atmospheric chemistry) (Bidleman et al., 2020). Climate-chemistry modeling is ongoing to project future nHC emissions, transport, and impact on the stratospheric ozone, with good comparison among models thus far (Hossaini et al., 2016; Bidleman et al., 2020).

Approximately half of the total bromine reaching the stratosphere in 2016 consisted of natural tribromomethane ($CHBr_3$) and other brominated nHCs, however, there is no indication of long-term trends so far (WMO, 2018). Most models predict an increase in nHC emissions over this century, but the impacts of this increase are uncertain. Increased nHC emissions may lead to a reduction in tropospheric ozone; this effect would then be carried over to the stratosphere due to an increase in convective transport, particularly in the tropics (Ziska et al., 2017). Stratospheric ozone loss from short-lived nHCs has nearly twice the radiative effect of long-lived (mostly anthropogenic) halocarbons, and therefore an increase in nHCs could have important impacts on future climate (Hossaini et al., 2015). On the other hand, offsetting factors such as increased tropospheric height, and greater chemical degradation and removal of particulate bromine, could mitigate the effects of higher ocean flux; in this case, the bromine from nHCs would not be a major source of stratospheric ozone depletion and climate forcing in the future (Falk et al., 2017).

Halocarbons are produced by an array of organisms including marine bacteria, phytoplankton, macroalgae and some invertebrate animals. In the Arctic, ice algae and microorganisms present in sea ice and frost flowers can release nHCs to the

atmosphere (Sturges, 1992; Granfors et al., 2013a, 2013b, 2014). In addition, climate change may impact areas where macroalgae are, or could be, commercially farmed; these farming operations and drying of the harvested crop may increase nHC emissions (Keng et al., 2020). Most commercial macroalgae farming occurs in tropical-subtropical regions (FAO, 2018), with recent expansion to the Nordic region (Hasselström et al., 2020).

2.2.7.2. Higher molecular weight halogenated natural products (hHNPs)

Similar to nHCs, hHNPs are both naturally- and anthropogenically-produced, but are of potential concern because, in many cases, their physical-chemical properties resemble those of POPs and other chemicals of concern. hHNPs are comprised of a highly diverse group of compounds; refer to Box 2.2.1 for a detailed list.

hHNPs have been reported in air, water and sediments from locations around the globe, including the Arctic (Figure 2.2.18). Bromoanisoles (BAs) are the most frequently reported hHNPs in air and water. Figure 2.2.19 summarizes the spatial distributions of two BAs, 2,4-dibromoanisole (2,4-diBA) and 2,4,6-tribromoanisole (2,4,6-triBA) in air and seawater, and demonstrates their presence at Arctic latitudes. BAs originate from bacterial O-methylation of bromophenols (BPs) that are produced by phytoplankton and macroalgae but are also released from anthropogenic activities including wastewater and seawater chlorination, and various industrial processes. BPs are ionized at seawater pH, but neutral BAs are volatile and found in air worldwide.

The few co-located measurements that exist for BAs in air and water indicate net sea-to-air exchange that occasionally approaches near equilibrium (Pfeifer et al., 2002; Wong et al., 2011; Bidleman et al., 2016). Nearly all the data in Figure 2.2.19 stem from single measurements or short-term campaigns. The only current air monitoring of 2,4,6-triBA is being done at stations in temperate and Arctic Norway, where annual means showed no trends between 2007 and 2018 (Bohlin-Nizzetto et al., 2019). A set of air measurements at the Arctic station Pallas, Finland showed a significant increase in 2,4-diBA between 2006 and 2012 ($p<0.05$); an increase in 2,4,6-triBA was also suggested, but was not significant ($p=0.064$) (Bidleman et al., 2017b).

Since hHNPs are produced in the ocean, transport via ocean currents seems likely (e.g. movement from productive coastal/shelf areas to the open ocean and tropical/temperate to polar regions). hHNPs are pseudo-persistent; always there, albeit with seasonal cycles. Continuous production implies continuous destruction, therefore, there must be efficient pathways for the breakdown of these compounds in the environment and metabolism by organisms (Vetter and Gribble, 2007). However, information on removal rates and processes within the Arctic are currently unknown, and ocean transport of hHNPs has scarcely been investigated.

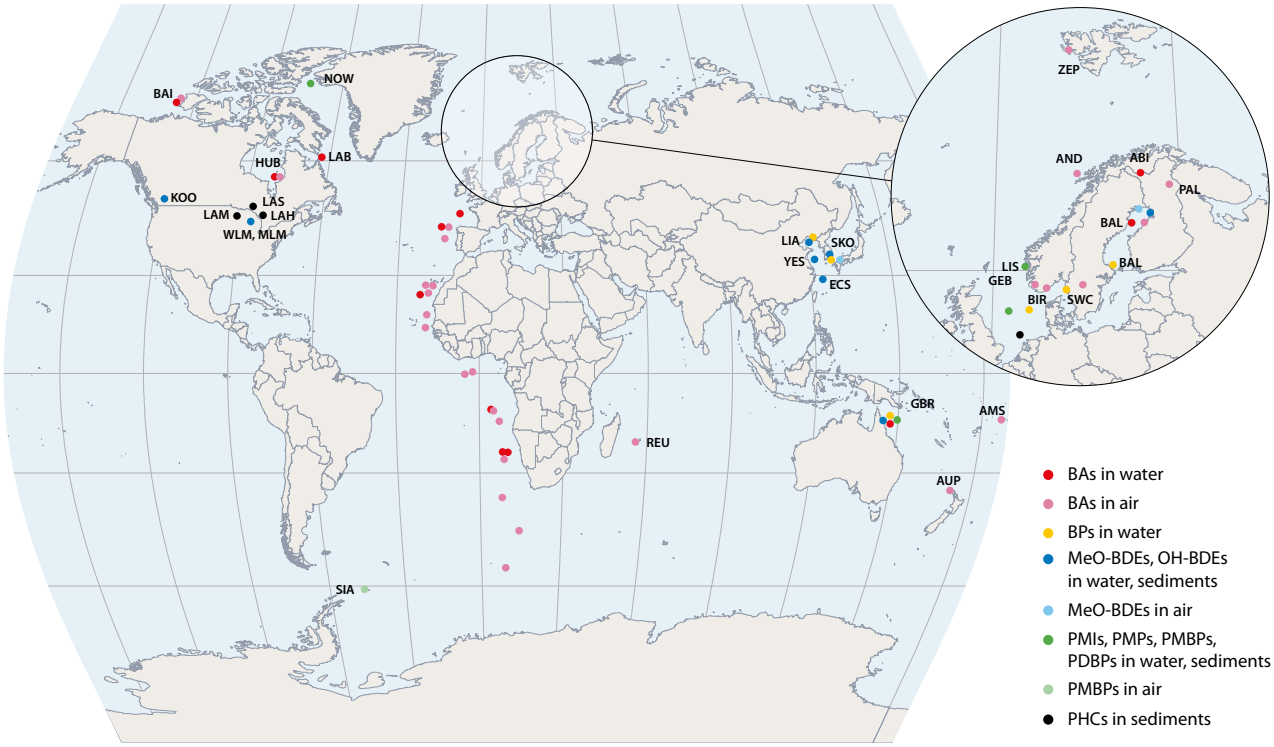


Figure 2.2.18. Locations where hHNPs have been reported quantitatively in air, water and sediments. See Box 2.2.1/Table 2.2.4 for expansion of chemical abbreviations. Location abbreviations: BAI = Banks Island, Canada; KOO = Kootany River, B.C., Canada; HUB = Hudson Bay, Canada; LAB = Labrador Sea, Canada; NOW = Northwater Polynya, Canada; LAM = Lake Michigan, U.S.A.; LAH = Lake Huron, Canada-U.S.A.; LAS = Lake Superior, Canada-U.S.A.; WLM, MLM = White and Muskegon Lakes, Michigan, U.S.A.; BIR = Birkenes, Norway; LIS = Lista, Norway; AND = Andøya, Norway; ZEP = Zeppelin Mountain (Ny Ålesund), Norway; BAL = Baltic Sea; ABI = Abisko, Sweden; PAL = Pallas, Finland; RÅÖ = Råö, Sweden; SWC = Swedish west coast; GEB = German Bight (North Sea); REU = Réunion; LIA = Liaodong Bay (Bohai Sea); ECS = East China Sea, YES = Yellow Sea; SKO = coastal South Korea (including Busan); GBR = Great Barrier Reef, Australia; AUP = Aupouri Peninsula, New Zealand; AMS = American Samoa; SIA = Signey Island, Antarctica. References available in Bidleman et al. (2020).

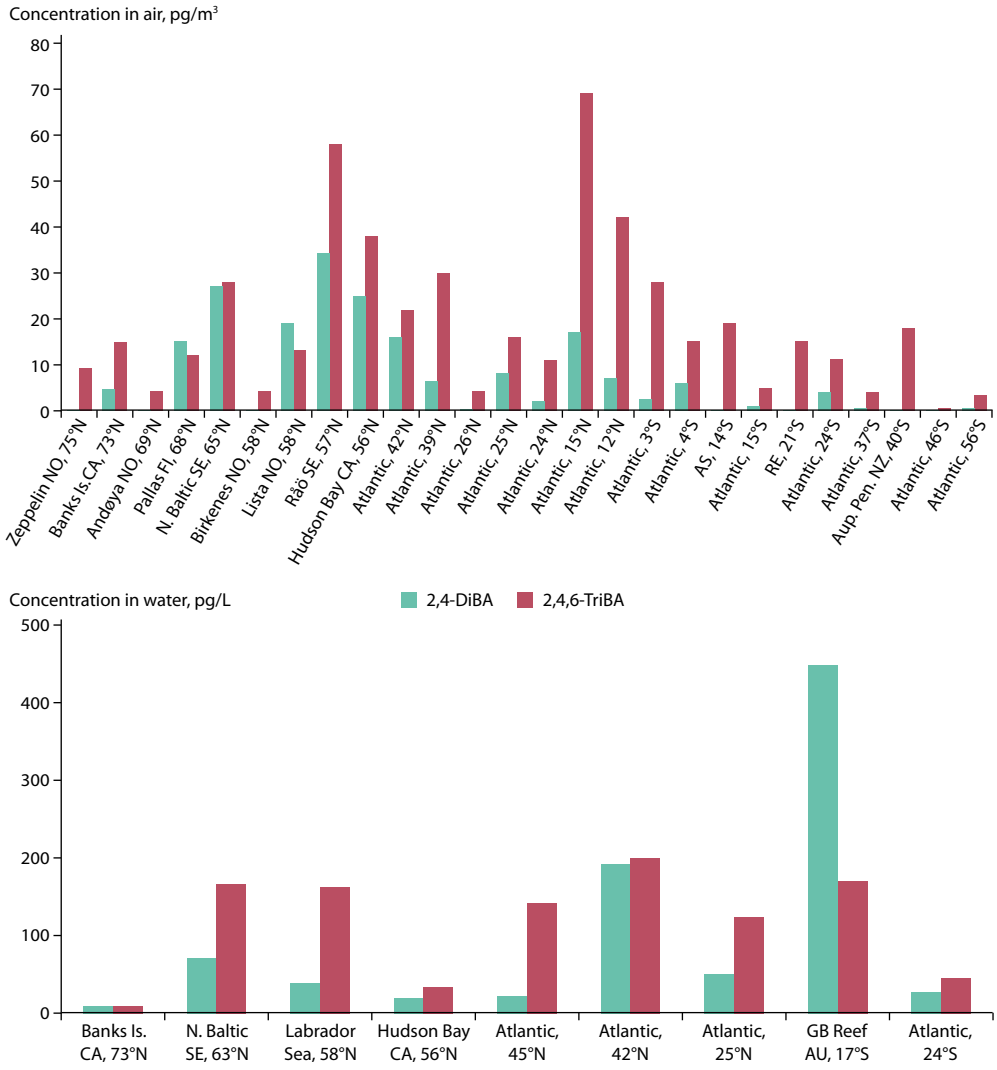


Figure 2.2.19 Distribution of 2,4-dibromoanisole (2,4-DiBA) and 2,4,6-tribromoanisole (2,4,6-TriBA) in air (top) and surface water (bottom) at indicated latitudes. See Figure 2.2.18 for locations and Bidleman et al. (2020) for data summary and references. Note the occurrence of these compounds at high latitude sites close to and within the Arctic.

The same organisms (bacteria, phytoplankton, and macroalgae) generate both nHCs and hHNPs. Production of these two compound classes is expected to respond similarly to climate-induced shifts in macroalgae range, phytoplankton and bacteria growth, and changes in chemical and physical stressors (Bidleman et al., 2020). How these changes will affect the levels and impact of hHNPs is unknown. Few temporal trends records for hHNPs exist which might be useful to infer climatic influence. So far, evidence for changing BA concentrations in air are weak or lacking (see above). There is some evidence for increasing methoxylated polybrominated diphenyl ethers (MeO-PBDEs) in recent times derived from the sediment record in the East China Sea, where MeO-PBDEs were correlated with phytoplankton lipid biomarkers (Fan et al., 2014).

Both nHC and hHNP emissions may also be affected by commercial macroalgae operations, with potential increased exposure to biota and humans (Haraguchi et al., 2016). Macroalgae are used for direct human consumption, in diets for farmed fish, in pharmaceuticals and personal care products, as stabilizers and emulsifiers, as "functional foods" that provide health benefits to people and animals, and as raw materials for biofuels (Holdt and Kraan, 2011; FAO, 2018). The macroalgae market is rapidly expanding, with a doubling of output between 2005-2015, and over 90% resulting from commercial operations (FAO, 2018). Such expansion may affect HNP emissions.

As climate change is expected to affect the production and emissions of hHNPs, baseline and temporal trend measurements in biotic and abiotic media are needed to evaluate future changes. Synergies can be sought through the analysis of hHNPs in air and water samples collected under long-term monitoring programs for POPs, and the use of archived biotic samples and sediment/ice cores to rebuild trends. While the nHCs scientific community is advanced in investigating sources, atmospheric and oceanic transport, and forecasting climate change impacts through modeling, these activities are nascent or non-existent for hHNPs. Collaborations between hHNP and nHC communities should be established, with joint measurement campaigns and an evaluation of hHNP transport, bioaccumulation, and fate in chemistry-climate models used for nHCs. Monitoring of hHNPs in producing species (i.e. macroalgae, phytoplankton, invertebrates) is also necessary to estimate existing stocks and temporal trends to better understand climate change impacts.

2.2.8 Conclusions

Contaminant pathways in the physical environment are changing; most notably the intra-Arctic transfer of POPs marked by re-mobilization of previously deposited 'reservoirs', including release from melting glaciers and permafrost (through slumping and erosion), as well as changing source types within and outside the Arctic. It is now apparent that the rate of decline of certain legacy POPs (particularly OC pesticides) in the Arctic atmosphere is slowing and can be attributed to revolatilization of 'old stocks' present in seawater, particularly with the decline in areal sea ice extent and changes in the cryosphere in general. This phenomenon has been observed at several of the air monitoring stations, but not all. It is important to recognize that climatic oscillations (e.g. AO) also affect the temporal trends of POPs in the Arctic atmosphere, as well as transport patterns with surface ocean currents. These oscillations provide wider teleconnections to temperate regions, and in a warming Arctic their influence on contaminant behavior and patterns of entry into the Arctic can be pronounced.

There are a number of contaminant transfer processes that are sensitive to a warming Arctic. These include the gas/particle distribution of semi-volatile compounds in the atmosphere, with atmospheric transport models simulating enhanced mobility for those chemicals shifted more towards the gas phase with warmer air temperatures. Enhanced deposition from the atmosphere (air-to-surface transfer) through increased scavenging of both gas and particle-bound chemicals brought about through higher rates of precipitation across large parts of the Arctic is likely, although empirical monitoring studies are limited. For the Arctic Ocean, increased primary productivity under climate change is enhancing the drawdown and transfer of contaminants from surface waters to deeper waters, through the process known as the 'biological pump'. The best evidence for this is from ship-based cruises in the western Arctic examining the atmospheric drawdown and transfer of PAHs from the surface ocean to deeper waters. A key feature of the changing cryosphere across the Arctic Ocean is the replacement of MYI by brine-rich FYI. This in turn is affecting contaminant dynamics in the sea ice system, as POPs and CEACs have been observed to accumulate, and even enrich in the brine fraction of FYI. This has implications for the subsequent exposure to sympagic organisms occupying brine channels and the lower parts of the ice in contact with the underlying seawater. As brine-rich FYI dominates ice coverage over large parts of the Arctic, the additional impacts of climate change, such as earlier or erratic thawing, could result in the more efficient delivery of contaminants to organisms at the base of the marine food web.

Climate change in the Arctic is clearly exacerbating intra-Arctic contaminant mobility and transfer between physical environmental compartments. There are now clear examples of contaminant release from glacier melt in the coastal fjords of Svalbard, as well as freshwater systems in the Canadian Arctic, particularly for watersheds dominated by glacier cover and melt (e.g. Lake Hazen, Ellesmere Island). These observations mirror the contaminant transfer observed with melting glaciers in mountainous areas in temperate regions. However, in the Arctic, re-mobilization of POPs in watersheds is also occurring due to permafrost degradation and erosion, resulting in generally higher and more variable contaminant concentrations in 'slump -affected' lakes in the western Canadian Arctic (where observational studies have been conducted). The relevance of such processes to biotic exposures in freshwater systems may be site- or regionally-specific, for example differing between high Arctic tundra and sub-Arctic boreal forest biomes, and therefore difficult to assess in other circumpolar regions. Site-specific processes in terrestrial and freshwater environments may introduce a stronger local or regional component into long-term time trends of POPs in the Arctic. Indeed, elevated concentrations of POPs in freshwater and marine systems as a consequence of this would then result in increased exposure to biota and respective food webs. Subsequent increases in POP concentrations in higher trophic level organisms are likely to be specific to certain geographic regions (where re-mobilization or altered contaminant pathways are clearly apparent). While increasing contaminant levels in these organisms may be a result of re-mobilization of contaminants, they may also be coincidently influenced by other ecological changes, including altered feeding habits from changes in prey abundance and/or behavior. These biological phenomena are explored in further detail in Chapter 2.3.

There is now evidence of indirect effects of climate change on Arctic and sub-Arctic systems, such as the increased frequency and magnitude of wildfires. Fires in the boreal forests of Canada, Alaska and Eastern Europe have resulted in short-term elevated air concentrations of PAHs and PCBs in the high Arctic. It is foreseeable that extreme weather events (e.g. severe rain events, snowstorms, and unseasonal warming in parts of the Arctic, that in turn can lead to forest fires or unusual melt events) will become more frequent due to climate change. Yet, knowledge of the role of such events on the overall transport and distribution pathways of POPs to Arctic terrestrial and marine environments is very limited.

There are additional emerging issues that are affecting contaminant dynamics in a warming Arctic. Microplastics are an important issue that is being addressed separately by AMAP (Box 2.2.2). However microplastic particles can transport POPs and CEACs, and thus serve as a likely source of these chemicals to Arctic systems. With regard to climate change-related effects, there is a lack of knowledge concerning the processes and rates of microplastic incorporation within sea ice, the resulting effect on sea ice properties, and the potential release of microplastics and associated contaminants entrained in the ice. Additionally, many hHNPs biosynthesized by bacteria, phytoplankton, and microalgae, possess POP-like properties and are present in the Arctic marine environment (Box 2.2.1). Globally their production is likely to increase with climate change and hence may provide an additional stress on higher trophic level organisms, alongside synthetic POPs/CEACs.

Increased human activities as an indirect effect of climate change (shipping, tourism, oil and gas development, fisheries) have the potential to increase local emissions within the Arctic. New primary sources (industries, communities, tourism) will contribute to a potential increase of CEACs, which clearly affect local areas in the vicinity of these sources. Elevated levels of some CEACs (PFAS, BFRs, OPEs, PAHs) have now been shown near Arctic communities. The relative proportion of contaminant input to the Arctic environment due to long-range atmospheric and/or oceanic transport versus local emissions

Box 2.2.2 Microplastic contamination and effects of climate change

Microplastics are tiny fragments of synthetic polymers (diameters <5 mm) arising from different sources. They are present in remote environments, including deep ocean sediments (Van Cauwenberghe et al., 2013; Bergmann et al., 2017a), terrestrial wilderness areas (Brahney et al., 2020), remote mountain lakes (Free et al., 2014; Zhang et al., 2016) and even Antarctica (Waller et al., 2017). Their presence in the Arctic arises through transport in ocean currents as well as the atmosphere (Evangeliou et al., 2020), although sources within the Arctic are also likely.

- In the Arctic, microplastic pollution has been discovered in sea ice (Obbard et al., 2014; Bergmann et al., 2017b; Peeken et al., 2018), snow (Bergmann et al., 2019), surface and sub-surface ocean waters (Lusher et al., 2015; La Daana et al., 2018) and deep-sea sediment (Bergmann et al., 2017a), as well as Arctic biota (Kühn et al., 2018; Morgana et al., 2018).
- A wide range of polymers, with many particles being in the lower size ranges (diameters <500 µm), have been identified in Arctic media, including in Arctic snow (Bergmann et al., 2019), suggesting multiple sources of microplastics, both local and long-range, exist (Evangeliou et al., 2020).
- Due to their presence in marine biota, their ability to sorb POPs and other pollutants, and potential to leach chemical additives and CEACs (Cole et al., 2011; Bakir et al., 2016), microplastics present a risk to Arctic ecosystems.
- Microplastics can accumulate in sea ice (Lusher et al., 2015; Geilfus et al., 2019), but particle concentrations show a very wide range in values, in part attributable to different measurement methodologies. For example, Obbard et al. (2014) observed particle concentrations of 0.04–0.24 particles/L in central Arctic Ocean sea ice, whereas much higher values of 1145–4270 particles/L were reported in sea ice from the Fram Strait (Peeken et al., 2018).
- Mesocosm studies have provided evidence that microplastic particles present at very high concentrations in sea ice and snow can alter the ice albedo and promote surface melt (Geilfus et al., 2019). However, there is a general lack of knowledge concerning the processes and rates of microplastic incorporation within sea ice, and any resulting effect on sea ice properties.
- Figure 2.2.20 illustrates a likely feedback loop of the effect of continued input of microplastics to the Arctic environment. It probably represents a worst-case scenario but highlights how increasing anthropogenic activities in the Arctic due to climate change could increase plastic pollution and enhance ice melt by reducing albedo, unless restrictions on plastic use and disposal are put into place.

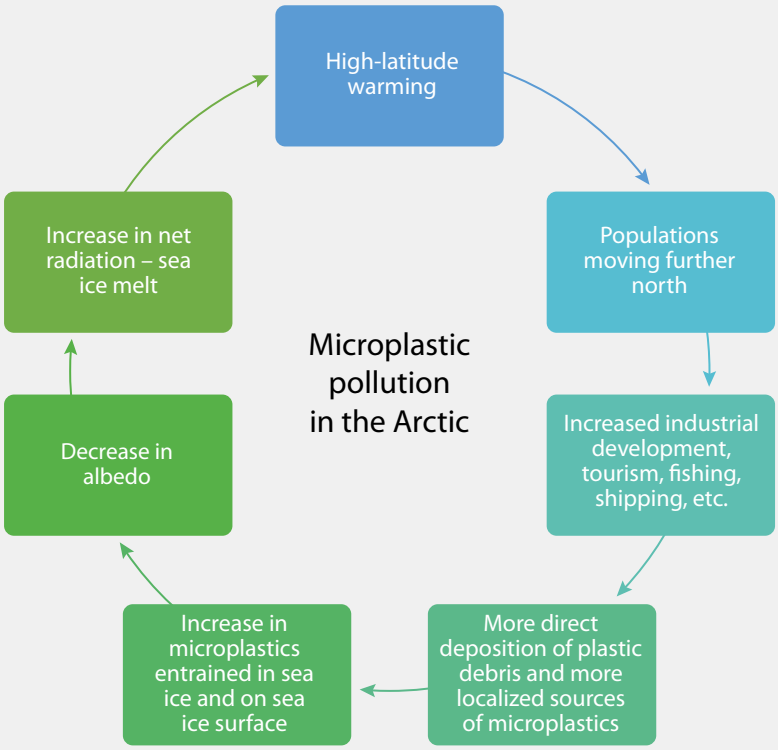


Figure 2.2.20 Feedback loop of climate change and microplastic pollution in the Arctic.

may change for certain chemicals due to significant changes in human activities as a result of warming, although further assessments are required to understand the contributions from these different sources. New CEACs are being identified in the Arctic atmosphere using cutting-edge analytical methodologies, although whether these chemicals arise through long-range transport into the Arctic or from local emissions (or a combination thereof) needs further investigation.

2.2.8.1 Findings and recommendations

Major observations of climate change influences on POPs and CEACs in the physical environment are summarized in Figure 2.2.21.

Finding: Changing human activities in a warming Arctic have altered the relative proportion of contaminant input to the Arctic environment from long-range atmospheric and/or oceanic transport versus local emissions. The nature of chemical usage and emissions has also changed (e.g. point sources of legacy POPs vs. diffuse sources of many CEACs; differences in physical-chemical properties of POPs and CEACs, etc.).

Recommendation: A reassessment of chemical hazard criteria that reflects changes in sensitivity to climate change is necessary to protect Arctic wildlife and humans from contaminants and contaminant mixtures that do not meet the Long-range transport, Persistence, Bioaccumulation and Toxicity criteria under the Stockholm Convention. In addition, more information is needed for CEACs, especially with respect to their environmental occurrence, physical-chemical properties, and transport and transformation processes, which ultimately will drive their distribution in the Arctic environment under a changing climate.

Finding: Retreating sea ice has enhanced air-water contaminant exchange. Increased river runoff and snow/ice melt has remobilized contaminants to marine and freshwater environments.

Recommendation: More research is needed to better understand the implications of accelerated melting and re-mobilization of contaminants on accumulation and exposure in marine and freshwater environments. Simultaneous multimedia assessments on contaminant redistribution (e.g. particulate bound vs. dissolved concentrations), and accumulation in food web organisms should be conducted to systematically quantify such impact.

Finding: Climate change is resulting in the replacement of multi-year ice with brine-rich first-year ice. Contaminants can accumulate, and even enrich, in the brine fraction of this new ice.

Recommendation: Further research encompassing a wider range of POPs and CEACs is needed to verify the replacement of multi-year ice with brine-rich first-year ice as a contaminant exposure pathway for sympagic organisms in both nearshore and open-ocean environments. The studies should include loadings of contaminants into ocean waters, as well as food web studies.

Finding: Studies on the re-mobilization of POPs due to permafrost degradation and erosion are currently limited to the Canadian Arctic. Biotic exposures in freshwater systems due to this process may be site- or regionally- specific, and therefore, cannot be applied to other Arctic regions. Site-specific processes in terrestrial and freshwater environments may introduce a

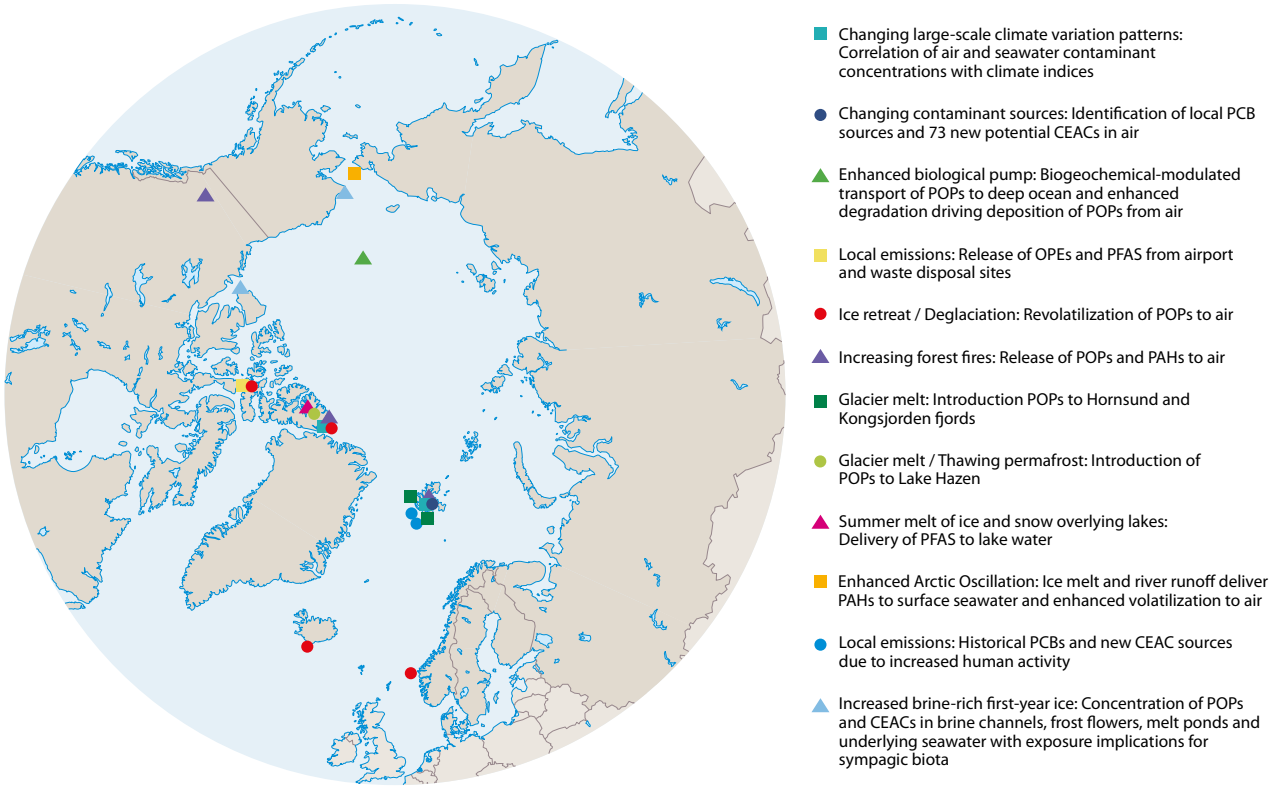


Figure 2.2.21 Locations of major observations of direct- and indirect- climate change influences on POPs and CEACs in physical environmental media of the Arctic.

strong local- or regional-component into long-term time trends of POPs in the Arctic.

Recommendation: It is important to identify key study locations in the Arctic where permafrost thaw and melting of terrestrial snow and glaciers impact aquatic environments (e.g. estuaries and lakes) and develop long-term studies on POPs and selected CEACs in these areas.

Finding: Increasing human and industrial activities in the Arctic due to warming will significantly increase the release of CEACs locally.

Recommendation: There is a need to re-evaluate the relative contributions of long-range transport versus local emissions to Arctic contamination for chemicals that might be increasingly used locally as a result of climate change-related impacts. CEACs, widely used substances in consumer and industrial products as reviewed by AMAP (2017), in particular, should be included in temporal trend monitoring. These studies should take into account the potential for current-use chemicals to have different sources and physical-chemical properties, which might call for monitoring in other media, such as seawater, soils, or vegetation.

Finding: The release of halogenated natural products (nHCs and hHNPs), which are generated by organisms (bacteria, phytoplankton and macroalgae), is expected to respond similarly to the climate-induced shifts in growth and range of these organisms, their chemical/physical stressors, as well as changes in locations of commercial macroalgae operations due to climate change.

Recommendation: It is recommended to expand monitoring of hHNPs, improve quality assurance in the measurements, gather and collaborate with the nHCs scientific community to advance knowledge on the abundance and distribution of hHNPs, and forecast climate change influences. Some hHNPs have toxic properties similar to those of anthropogenic contaminants, therefore it is recommended to evaluate the toxicity of natural-anthropogenic compound mixtures.

Finding: Microplastics, as well as POPs and other pollutants associated with them, have been discovered in Arctic sea ice, snow, ocean waters and deep-sea sediments, and Arctic wildlife. High concentrations of microplastic particles on sea ice and snow can alter the ice albedo and promote surface melt.

Recommendation: The role of microplastic particles present in snow and ice as climate forcers requires further study. In addition, the role of microplastics as vectors for contaminant transport, and the exposure of Arctic biota to microplastics needs further investigating. Future work under the AMAP POPs Expert Group and the AMAP Litter and Microplastics Expert Group should be better coordinated in this respect.

Finding: Extreme weather events (e.g. severe rain events, snowstorms, and unseasonal warming in parts of the Arctic – that in turn can lead to forest fires or unusual melt events) will become more frequent due to climate change.

Recommendation: Studies are needed that link records of extreme weather events to the fate of contaminants in the Arctic. The importance of such events on the emission of POPs and CEACs, and their deposition and distribution to Arctic terrestrial, freshwater, and marine environments should be quantified.

2.3 How does global climate change influence accumulation of POPs and CEACs in Arctic food webs?

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2.3.1 Introduction

Global climate change affects physical, biological, and ecological processes in the environment, and thus has potential to influence the uptake and fate of persistent organic pollutants (POPs) in biota and food webs through multiple mechanisms (AMAP, 2003). As climate change is occurring most quickly and with the greatest amplitude in polar regions, and is predicted to continue to do so under future scenarios (IPCC, 2019), the effects of climate change on POP bioaccumulation through food webs are best studied in these ecosystems.

The potential effects of climate change on the food web accumulation of contaminants in polar regions was first identified just after the turn of the century (AMAP, 2003). As summarized in UNEP/AMAP (2011), the predicted effects of climate change on contaminant availability and subsequent uptake and bioaccumulation were investigated with various modeling studies constrained by Intergovernmental Panel on Climate Change (IPCC). Prior to that, the focus was predominantly on understanding how the unique characteristics of polar ecosystems influence pollutant uptake and accumulation (AMAP, 1998; Borgå et al., 2004). In the decade that followed, national and international efforts including the International Polar Year, obtained empirical data on the effects of climate change on POP food web accumulation in the Arctic, as summarized for marine ecosystems by McKinney et al. (2015).

The bioavailability, uptake, bioaccumulation, and fate of POPs in Arctic ecosystems are affected by a multitude of complex and interacting factors, including environmental conditions, such as temperature, precipitation, and presence of sea ice; POP physicochemical properties, such as hydrophobicity and recalcitrance; and a number of biological factors, including physiological and ecological variables such as phenology, energy allocation and lipid dynamics, reproductive strategy, body size, age, sex, life-cycle stage, biotransformation capacity, habitat use, migration and feeding ecology (Figure 2.3.1;

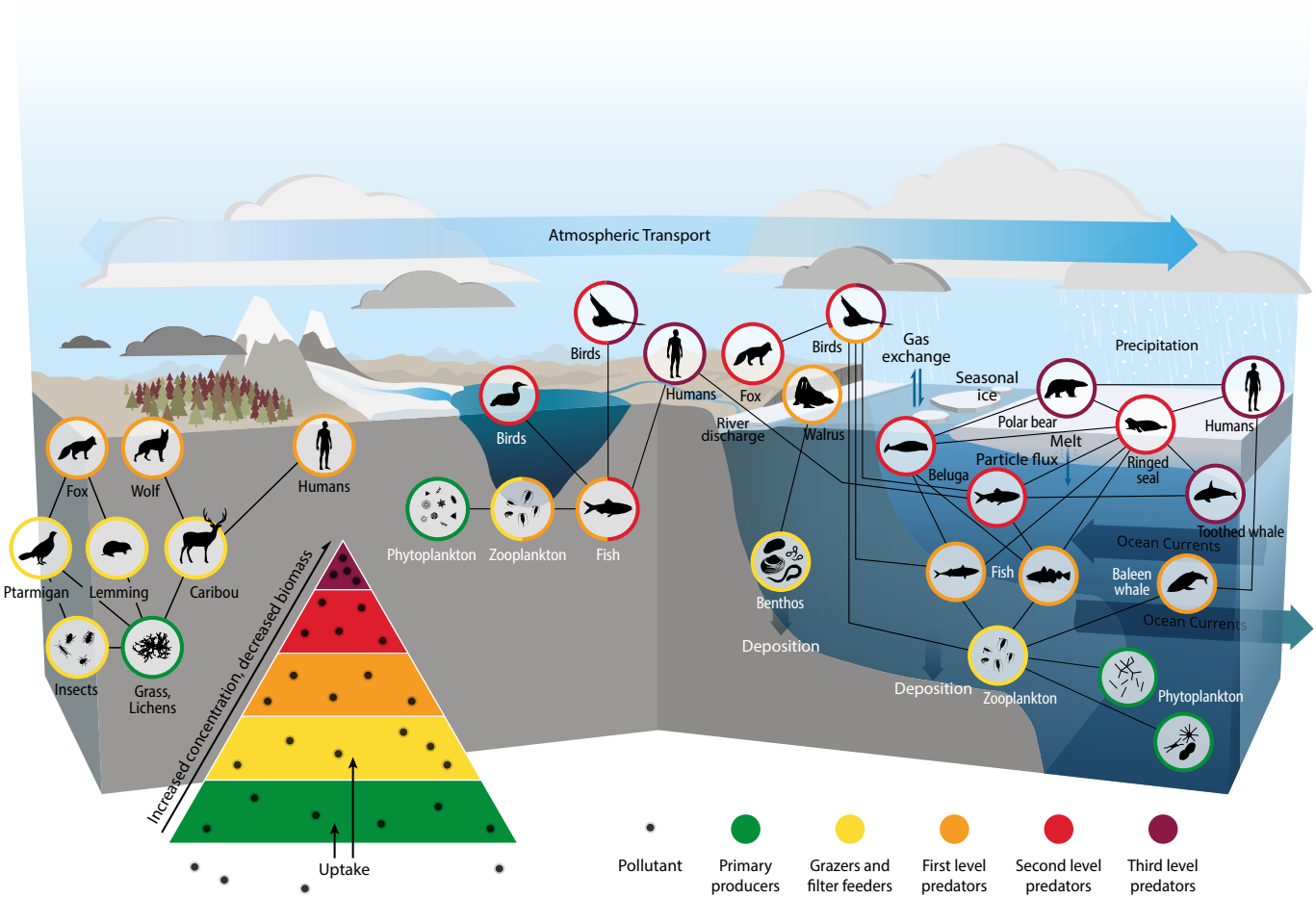

Figure 2.3.1 Pictorial representation of the physical and ecological pathways of persistent organic pollutants moving into and through the Arctic environment. Global climate change may influence the depicted pathways, and in turn, the dynamics and fate of Arctic contaminants. Adapted from Macdonald et al. (2005a).

reviewed in Borgå et al., 2004). POPs taken up by Arctic biota may therefore be directly impacted by physical changes in the environment, such as changes in sea ice extent, thickness, age (i.e. multi-year vs. first-year ice), length of the ice season in seasonal ice regions, presence of glaciers, snow cover and permafrost, as well as increased freshwater runoff and altered nutrient availability (Perovich and Richter-Menge, 2009). Additionally, biological and ecological changes, including increased primary production, reduced population sizes of some ice-dependent species, northward range shifts of sub-Arctic and temperate marine and terrestrial species, and altered trophic structuring can influence POP dynamics in Arctic food webs (Post et al., 2009, 2013; Fossheim et al., 2015; Pecl et al., 2017).

In this chapter, we review the current state of knowledge regarding climate change-driven physical and ecological factors impacting contaminant fate and accumulation in Arctic species and food webs, with a focus on the POPs and CEACs defined in Chapter 1 (Table 1.1). We summarize findings from available studies published over the past decade since the previous UNEP/AMAP (2011) report to address two overarching questions:

Table 2.3.1 Summary of predicted and empirical evidence, conclusions and knowledge gaps for global climate change-related effects on contaminant exposure and bioaccumulation in Arctic food webs. Arrows signify evidence of increased (↑), decreased (↓), mixed (↑↓), or no influence (↔) of environmental or ecological change.

Climate-related change	Effect on contaminant exposure and bioaccumulation in Arctic biota and food webs	
	Predicted	Empirical/Correlational
Abiotic changes		
Increased temperature ↑↓ Chemical fate and behavior	↑ LRT ↑ Mobilization of POPs from primary and secondary sources ↔ Contaminant uptake and elimination rates in fish (modeled) ↔ Bioaccumulation in Barents Sea food web (modeled)	↑ LRT ↑ Mobilization of POPs from primary and secondary sources
Altered climate patterns ↑↓ Atmospheric circulation ↑↓Ocean circulation ↑↓ Wind and precipitation patterns	None identified	POPs ↑ in gulls, ringed seals and landlocked Arctic char with AO+/NAO+ in preceding year POPs ↓ in gulls with AO+ in current winter POPs and/or OCPs ↑ or ↓ in murres and/or fulmars 4-9 years after high rainfall and/or NAO+ or NAO- Declining trends of some POPs in ringed seals slowed in years with greater influx of Atlantic currents
Reduced sea ice ↓ Sea ice cover ↓ Sea ice season ↓ Multi-year ice ↑ First-year ice	None identified	Pesticides ↑↓ in zooplankton and ice fauna in areas with more sea ice in the Barents Sea Lipophilic POPs ↑ in ringed seals in years with early sea ice break up in the Beaufort Sea PCB-153 ↑ in ringed seals in years with shorter duration of sea ice in West Greenland Lipophilic POPs ↑ in ringed seals with high total or multi-year sea ice in the Canadian Arctic Lipophilic POPs ↑ in polar bears in areas and seasons with low sea ice extent around Svalbard Lipophilic POPs and PFAS ↑ in polar bears with high quality sea ice habitat β-HCH and PFAS ↑ in Arctic foxes with increasing sea ice cover
Increased terrestrial runoff to surface waters ↑ Glacial and snow meltwater discharge ↑ Precipitation and associated run off	↑ Mobilization of POPs from secondary sources ↑ Release of POPs to lakes and coastal regions ↑ Darkening and browning of lakes and rivers and coastal waters leading to ↑↓ changes to trophic structure and transfer of POPs	POPs ↔ in Arctic char in waters with glacial discharge or increased turbidity POPs ↓ in littoral amphipods with increasing run-off during melt season

(1) What are the climate change-driven physical and/or ecological processes influencing POPs in Arctic wildlife? and (2) How is climate change influencing the levels of POPs in Arctic biota and food webs? The processes described herein are connected to those reviewed in the chapters on the abiotic environment (Chapter 2.2) and temporal trends in biota (Chapter 2.4). The main findings of this chapter are summarized in Table 2.3.1.

When examining the effects of climate change on food web accumulation of POPs, it is important to recognize that observed changes are the cumulative result of multiple, coinciding processes, including changes in POP emissions and sources (addressed in Chapters 2.1 and 2.2), environmental changes that affect contaminant exposure, bioavailability and accumulation (reviewed herein), and alterations of food web structure, transfer efficiency of contaminants, animal growth, activity and reproduction that affect food web accumulation (also reviewed herein). As a result of these collective changes, temporal trends in biota can be affected (addressed in Chapter 2.4) that can have implications for regulatory policies.

References	Conclusions	Knowledge Gaps
Chapter 2.1 Chapter 2.2	Changes in sources and emissions could impact exposure in biota and will be important for interpreting observed changes	
Gouin et al., 2013	Climate-related temperature increases may have minimal impacts on bioaccumulation, due to species-specific biotransformation capacities and thermal tolerances	
Borgå et al., 2010	Seasonal variations in temperature could influence bioaccumulation on smaller time scales	
Bustnes et al., 2010 Rigét et al., 2013 Houde et al. 2019 Cabrerizo et al., 2018b Foster et al., 2019	Higher concentrations of POPs in Arctic biota observed following NAO+/AO+ states and influx of Atlantic oceanic currents suggest higher exposure following transport of air and water masses from North America and Europe	Unknown underlying mechanisms of statistical relationship between AO/NAO and POPs in biota No data for terrestrial species Effects of intermittent extreme weather events unknown Little understanding of the variance allocation to confounding factors for large regional differences
Carlsson et al., 2014 Borgå et al., 2002a,b Gaden et al., 2012 Rigét et al., 2013a Houde et al., 2019 Tartu et al., 2017b Tartu et al., 2017a Routti et al., 2017 Tartu et al., 2018 Blévin et al., 2020 Andersen et al., 2015a Routti et al., 2017	Melting of multi-year sea ice may release stored contaminants into the Arctic marine food web or may dilute and reduce contaminants at base of the food web Separating the direct effects of declining sea ice cover on biota POP levels from the indirect effects from changes in food web structure and function are difficult	Contrasting results indicate sea ice may be a source of POPs/CEACs or dilute POP/CEACs Little is known regarding the match/mismatched of ice melt and biological production and the effect on POP/CEAC accumulation Differences between POPs and CEACs Processes largely unknown apart from biotic processes
Cabrerizo et al., 2018b	Findings are based on the effects of glacial sources on Arctic freshwater biota and food webs are limited	Effect of terrestrial-deposited snow and melting glaciers on POP accumulation in receiving freshwater, coastal marine, and terrestrial food webs
Skogsberg, 2019	Increased runoff may dilute and reduce bioavailable fraction of contaminants at base of food web	Net result of terrestrial runoff on contaminant accumulation in Arctic biota still unknown due to large seasonal variations

Table 2.3.1 continued

Climate-related change	Effect on contaminant exposure and bioaccumulation in Arctic biota and food webs	
	Predicted	Empirical/Correlational
Freshwater hydrology and permafrost thaw ↓ Ice cover ↑ Permafrost thaw ↑ Suspended sediment and water turbidity ↑ Primary productivity	None identified	POPs ↑ in sedimentary organic matter of slump-affected lakes POPs ↑ in amphipods from slump-affected lakes
Seasonality changes ↑ Earlier ice melt ↑ Open water season ↓ Later ice freeze up	None identified	PCBs ↑ in later hatching geese eggs POPs ↑↓ in zooplankton, fish and seabirds with seasonal changes in an Atlantic fjord
Ecological changes		
Increased primary production in marine and freshwater	↓ bioaccumulation due to less bioavailability of dissolved POPs Systematic ↓ bioaccumulation due to lower bioconcentration in phytoplankton	
Changing species interactions: Marine-to-terrestrial prey shifts	None identified	↓ 30% in CHL for Southern Beaufort Sea polar bears; decrease not reaching statistical significance for PCB, HCH, ClBz and increase not reaching statistical significance for DDT ↓ PCBs, OCPs, PBDEs, PFAS in Arctic foxes from Svalbard, but long term changes in diet did not affect temporal trends of POPs ↓ PCBs, OCPs, PBDEs, PFAS in polar bears from Svalbard, but long term changes in diet did not affect temporal trends of POPs
Changing species interactions: Arctic to sub-Arctic prey shifts	None identified	↑ instead of ↓ trend for PCBs and CHLs in Western Hudson Bay polar bears, faster rate of ↑ for PBDEs and β-HCH, faster rate of ↓ for DDTs Non-significant slower rate of ↓ for PCBs and CHLs in East Greenland polar bears, non-significant faster rate of ↑ of PBDEs. ↔ effect for HCB and DDTs ↑ biomagnification when transient/sub-Arctic species included in the food web
Changes in energy intake/ expenditure: ↑ Energy needs ↓ Body condition	None identified	↑ POPs during incubation in eiders with low body condition ↑ POPs in polar bears and Arctic foxes from Svalbard, but long-term changes in body condition do not affect temporal trends of POPs ↑ POPs in polar bears in the Barents Sea ↓ Sea ice led to ↑ energy expenditure and ↑ Hg in seabirds due to more time flying, less time underwater, and deeper and longer dives
Changes in behavior and migration patterns	↑ Exposure expected due to migration and higher trophic level diet (i.e. elevated POP levels in migratory seabirds)	Delayed migration timing and routes affect contaminant transfer to offspring/eggs: heavier PCBs ↑

AO: Arctic oscillation; CC: climate change; CHL: chlordane; ClBz: chlorobenzene; CEAC: chemical of emerging Arctic concern; DDT: dichlorodiphenyltrichloroethane; HCH: hexachlorocyclohexane; LRT: long-range transport; NAO: North Atlantic oscillation; OCP: organochlorine pesticide; PBDE: polybrominated diphenyl ether; PCB: polychlorinated biphenyl; PFAS: per- and polyfluoroalkyl substances; POP: persistent organic pollutant

References	Conclusions	Knowledge Gaps
Eickmeyer et al., 2016	Findings based on limited studies from small lakes	Broader suite of lakes need to be studied
D'Onofrio, 2014	Sedimentation of organic matter and particulates facilitate the delivery of associated POPs/CEACs from the water column to benthic communities	
Hitchcock et al., 2019a	Changes in seasonal timing may impact migration routes and/or relative time spent at more contaminated wintering grounds	Drivers of seasonal contaminant shifts in food webs (e.g. sea ice and meltwater) still unknown and are large confounding factors
Hallanger et al., 2011a,b,c	Seasonal effects may differ between POPs in relation to physiochemical properties (e.g. volatility, water solubility)	
Borgå et al., 2010	Current understanding of the net effect of increasing primary production on contaminant accumulation is limited	Effects of regional boosts in primary production and biodilution are unknown Little is know regarding the cascading effects on trophic interactions in the food web
Atwood et al., 2017 (Southern Beaufort Sea polar bear) Andersen et al., 2015a Routti et al., 2017 Chapter 2.4 Tartu et al., .2017a, b Lippold et al., 2019 Routti et al., 2017	Studies to date suggest a shift from marine to terrestrial prey is associated with decreasing POP levels	Knowledge of the mechanisms and processes underpinning observed correlations is limited
McKinney et al., 2009 (Western Hudson Bay polar bear) McKinney et al., 2013 (East Greenland polar bear) McKinney et al., 2012	Difficult to conclude as effects appear to depend on the particular prey shift occurring and may be contaminant-specific	Effects of climate-related changes in food webs and species interactions on contaminant accumulation are largely unknown and would benefit from future application of modeling approaches
Bustnes et al., 2012 Baert et al., 2013 Tartu et al., 2017b Andersen et al. 2015a Lippold et al., 2019 Routti et al., 2017 Blévin et al. 2020 Amélineau et al., 2019 Elliott and Fernie 2019	Difficult to conclude as most studies focus only on the relationship between body condition and POP levels (e.g. lipid depletion increasing POP concentrations)	Understanding of the effects of climate-related changes in behavior and migration on POP/CEAC accumulation is limited
Baert et al., 2013	Models suggest higher POP concentrations in migrating seabirds	
Hitchcock et al., 2019a	Behavior and migration changes lead to differences in diet and condition which affect POP/CEAC exposures	

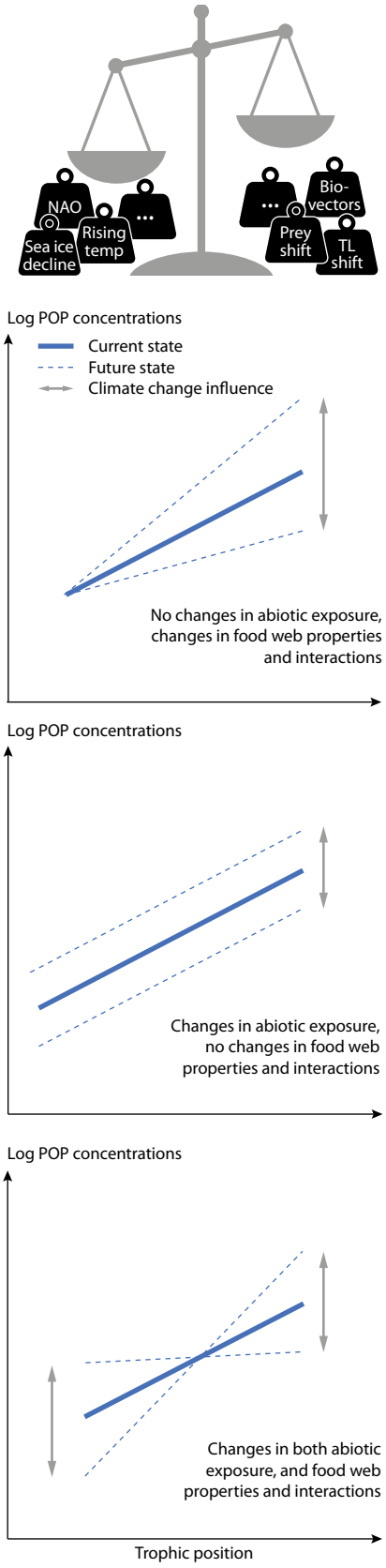

Figure 2.3.2 Conceptual figures showing the concentrations of POPs in relation to trophic position, in the case that climate change influences food web properties or interactions (upper), abiotic exposures (middle), or both food web properties/interactions and abiotic exposures (lower). The solid line represents the present log-normal relationship between the trophic position of the organisms and accumulated contaminant concentrations, while dotted lines reflect the projected window of biomagnification because of the influence of climate change. NAO = North Atlantic Oscillation; TL = trophic level.

Climate change is projected to concurrently alter many of these processes, potentially resulting in decreased or increased contaminant exposure at the base of the food web and changes to food web accumulation (Figure 2.3.2). Mechanistic process-oriented models can add to the understanding of how the changing contributions and variations of different parameters such as primary and secondary emission sources, transport processes, temperature, lipid dynamics, and food web structure, both alone and in combination, could affect the uptake, elimination and overall bioaccumulation of contaminants (AMAP, 2016a).

Further, we address how climate change, in combination with other stressors might alter POP toxicity (e.g. climate-induced toxicant sensitivity). Finally, we provide conclusions on our current understanding of climate change influences on POPs in Arctic species and food webs, identify knowledge gaps, and provide recommendations for future research and monitoring. Research findings for each topic are reviewed in order, from low- to high- trophic levels for marine, terrestrial and freshwater ecosystems, when studies were available, and include results from empirical and model-based studies.

2.3.2 How do physical changes to the environment affect POP exposure and bioaccumulation in Arctic biota?

2.3.2.1 Temperature

Temperature is a central abiotic driver in the environment that regulates and limits biological processes (e.g. development, growth, reproduction), and plays a key role in contaminant dynamics, affecting the physical-chemical properties of a chemical, as well as the biological and physiological processes regulating xenobiotic exposure (i.e. uptake, metabolism, and elimination), especially in poikilothermic organisms. Temperature is also a main driver of climate change, and is expected to continue to increase in the future. Under medium- to high- greenhouse gas emission scenarios, Arctic fall-winter temperatures of 4 to 5°C above end of 20th century temperatures are predicted for mid-21st century (AMAP, 2017b). To date, research on the direct relationship between temperature and POP uptake and elimination rates in Arctic biota is still scarce (Alava et al., 2017). Uptake and elimination are both likely to increase with increasing temperatures due to an elevation in metabolism (e.g. Noyes and Lema, 2015), however, this relationship may be limited by species-specific biotransformation capacities. In one study, the bioaccumulation potential of POP-like and hypothetical chemicals modeled in the food chain of a non-Arctic fish species, the round goby (*Apollonia melanostomus*) projected only a minimal impact of climate change on temperature-dependent uptake and elimination rates (Gouin et al., 2013). This study stressed the seasonal variation of bioaccumulation, and that when the environmental temperature crosses the species-specific minimum or maximum critical temperature for various physiological functions, uptake and elimination rates may be greatly affected (Figure 2.3.3). The resulting effect on bioaccumulation depended on the season, with higher accumulation in spring and autumn, and lower accumulation

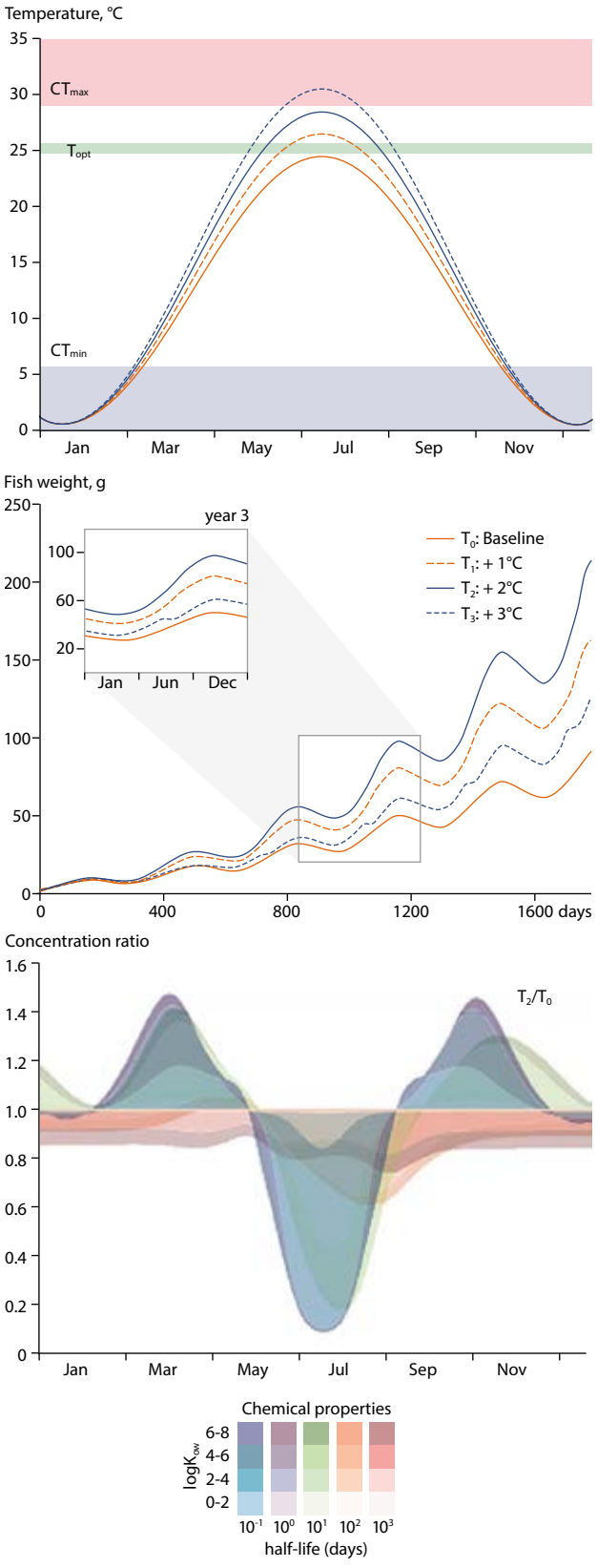

Figure 2.3.3 Modeled effects of climate change on bioaccumulation in fish (round goby, *Apollonia melanostomus*) in Lake Erie, showing the variation in bioaccumulation occurring with changes in seasonal temperatures, chemical octanol-water partitioning coefficients (K_{OW}) and chemical half-lives. Shown are the interactions of projected environmental temperature scenarios (baseline Lake Erie annual temperature (T_0) plus one (T_1), two (T_2), or three (T_3) degree(s) Celsius) with (upper) species thermal ranges (lower critical thermal limit (CT_{min}), upper critical thermal limit (CT_{max}), optimal temperature for growth (T_{opt})) and (middle) bioenergetics impacting growth, resulting in (lower) extremely seasonal bioaccumulation, which can exceed or subceed baseline thresholds. Adapted from Gouin et al. (2013).

in summer when growth was high. Similarly, a modeling study of the Barents Sea food web concluded that IPCC-predicted annual temperature increases due to climate change would elevate some processes and decrease others, resulting in only small net effects on bioaccumulation (Borgå et al., 2010).

In addition to its effects on chemical uptake and elimination rates, temperature also affects the bioavailability of, and thus exposure to, contaminants in the Arctic. Warmer temperatures can increase mobilization and long-range transport of POPs from primary emissions (see Chapter 2.1; Lamon et al., 2009) and re-mobilization from secondary sources, such as soil, melting permafrost, seawater, glacial ice and sea ice (see Chapter 2.2; Ma et al., 2011, 2016). For example, Cabrerizo et al. (2018b) found indications that soil from Resolute Lake in the high Canadian Arctic might act as net source of polychlorinated biphenyls (PCBs) to the atmosphere, demonstrating re-mobilization from secondary sources. Additionally, long-term temporal trends of PCBs in Arctic air measured at Alert (Canada), Stórhöfði (Iceland), Zeppelin (Svalbard) and Pallas (Finland), between 1993 and 2012 show a lower rate of decline in recent years and an increase of PCBs in air after the year 2000 (Hung et al., 2016). This increase may be connected to re-emissions of PCBs from secondary sources (e.g. oceans, melting ice and permafrost) due to higher ambient temperatures. The changing of environmental contaminant sinks to contaminant sources could have major effects on the exposure, and thus bioaccumulation potential of POPs in wildlife.

It is important to note that both observed and predicted changes in the environmental fate of chemicals strongly depend on compound-specific physical-chemical properties, such as volatility and water solubility. Consequently, even closely related compounds may respond differently to temperature changes. For instance, a review of temporal trends of hexachlorocyclohexanes (HCHs) found 80% of the long-term time series published for α-HCH in the Arctic show a decreasing trend, whereas no significant trends were observed in 64% of the long-term time series reported for β-HCH (Rigét et al., 2019). Both chemicals were phased out in the early 2000s, but their environmental concentrations vary due to differences in their Henry's law constants, which change with temperature, and long-range transport (LRT) processes. α-HCH is transported via air, whereas β-HCH is transported via ocean currents, and as a result, there is a time-lag for β-HCH entering the Arctic compared to α-HCH.

Exposure is also dependent on an organism's biotransformation capacity for specific chemicals. Biotransformation capacities can vary between species and individuals in accordance with a number of factors, including physiological differences (inclusive of thermal tolerances), as well as variations in life history strategies and age.

2.3.2.2 Climate patterns

Large-scale climate patterns (i.e. atmospheric circulation, oceanic currents, wind and precipitation patterns), along with local and regional weather, influence the LRT of POPs (Chapter 2.1; Macdonald et al., 2005a; Ma et al., 2016), and thus their concentrations in Arctic abiotic compartments

(Chapter 2.2), and their subsequent uptake and accumulation in Arctic wildlife. Yet, as climate changes extend across decadal time scales and geopolitical boundaries to impact ecological systems in complicated and interlinked ways, it is not necessarily easy, or even feasible, to identify individual meteorological variables influencing biotic POP levels. For this reason, metrics based on a combination of variables and reflective of broad-scale climate variations, such as climate oscillation indices, are often used to explore associations between climate and ecological changes, including POP temporal trends in biota (see Chapter 2.4). In particular, the Arctic Oscillation (AO) and North Atlantic Oscillation (NAO) indices have been associated with changes in environmental contamination in Arctic studies; for a description of these indices refer to Box 1.1 found in Chapter 1.

Modeling and correlative studies strongly suggest that climate change affects the uptake and accumulation of POPs in Arctic wildlife through altered wind and precipitation impacts on contaminant transport or wet deposition (see Chapter 2.1). Correlations have been documented between the AO or NAO indices and contaminant trends in glaucous gulls (*Larus hyperboreus*) in the Norwegian Arctic and ringed seals (*Pusa hispida*) in the Greenlandic and Canadian Arctic (Bustnes et al., 2010; Rigét et al., 2013a; Houde et al., 2019). Glaucous gulls from Bjørnøya, Svalbard, showed positive relationships between POP concentrations and AO conditions of the preceding summer and preceding winter (Bustnes et al., 2010). Concentrations of POPs in ringed seals from West Greenland were also positively associated with winter AO conditions (Rigét et al., 2013a). Similarly, POP concentrations in ringed seals from the Canadian Arctic Archipelago and Hudson Bay were positively related to AO, and NAO states, respectively, in the preceding year (Houde et al., 2019). These findings indicate that POP concentrations were higher in gulls and seals following years with greater air mass transport from North America and Europe toward the Arctic. In glaucous gulls, negative relationships between concentrations of POPs and AO in the current winter were suggested to be related to changes in diet or overwintering areas (Bustnes et al., 2010). The cold winters associated with a negative AO phase (AO-) may decrease the availability of high trophic level prey during the following summer; alternatively, cold winters may force glaucous gulls to migrate further south, exposing them to higher levels of POPs.

Climate-related increases in precipitation were previously predicted to increase the deposition of POPs through scavenging (AMAP, 2003). Consistent with this prediction, organochlorine contaminant concentrations in thick-billed murre (*Uria lomvia*) and northern fulmar (*Fulmarus glacialis*) eggs in the Canadian Arctic were strongly related to the NAO and/or rainfall amounts between 1975 and 2014 after controlling for differences in chemical partitioning characteristics, feeding ecology, and chemical emissions, which explained the majority (70% in murres; 77% in fulmars) of variability in egg organochlorine contaminant concentrations (Foster et al., 2019). More specifically, years of higher rainfall were followed by years with thick-billed murre eggs having higher concentrations of chlorobenzenes, octachlorostyrene, dieldrin, DDE and most PCBs, and lower concentrations of heptachlor epoxide, oxychlordane, *cis*-nonachlor, PCB-170 and PCB-180. In fulmars, years experiencing positive NAO phase (NAO+) conditions, when industrial chemicals show enhanced transport to the Arctic from eastern North America and Eurasia, were followed by years when eggs contained higher concentrations of chlorobenzenes, *trans*- and *cis*-nonachlor, dieldrin, photomirex and mirex. In contrast, negative NAO phase (NAO-) conditions were associated with birds laying eggs with lower concentrations of these chemicals. A four- to nine-year time lag occurred between the occurrence of the climate pattern and the apparent influence on POP concentrations deposited in the bird eggs (Foster et al., 2019). POP concentrations in landlocked Arctic char (*Salvelinus alpinus*) from the Canadian Arctic were also positively correlated with interannual variations of the NAO (Cabrerizo et al., 2018b). Greater amounts of precipitation will also impact freshwater ecosystems though increased terrestrial runoff as discussed in further detail in Section 2.3.2.4.

Changes in the movement of water masses (e.g. oceanic currents) can also influence contaminant transport to, and within, the Arctic, and thus affect exposure to biota. From 1994 until 2010, some POPs, including PCB-52, PCB-153, *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE), hexachlorobenzene (HCB), and α- and β-HCHs decreased in the blubber of ringed seals from central West Greenland (Rigét et al., 2013a). β-HCH, a chemical known to undergo long-range oceanic transport, showed the slowest decline, which was correlated to changes in salinity resulting from the dominance of the certain currents around Greenland. That is, years dominated by warm, saline water experienced a greater influx of the Irminger current, an Atlantic water branch, around West Greenland, and potentially with it, a higher influx of ocean-transported POPs. Similarly, an increased flux of warm, saline Atlantic water into the European Arctic (Polyakov et al., 2017) will likely increase oceanic contaminant transport into the High Arctic. Passive samplers deployed at different depths in the Fram Strait, an important deep-water channel to the Arctic Ocean, revealed a net influx of 0.16 megagrams of PCBs per year from the Atlantic into the Arctic, and a net export of HCB and HCH from the Arctic into the Atlantic Ocean, highlighting the continuous omnipresence of these chemicals (Ma et al., 2018).

2.3.2.3 Sea ice

Early modeling efforts forecasted that less sea ice and more open water would increase the exchange of chemicals from the atmosphere to surface waters, especially for POPs still undergoing net loading to the Arctic Ocean, like PCBs and toxaphene (Macdonald et al., 2005a). A comparison of pesticide concentrations in zooplankton (*Calanus* spp.) found higher chlordane (CHL) concentrations, but lower HCB concentrations, in those from ice-covered Arctic water masses than those from Atlantic water masses without ice cover (Carlsson et al., 2014).

Several studies have shown relationships between sea ice parameters and POP concentrations in marine mammals (Table 2.3.1, Box 2.3.1), which may be related to various

Box 2.3.1 Case Study: Sea ice conditions and contaminant exposure in coastal and offshore polar bears in the Barents Sea

Polar bears' main habitat is sea ice that they use for hunting seals and for migration. The marginal ice zone with intermediate sea ice concentrations is particularly preferred by polar bears (Lone et al., 2018). Due to climate change, polar bear habitat is undergoing drastic changes (Durner et al., 2009; Stern and Laidre, 2016; Lone et al., 2018). Over the last three to four decades the sea ice-free period has lengthened by three to nine weeks in most regions and by seventeen weeks in the Barents Sea (Stern and Laidre, 2016). Polar bear subpopulations have different strategies to cope with seasonal changes in declining sea ice, and some exhibit bimodal strategies. When sea ice retreats, 'coastal' bears stay on land and feed on land-based foods, if any are present, whereas 'offshore' bears undergo long migrations to follow or reach the retreating marginal ice zone, which can be located far from coastal areas (Mauritzen et al., 2001; Rode et al., 2015). Offshore bears from the Barents Sea follow the ice towards the northeastern Barents Sea, whereas coastal bears stay close to the island of Svalbard year-round (Mauritzen et al., 2001; Tartu et al., 2018). These strategies have consequences for contaminant exposures in polar bears. A study on Barents Sea polar bears sampled in the 1990s first showed that PCB concentrations were higher in offshore polar bear females compared to coastal ones (Olsen et al., 2003). The authors suggested that the higher contaminant intake of the offshore bears was related to their larger home ranges, higher energetic requirements, and thus greater food intake. Later analysis (2000–2014) indicated that offshore bears had similar concentrations of lipophilic POPs to the coastal bears, but that they were fatter than the coastal bears (Figure 2.3.4a; Tartu et al., 2018). As lipophilic POPs are more concentrated in thin bears (Tartu et al., 2017b), any effect of habitat use was likely masked by body condition differences. PFAS concentrations were, however, higher in offshore bears (Figure 2.3.4a; Tartu et al., 2018). Concentrations of both lipophilic POPs and PFAS increased with stable isotope values of carbon and nitrogen, indicating bears feeding on higher-trophic position and marine-based prey had higher levels of these pollutants. Although average stable isotope levels were similar for coastal and offshore polar bears, offshore bears had a narrower range of isotopic values indicating a diet of exclusively high trophic level marine prey (Tartu et al., 2018). A follow-up study that selected offshore and coastal individuals with similar body condition found that both PFAS and lipophilic POPs were higher in offshore bears (Figure 2.3.4b; Blévin et al., 2020). Compound-specific and bulk stable isotopes ($\delta^{15}N$ and $\delta^{13}C$), home range, field metabolic rates (based on telemetry), and contaminant levels in harp seal prey from different locations cumulatively indicated that higher POP concentrations in offshore bears were related to a combination of factors, including the consumption of greater proportions of high-trophic level and marine-based prey, higher levels of POPs in prey species, larger energy requirements, and distribution in marginal ice zones (Figure 2.3.4b).

Sea ice conditions also vary within the Svalbard archipelago. The west and northwest coasts are nearly ice free most of the year, whereas the amplitude of sea ice extent is large on the east coast of Svalbard (Prop et al., 2015). These variations in ice conditions have also been found to be related to differences in polar bear body condition, diet, and levels of POPs. As lipophilic POPs are more concentrated in thinner bears, concentrations of these compounds were higher in polar bears sampled from areas with poor sea ice conditions that were thinner compared to bears sampled from areas with good sea ice conditions (Bourgeon et al., 2017; Tartu et al., 2017b). This finding was observed despite the fact that the thinner bears fed at a lower trophic level (Tartu et al., 2016; Tartu et al., 2017b). The same pattern was also reported at a seasonal scale. Polar bears sampled after a winter with poor sea ice conditions were thinner and had higher levels of lipophilic POPs than bears sampled after good winter sea ice conditions or after the summer season (Tartu et al., 2017b). PFAS concentrations, on the other hand, were mainly related to diet and fasting status (Tartu et al., 2017a). Concentrations of circulating PFAS increased with higher intake of upper trophic marine diet items. PFAS concentrations were also higher in fasting vs. non-fasting individuals (Tartu et al., 2017a).

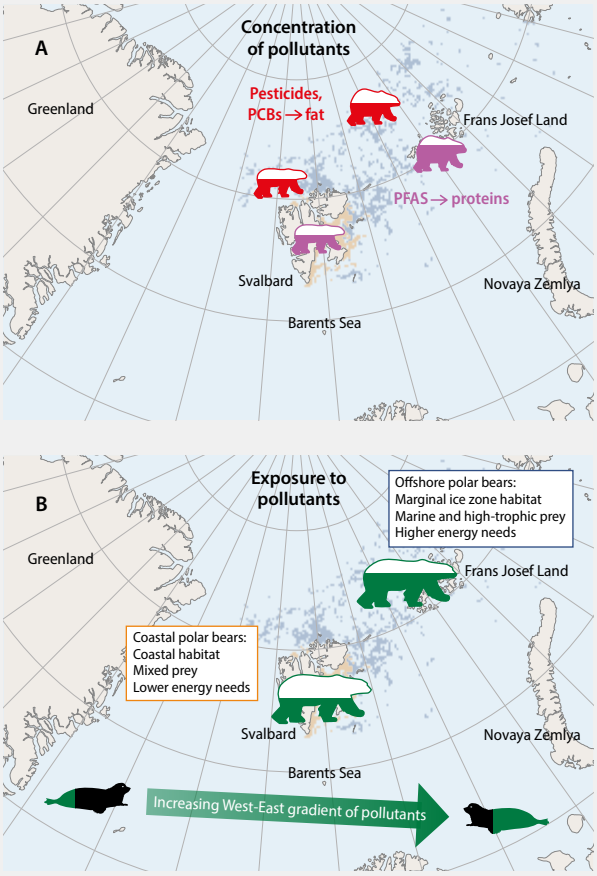

Figure 2.3.4 Combination of factors influencing differences in contaminant exposure and tissue concentrations of coastal and offshore polar bears from the Barents Sea. (a) Concentrations of lipid-soluble POPs are similar in fatter, offshore polar bears than in thinner, coastal bears from the Barents Sea, whereas PFAS concentrations are more elevated in the offshore bears (Tartu et al., 2018). (b) When comparing offshore and coastal bears of similar condition, however, offshore polar bears are exposed to higher levels of POPs than coastal polar bears due to differences in feeding habits, energy expenditure, and geographical distribution (Blévin et al., 2020). Tracks of offshore polar bears shown in blue and tracks of coastal polar bears shown in orange.

processes. Studies on ringed seals from several areas of the Canadian Arctic have reported significant relationships between concentrations of PCBs and/or chlorinated pesticides and sea ice coverage (Houde et al., 2019). The results mostly suggest that contaminants accumulate in ringed seals to a higher extent in years with greater total sea ice coverage and/or years with more multi-year sea ice (MYI). This may be related to the ability of sea ice to facilitate the delivery of organic contaminants to Arctic marine food webs (Pućko et al., 2015), as sea ice, along with snowpack and glaciers, are reservoirs for organic contaminants. In addition, re-mobilization of contaminants from sea ice and water reservoirs is increasing as a consequence of increasing temperatures and decline of sea ice (Ma et al., 2011). However, changes in sea ice phenology also affect the prey composition of ringed seals (Houde et al., 2019). Moreover, in contrast to the positive correlations between contaminants and total and MYI sea ice coverage, first-year sea ice (FYI) coverage was negatively correlated with POP levels in ringed seal blubber, therefore, the authors suggested that accumulation of contaminants in the cryosphere may also depend on the age of sea ice (Houde et al., 2019). Higher levels of POPs have also been associated with shorter sea ice seasons in ringed seals from the Beaufort Sea and Greenland, possibly related to variations in feeding opportunities (Gaden et al., 2012; Rigét et al., 2013a). Although changes in prey composition were not evaluated in these studies, in both cases, the authors suggested POP changes could be related to prey type.

Studies from the Barents Sea have reported higher levels of lipophilic POPs and per- and polyfluoroalkyl substances (PFAS) in polar bears (*Ursus maritimus*) that use high-quality sea ice habitats (i.e. the marginal ice zone) in eastern Svalbard/Barents Sea, compared to bears using habitats with less or no sea ice in western Svalbard/Barents Sea (Routti et al., 2017; Tartu et al., 2017a, 2018; Blévin et al., 2020). These differences may be influenced by the presence/absence of sea ice (see Chapter 2.2), in addition to other biotic factors, which are discussed in Section 2.3.3. The presence of sea ice likely increases the uptake of atmospherically-deposited POPs into the marine food web (Tartu et al., 2017a; Blévin et al., 2020). For example, PFAS deposited on surface snow are released and concentrated into the meltwater at the base of the snowpack by the end of the melting period (Codling et al., 2014), which is followed by phytoplankton blooms, and subsequently large increases of zooplankton biomass (Søreide et al., 2010). In contrast, PFAS deposited on open water surfaces are diluted by sea water and found at lower concentrations than in meltwater (Kwok et al., 2013). Concentrations of lipophilic POPs in Barents Sea polar bears were also related to sea ice extent variations on a seasonal scale (Tartu et al., 2017b). Plasma and adipose tissue concentrations of POPs were higher during seasons and in areas where sea ice conditions were poor. These relationships were, however, mainly related to changes in body fat, and are further discussed in Section 2.3.3.3 and the case study in Box 2.3.1.

For Arctic foxes (*Vulpes lagopus*) on Svalbard, concentrations of β-HCH and several PFAS increased with increasing sea ice availability (Andersen et al. 2015a; Routti et al., 2017). However, these relationships were related to an increased availability of seals as further discussed in Section 2.3.3.2.

2.3.2.4 Terrestrial runoff to surface waters

In many regions of the Arctic, climate change is expected to increase the amount of precipitation received in the form of rain. This change will increase the runoff of terrestrial-derived organic material from land to surface waters, leading to the browning of Arctic and sub-Arctic lakes and rivers, and darkening of coastal waters (Macdonald et al., 2005a; Dupont and Aksnes, 2013; Wauthy et al., 2018). Terrestrial runoff can carry organic contaminants (Ripszam et al., 2015) and alter the bioavailability of contaminants by increasing loads of particulate organic matter, and thus influence contaminant exposure in freshwater biota. In addition, elevated terrestrial inputs may alter coastal food web dynamics by presenting zooplankton with lower-quality organic matter (Andersson et al., 2018). If fed upon by zooplankton, the terrestrial energy input introduces an additional trophic level and trophic transfer into the food web (Jonsson et al., 2017). Darkened coastal waters due to terrestrial input might further alter predator-prey relationships by decreasing the hunting efficiency of optical predators such as fish, in favor of tactile predators, such as jellyfish (Aksnes et al., 2009). Such changes in trophic interactions will ultimately alter the transfer of energy and contaminants. Thus, increased terrestrial runoff will likely increase POP exposures and alter trophic transfer in the food web (Figure 2.3.2).

In polar regions, runoff from terrestrial systems includes meltwater from snow and glaciers, which can result in seasonal pulses of freshwater with contaminants to terrestrial, freshwater, and coastal ecosystems. Snow melt releases recently atmospherically-deposited contaminants (Wania et al., 1998), as found for PAHs in Antarctica (Casal et al., 2018b, 2019), whereas glacial melt transfers stored contaminants to the terrestrial system, coastal waters, and lake sediments (Hermansson et al., 2010; Garmash et al., 2013; Lehnherr et al., 2018). However, in the Adventfjorden on Svalbard, which receives riverine input containing runoff from both a large tundra valley and glacier, benthic marine amphipods (*Gammarus setosus*) showed decreasing total PCB concentrations seasonally from April to August (Skogsberg, 2019). The decrease in total PCB concentrations in amphipods coincided with an increase in terrestrial organic matter from riverine runoff, which may have both diluted and lowered the bioavailable fraction of contaminants at the base of the food web. However, less hydrophobic PCBs peaked in amphipods in May/June, coinciding with snow melt. May and June dietary data (i.e. fatty acids) suggested pelagic diatoms made up a high proportion of the amphipod diet, and thus may be the link between the PCBs in snow melt and those detected in the the amphipods (Skogsberg, 2019). The net result of terrestrial runoff on contaminant accumulation in Arctic ecosystems is not known to date, and requires further investigation (McGovern et al., 2019). In addition, increased temperatures in polar regions will enhance permafrost thawing, releasing another pool of organic matter (see Section 2.3.2.5 and Box 2.3.2).

Box 2.3.2 Case Study: Effects of permafrost thaw on Arctic char condition

Two lakes, East Lake and West Lake, on Melville Island in the central Canadian Arctic Archipelago have been the subjects of a long-term study examining the effect of permafrost thaw and degradation on lake water (Roberts et al., 2017). Since 2008, West Lake has been increasingly impacted by permafrost degradation and subaqueous slumps, which together increased turbidity 50- to 100-fold compared to the nearby East Lake by 2016 (Figure 2.3.5, left). Over this same time period, the physical condition of the resident Arctic char in both lakes changed, as determined using condition factors (K), a standard measure of fish health calculated using an individual's mass relative to its length. Whereas condition factors increased in char from East Lake, they declined in char from the permafrost thaw-impacted West Lake (Figure 2.3.5, left). With turbidity >100 times higher than in East Lake, visibility in West Lake has been substantially reduced, limiting the ability of visual predators such as Arctic char to feed, which likely explains the lake's declining fish condition. Alternatively, the increased fish condition in East Lake may be due to warmer water temperatures and reduced ice cover, although this requires further investigation. Analysis of carbon stable isotopes ($\delta^{13}C$) and nitrogen stable isotopes ($\delta^{15}N$) showed that mean $\delta^{13}C$ values (± SD) in adult char from East Lake (-27.27±0.81 ‰; n=98) were depleted compared to those of West Lake char (-24.73±1.17 ‰; n=97), indicating greater terrestrial and benthic carbon inputs to West Lake (Muir, unpubl. data). Also, mean $\delta^{15}N$ values (± SD) were lower in West Lake char (10.1±0.98 ‰) compared to those in East Lake char (11.2±0.50 ‰), suggesting differences in food sources. By comparison, fish condition has not changed significantly in three of four Arctic char populations in lakes of similar size on Cornwallis Island in the central Canadian Arctic Archipelago (Figure 2.3.5, right) (Hudelson et al., 2019), despite similar warming temperatures, especially during the period 2008 to 2012.

The physiological condition of Arctic char in Lake Hazen, the largest lake by volume north of the Arctic Circle, was also reported to have declined from 1981 to 2015 based on the overall trend in K values (Lehnherr et al., 2018). This trend also coincided with increased turbidity in the lake, arising from increased discharge of sediment-rich glacier-fed rivers, which was particularly apparent between 2008–2013. However, it should be noted the conclusion of Lehnherr et al. (2018) – that ecological changes have resulted in a significant decline in the condition of Lake Hazen char – has been challenged, with independent statistical analysis of the dataset suggesting no significant trend exists (Moore et al., 2018). Nevertheless, trends in the phytoplankton ecology of Lake Hazen have also been observed based on paleolimnological studies, with the planktonic diatom, *Cyclotella sensu lato*, increasing in relative abundance and supplanting benthic species by the late 1990s, reflecting longer ice-free periods in the lake. Thus, the food web of Lake Hazen is undergoing changes, but further monitoring is needed to establish the effects on Arctic char, the lake's top predator. Concentrations of lipid-normalized ΣPCBs and ΣDDTs in Arctic char in Lake Hazen declined over the period 2001–2015 but were positively correlated with NAO conditions (Cabrerizo et al., 2018b; see Chapter 2.4). However, trends of POPs in Arctic char in Lake Hazen were not related to the increased glacial discharge or turbidity.

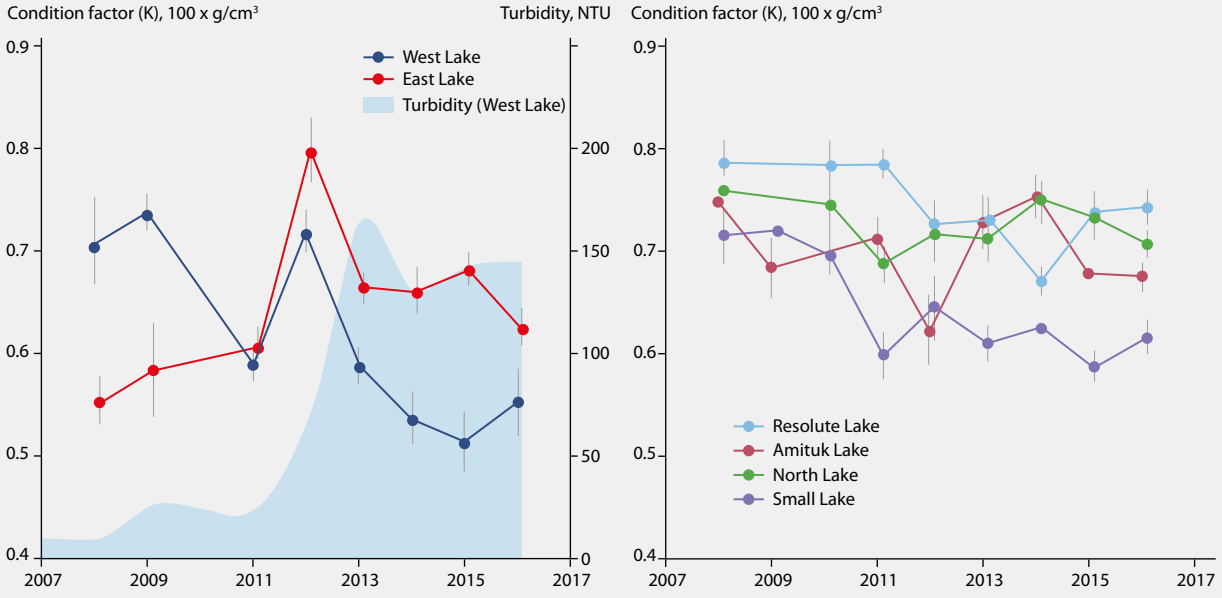

Figure 2.3.5 Condition of land-locked Arctic char in Canadian lakes. (Left) Condition factors K for landlocked Arctic char in the clear East Lake and turbid West Lake (2008–2016). Turbidity in West Lake is shown as a shaded background. Turbidity values in East Lake were <5 NTU and not shown. Data from Roberts et al. (2017). (Right) Condition factors of adult Arctic char from four other lakes on Cornwallis Island in the central Canadian Arctic Archipelago for comparison. Data from Hudelson et al., (2019).

2.3.2.5 Freshwater hydrology and permafrost thaw

As the climate warms, Arctic lakes and surrounding terrestrial environments are undergoing numerous changes that may impact their food webs and fish populations. These changes will ultimately affect POP inputs to lake surface waters and catchments, as well as bioaccumulation and food web biomagnification. For a detailed overview of the impacts of climate change on Arctic terrestrial hydrology and lake ecosystems, readers are referred to reviews of Wrona et al. (2013), Vonk et al. (2015), Prowse et al. (2015) and Bring et al. (2016). Of particular relevance to POP bioaccumulation in Arctic lake food webs are changes in primary productivity, biogeochemical cycles and chemical transport pathways, seasonal phenology, and species compositions, which are driven by declining lake ice cover, thawing permafrost, increased early- and late-season precipitation in the form of rain, and warming average annual temperatures and summer air temperatures (Wrona et al., 2013). These changes are briefly discussed below in the context of impacts on freshwater food webs, and ultimately on bioaccumulation of POPs. Note that such changes and processes are likely highly relevant for coastal marine waters as well.

A general decline in the duration of ice cover has been observed in Arctic lakes greater than one squared kilometer in area. Šmejkalová et al. (2016) surveyed satellite imagery for 13,300 lakes across the circumpolar Arctic from 2000–2013 and found significant trends towards earlier ice break-up, ranging from -0.6 days/y in northern Alaska, to -0.1 days/y in northern Scandinavia. Lakes of this size generally have fish populations with salmonids, and especially Arctic char, predominating in regions above the treeline (Rigét et al., 2000a; Hershey et al., 1999, 2006). These changes are likely to affect water temperatures and within-lake biogeochemical cycles, leading to increases in primary productivity and consequently, changes in lake trophic relationships. Shifts in the diatom community structures of High Arctic lakes have been rationalized primarily by changing climatic factors such as shorter durations of ice coverage, longer growing seasons, and more favorable thermal stratification patterns (ACIA, 2005).

Climate warming is also thawing permafrost in lake catchments and shorelines resulting in changes to lake and river water chemistry. Permafrost thawing results in inputs of dissolved organic carbon (DOC), inorganic solutes including nutrients such as ammonium, nitrate, and phosphate and major ions such as Ca^{2+}, Mg^{2+} and SO_4^{2-}, and suspended solids to streams and lakes. Nonetheless, limnological properties are highly influenced by the regional surroundings (Vonk et al., 2015). Paleolimnological and water chemistry studies of retrogressive thaw slump (RTS; Burn and Lewkowicz, 1990) affected lakes located east of the Mackenzie River delta area of the Canadian Arctic have shown increases in the abundance and diversity of periphytic diatoms consistent with increased water clarity and subsequent development of aquatic macrophyte communities (Thienpoint et al., 2013). There is broad agreement from several studies of lakes in this region that higher concentrations of Ca^{2+}, Mg^{2+}, SO_4^{2-} and other ions, lower DOC, and increased water clarity occur in RTS-impacted lakes compared to unimpacted lakes (Kokelj et al., 2005; Thompson et al., 2012; Houben et al., 2016). The decline in DOC was attributed to the sedimentation of organics and particulates from the water column (Houben et al., 2016). In related work, Eickmeyer et al. (2016) found that reduced DOC in slump-affected lake water resulted in higher concentrations of POPs in sedimentary organic matter. Although the impacts of these changes on the bioaccumulation of POPs in these lake food webs have not been investigated in detail, D'Onofrio (2014) found amphipods (*Gammarus* sp.) from slump-affected lakes had higher mean ΣPCBs and ΣDDTs (27.5 ng/g lw and 18.5 ng/g lw, respectively) than those from unaffected lakes (17.0 ng/g lw and 10.9 ng/g lw, respectively). Additionally, amphipod POP concentrations increased with the percentage of the catchment slumped (D'Onofrio, 2014). Mean ΣHCHs also differed between amphipods from slump-affected lakes (7.0 ng/g lw) and unaffected lakes (5.1 ng/g lw), although less so than for the more hydrophobic POPs. These water chemistry changes resulting from permafrost thawing and run-off might also affect the physiological condition of freshwater fish (Box 2.3.2).

Other studies in northern Alaska (USA), northern Quebec (Canada), Siberia (Russia) and northern Sweden focused on tundra streams have shown elevated suspended sediments and DOC downstream of thermokarst activity (Abbott et al., 2015; Vonk et al., 2015).

2.3.2.6 Seasonality

The large seasonal variation of solar irradiance in the Arctic not only guides day length and temperature, but also wind and precipitation patterns. Seasonality therefore has strong effects on both physical and biological processes during the year. Although climate change does not affect irradiance, the annual light cycle, and day length *per se*, the increase in temperature due to climate change will lead to alterations in the seasonality of physical drivers, such as terrestrial snow melt and runoff, and sea ice thickness, distribution and break-up, which in turn, will affect the timing of ecosystem responses, such as phytoplankton blooms and availability of breeding grounds. Thus, the window of the productive season, to which Arctic species have adapted their life histories and trade-offs, will be altered, with implications for the growth, reproduction, energy allocation, and migration of biota (Varpe, 2017).

Climate change might affect exposure, accumulation and effects of POPs on wildlife through shifts in the seasonal timing of their biological responses (e.g. migration, availability of prey, reproduction). In seabirds, POP levels measured in parents and their offspring at summer Arctic breeding grounds reflected higher exposures received from the industrialized and agricultural winter grounds they migrated from (Baert et al., 2013; Bourgeon et al., 2013; Leat et al., 2013). Changes in migration timing and the relative time spent at winter and breeding grounds could therefore affect POP exposure. Although links to POPs have not been examined, climate-associated changes in the timing of Arctic cetacean migrations have also been documented (Hauser et al., 2017; Druckenmiller et al., 2018). Due to climate change, the Arctic will experience increasingly earlier springs, with an earlier availability of breeding grounds and food (Post et al., 2009). If migrating birds do not respond to this earlier onset of spring, they may experience an increasing mismatch between the timing of resource availability and their arrival at breeding grounds, eventually resulting in poorer condition and reduced fitness (Saino et al., 2011). Birds may respond to this shift in

Box 2.3.3 Case Study: Climate change, seasonality and contaminant transfer in the pelagic food web of an Arctic fjord

Unravelling the effects of climate-induced changes in contaminant transfer is complicated by the strong seasonality of Arctic ecosystems. However, there is still a need for empirical knowledge on the effect of climate change on contaminant disposition in ecosystems (Nizzetto et al., 2010; Ruus et al., 2010). On Svalbard, an effort to elucidate the effects of climate change on contaminant bioaccumulation and biomagnification in a pelagic food web was conducted between 2007–2008 using a 'space for time' approach where two or more contrasting sites embodying characteristics representative of present-day and expected future conditions are used to model potential changes over time. Contaminant transfer in the pelagic food web of two fjord systems, Kongsfjorden (Atlantic fjord, representative of future Arctic climate conditions) and Liefdefjorden (Arctic fjord, representative of present-day Arctic climate conditions), were studied with regards to seasonal changes in POP food web bioaccumulation (only Kongsfjorden) and differences between POP bioaccumulation in the food webs of the two fjord systems (Figure 2.3.6) (Hallanger et al., 2011a, b, c).

At Kongsfjorden, seasonal differences in POP levels were observed in zooplankton (decrease from May to October), various fishes (different trends depending on species), little auks (*Alle alle*; similar to trends observed in zooplankton), and kittiwakes (*Rissa tridactyla*; different trends depending on contaminant) (Hallanger et al., 2011a, c). Given the diversity of seasonal changes in POPs experienced by different trophic levels and species of the fjord food web, knowledge of seasonal variation will be essential for differentiating climate change-induced alterations from seasonal changes in POP bioaccumulation and biomagnification.

Differences between the Atlantic- and Arctic-influenced fjords were observed both with regards to POP transfer and fjord-specific characteristics. Higher POP concentrations and bioaccumulation factors (BAFs) were generally observed in zooplankton species from Kongsfjorden. However, there were compound-specific differences, with Liefdefjorden having higher concentrations and proportions of the more volatile compounds, such as α-HCH and chlordanes (Hallanger et al., 2011b). These differences were attributed to different water masses, ice cover, and freshwater input: all factors which have clear climate relevance but may represent confounding factors associated with the fjords. The seasonal input of freshwater due to the melting of glaciers and snow was earlier in southern Kongsfjorden than northern Liefdefjorden. Further, Kongsfjorden did not have sea ice during winter, while the sea ice in Liefdefjorden broke up shortly before sampling. These differences made the two fjords out of phase with each other with regard to release of volatile POPs from sea to air. Delayed volatilization of compounds in Liefdefjorden due to the later melting of snow, glacial ice and sea ice resulted in higher concentrations/exposure in seawater in Liefdefjorden compared to Kongsfjorden for these compounds (Hargrave et al., 1997, Lohmann et al., 2009; Hallanger et al., 2011b).

Therefore, the differences observed in zooplankton and the food webs of the two fjords could be due to seasonal variation and not climate change (Hallanger et al., 2011b). Overall, there seemed to be a seasonal lag in the peak concentrations detected in biota beginning with an early season peak in zooplankton and progressing to mid- and late- season peaks in fish and birds. Environmental changes in contaminant exposure appear to be reflected faster in zooplankton than in longer-lived, higher trophic-level organisms (Borgå et al., 2005). Thus, extrapolating the results of this study to approximate future climate change effects should be done with caution. Space-for-time approaches require two contrasting sites that are comparable with regards to other potential confounding factors within- or between- sites. This was not achieved within these two fjord systems. Additional investigations with sampling taking place over several years, and/or using Arctic and Atlantic locations outside the fjords that are not influenced by sea ice and meltwater, could be more suitable for elucidating climate change influenced alterations in bioaccumulation and biomagnification.

Characteristics	Kongsfjorden	Liefdefjorden
Representative climatic conditions	Future High-Arctic	Current High-Arctic
Sea ice cover	No	Yes, broke just before sampling
POP concentrations in zooplankton	Higher	Lower
Bioaccumulation factors	Higher	Lower
Volatile compound concentrations and relative proportions	Lower	Higher

Figure 2.3.6 Comparison of environmental conditions, contaminant exposure and bioaccumulation in zooplankton from the pelagic food web of two fjords on Svalbard: Kongsfjorden (Atlantic fjord with conditions representative of future Arctic climate) and Liefdefjorden (Arctic fjord with conditions representative of present-day Arctic climate). Map from the Norwegian Polar Institute (toposvalbard.npolar.no).

phenology by migrating north earlier, which could also affect their migration route, timing, and energy used for reproduction at breeding sites (i.e. income vs. capital breeders), and thus, the POPs accumulated and transferred to offspring. Animals that rely mostly on stored reserves for breeding purposes (e.g. egg formation) are considered "capital breeders", and species relying mostly on concurrent intake for reproduction are "income breeders"; there is a continuum across species in using stored nutrients for breeding (Jaatinen et al. 2016). In the terrestrial barnacle goose (*Branta leucopsis*), PCB levels were slightly increased in later-hatching eggs, suggesting females arriving later to Arctic breeding grounds spend more time consuming vegetation at more-contaminated overwintering grounds, compared to earlier-arriving females that predominantly rely on less-contaminated vegetation at the breeding grounds to accumulate the energy reserves for reproduction and egg laying (Hitchcock et al., 2019a). Based on this example, animals arriving at breeding grounds earlier and relying on the resources consumed there for reproduction, might be expected to have offspring with lower contaminant concentrations, as a result of income breeding adaptations and feeding on less-polluted prey.

In the marine ecosystem, the seasonal melting of marginal ice zones influences the timing of algal blooms. The earlier reduction in sea ice extent and volume can lead to a mismatch in the timing of energy availability for secondary producers such as copepods, a key species in the Arctic marine food web (Søreide et al., 2010). This may in turn alter exposures to POPs in these consumers and their predators. Seasonal changes in a pelagic food web can offer insight into climate change impacts in this regard (Box 2.3.3, previous page).

2.3.3 How do ecological changes affect POP exposures in Arctic biota and food webs?

2.3.3.1 Primary production

Climate change is expected to affect both the seasonal timing (phenology) and the amount of terrestrial and aquatic primary production. The abundance of primary production will still vary seasonally, however, the overall biomass is predicted to change (IPCC, 2013). This has implications for the bioavailability of POPs entering the base of the food web, as increased primary production leads to more particulate matter and particle-bound POPs, and conversely, less freely-dissolved POPs bioavailable for direct uptake from water (Borgå et al., 2010). Although dietary uptake of POPs occurs via the particle phase (i.e. algae surface), the average POP concentration in the algae (i.e. in particulate matter) should be lower due to dilution in a larger overall pool of biomass. How, and if, these effects will counteract each other is unknown. In addition, the proportion of POPs subjected to sedimentation into the benthic systems could increase, which all else being equal, would result in higher POP exposure in benthic ecosystems through benthopelagic coupling (Van den Brink et al., 2011; Wagner et al., 2019).

In consumers at lower trophic levels of marine and freshwater food webs, POP exposure occurs both through direct uptake from water and through dietary exposure (Hargrave et al., 2000; Fisk et al., 2003; Nizzetto et al., 2010; Hallanger et al., 2011a). Concentrations of POPs in Arctic marine zooplankton from Lancaster Sound in the Canadian Arctic decreased in the open water period, being lowest in August-September (Hargrave et al., 2000). This was due in part to reduced water and algae organochlorine contaminant concentrations, but may also have been related to increasing zooplankton lipid content during this time. Similar trends were observed in Arctic zooplankton from Kongsfjorden, Svalbard, with overall POP levels decreasing from May to October (Box 2.3.3; Hallanger et al., 2011a). In the western Canadian Arctic, burbot (*Lota lota*) from the McKenzie River showed increasing PCB concentrations from 1985–2009, despite decreasing or stable atmospheric concentrations over the same time period (Carrie et al., 2010). The authors showed evidence that the rising PCB concentrations in these freshwater fish were likely related to increases in primary production and 'algal contaminant scavenging' into the food web (Carrie et al., 2010).

2.3.3.2 Species interactions

Climate is a major factor constraining the distribution of species on Earth, and consequently, climate change is shifting the composition of the planet's ecological communities. Species are undergoing global redistribution towards cooler regions, including the poles, higher altitudes, and greater aquatic depths (Pecl et al., 2017). Movements into these areas are being undertaken to maintain thermal tolerances and in response to other species' movements, such as predators following range-shifting prey (Chen et al., 2011; Poloczanska et al., 2013). The intensity of these climate-driven species invasions, as well as local extinctions, are predicted to be greatest in the Arctic and Antarctic (Cheung et al., 2009). Thus, the formation of 'no-analog' communities, those with compositions not found under current conditions (Williams et al., 2007; Urban et al., 2012), may be especially pronounced in polar regions and possibly other cold regions such as the Tibetan Plateau.

Increased temperature and ocean current velocities due to climate change are leading to changes in the northern boundary of various species, with Arctic species generally following the receding marginal ice zone northward and boreal species increasing their presence and abundance in the Arctic, a phenomenon referred to as 'borealization' (Fossheim et al., 2015; Oziel et al., 2020). Borealization will lead to changes in the species composition of Arctic food webs, and thus changes in trophic interactions, and ultimately alterations in biomagnification. This concept is referred to as 'Atlantification' in the marine environment of the European Arctic and 'Pacification' in the western North American Arctic. Altered community composition gives rise to novel interactions among species, such as new competitive and predatory interactions. Disturbed predator-prey interactions, in particular, are expected to have outsized consequences for ecological communities through trophic cascades (Zarnetske et al., 2012). These changing vertical interactions can be anticipated to impact POP burdens in biota, since most exposures in consumers occur largely via the diet (Borgå et al., 2004; Hallanger et al., 2011a; McKinney et al., 2011). Moreover, given that the greatest rates of species invasions, at least for marine ecosystems, are projected to occur in polar regions (Cheung et al., 2009), and rapid Arctic borealization has already

been documented (e.g. in the Barents Sea; Fossheim et al., 2015), we can speculate that polar marine wildlife may be more at risk from climate-driven ecological changes and POP exposures than other regions (Borgå, 2019).

With the current lack of understanding of how climate affects food web structure and the strength of interspecific interaction for most species, the ability to forecast shifting biotic interactions under climate change remains limited (Urban et al., 2017). Hence, models exploring the future impacts of shifting food webs on POPs are non existent. However, in the past decade, several studies have empirically examined the role of changing trophic relationships on tissue POP concentrations in Arctic species and food webs. The current state of knowledge is based on research from Canadian, Alaskan (USA), Greenlandic (Denmark), and Norwegian Arctic ecosystems, however, data from the Russian Arctic are lacking. Thus far, most published research on the impacts of shifting species interactions on POPs has focused on marine environments, with marine mammal and seabird studies predominating, but a few studies on marine fishes and a terrestrial mammal (Arctic foxes) have also been performed; findings from these studies are summarized below.

In three regions of varying latitudes within the eastern Canadian Arctic, concentrations of PCBs, organochlorine pesticides (OCPs), and various flame retardants were compared between marine prey fish communities, including native and redistributing non-native species (Pedro et al., 2017a). The comparisons particularly focused on the keystone species, Arctic cod (*Boreogadus saida*), and the sub-Arctic 'replacement' species, capelin (*Mallotus villosus*) and sand lance (*Ammodytes* sp.). Although not all species were collected in all regions, native benthic species, specifically sculpin (*Cottoidea* sp.) and northern shrimp (*Pandalus borealis*), showed the highest concentrations of ΣPCBs and ΣOCPs of all species. Nonetheless, capelin, and to a lesser extent, sand lance, showed higher concentrations of many POPs than Arctic cod (Figure 2.3.7). Furthermore, neither feeding habitat (based on $\delta^{13}C$ values), trophic position (based on $\delta^{15}N$ values), or fish length accounted for this difference. This finding, along with particular congener/compound pattern differences among the species (e.g. higher proportion of DDT vs DDE in capelin relative to Arctic cod), suggested that the higher POP concentrations in invading sub-Arctic capelin may, at least in part, be a consequence of them acquiring temperate contaminant signatures during seasonal migrations. Thus, these species may act as climate-driven contaminant biovectors into Arctic marine food webs.

Similarly, both transient marine fish and mammals showed higher POP concentrations than similar trophic-position Arctic-resident species in a study of the food web of Cumberland Sound, Canada (McKinney et al., 2012). Elevated concentrations of several current-use pesticides (chlorothalonil, chlorpyrifos, dacthal, and endosulfan) were also found in the capelin of Cumberland Sound relative to other species at a similar trophic position (Morris et al., 2016). Nonetheless, the differences in POP concentrations between these native and non-native replacement fishes were small (two-fold or less), suggesting changes in these forage fish communities may have a limited impact on POP concentrations in piscivorous predators (i.e. predator fish, marine mammals, and seabirds). Direct impacts of these prey fish changes on predator POP concentrations have been investigated for a few seabird species (as discussed further below), but not for any piscivorous fish or marine mammals to date.

Killer whales (*Orcinus orca*), perhaps the most contaminated species on Earth with respect to POPs (Ross et al., 2000), are undergoing a northward range shift into Arctic and sub-Arctic waters in multiple circumpolar locations (Higdon et al., 2012, 2014; Clarke et al., 2013; Rosing-Asvid, pers comm.). Within the eastern Canadian Arctic and around southeast Greenland, they appear to be targeting marine mammals for food, which may differ from their presumably more typical diet of fish in the North Atlantic (Ferguson et al., 2012; Higdon et al., 2012; Reinhart et al., 2013; Bourque et al., 2018). However, recent studies have suggested that some North Atlantic killer whale populations may also eat prey other than fish. Killer whales sampled between 2012–2014 around southeast Greenland showed mean blubber concentrations of PCBs (40 mg/kg lw)

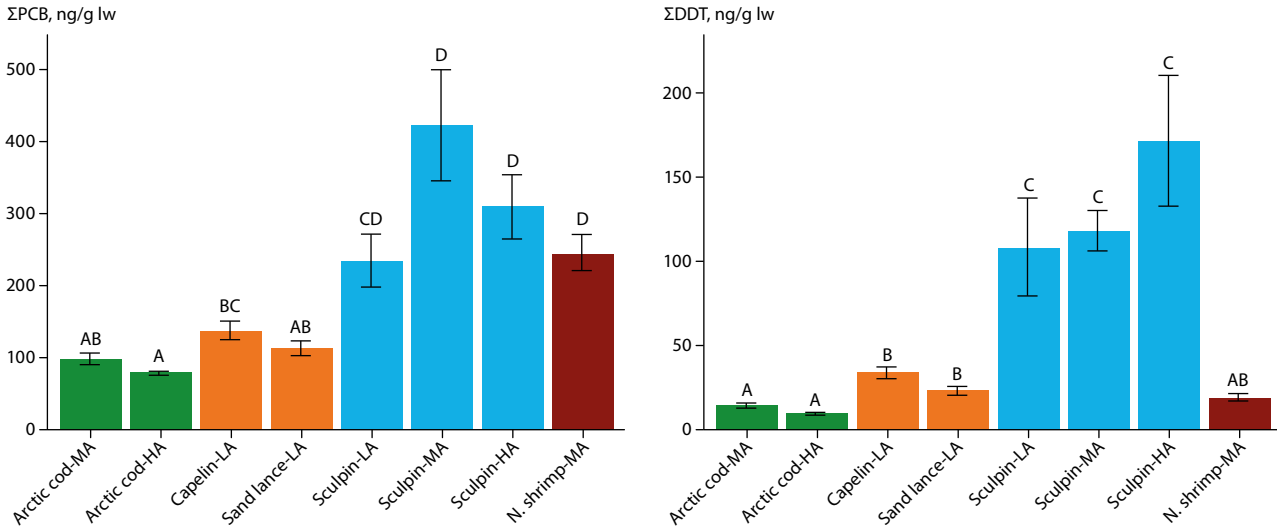

Figure 2.3.7 Mean (+/- SE) lipid weight concentrations of PCBs (left) and DDTs (right) in muscle of marine forage fish and invertebrates of the low- (LA), mid- (MA), and high- (HA) Canadian Arctic from 2012–2014. Arctic cod (green), the two sub-Arctic species, capelin and sand lance (orange), sculpin (blue), and northern shrimp (red) are shown. Significant differences in contaminant concentrations among species are indicated by different letters above each bar. Note that the y-axes scales differ. Adapted from Pedro et al. (2017a).

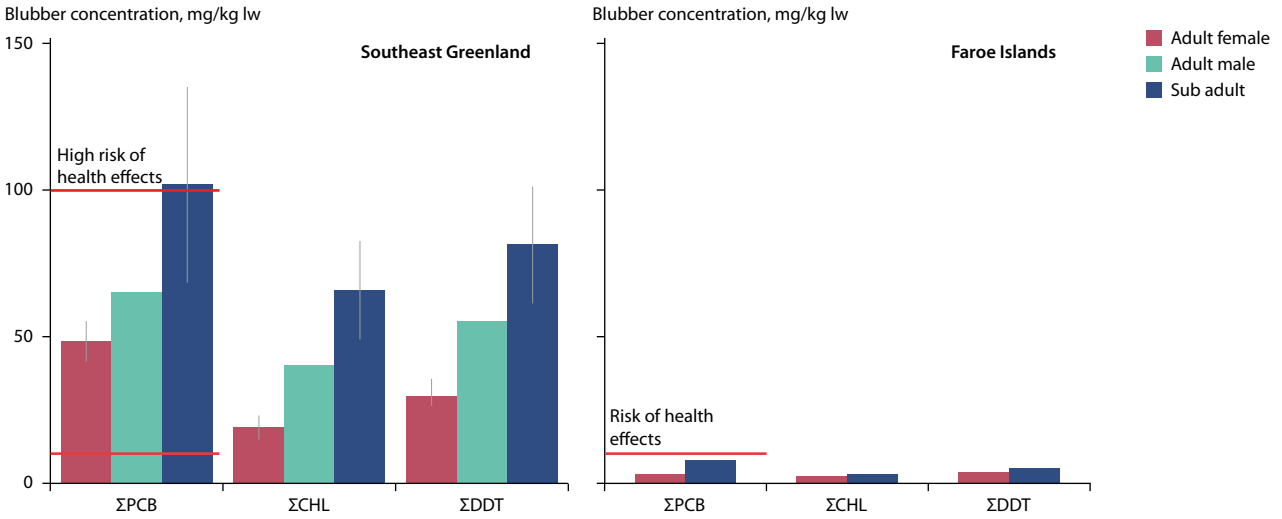

Figure 2.3.8 Mean concentrations (±SE) of ΣPCBs, ΣCHLs, and ΣDDTs (mg/kg lw) in blubber of eighteen killer whales sampled in 2012-2014 in southeast Greenland (left) and two killer whales sampled in 2008 in the Faroe Islands (right). Red horizontal lines indicate thresholds for risk (10 µg/g lw), and high risk (100 µg/g lw) of health effects associated with PCB exposure as reported by Dietz et al. (2019). Adapted from Pedro et al. (2017b).

and OCPs (70 mg/kg lw) (Pedro et al., 2017b) that were an order of magnitude higher than those found in killer whales sampled over similar years in the Faroe Islands. This substantial difference in contaminant burdens was suggested to be due, at least in part, to North Atlantic whales increasingly feeding on marine mammals in new Arctic habitats (Figure 2.3.8).

Recent studies using stable isotopes as dietary markers in a killer whale population off the Norwegian coast also indicate that some whales may feed on marine mammals (Jourdain et al., 2020) resulting in elevated contaminant levels (Andvik et al., 2020). Analysis of PCBs indicates 100% of the seal-eating killer whales were above the threshold for risk of health effects, compared to 43% of fish-eating killer whales (Figure 2.3.9; AMAP, 2018b).

With climate change has come a shift in the dietary composition of seabirds in the Canadian Arctic (Braune et al., 2014) and East Greenland (Amélineau et al., 2019), that in turn, has likely contributed to changes in their exposure and accumulation of POPs. In the Canadian Arctic, and particularly the Hudson Bay-Hudson Strait region, the diet of thick-billed murres has

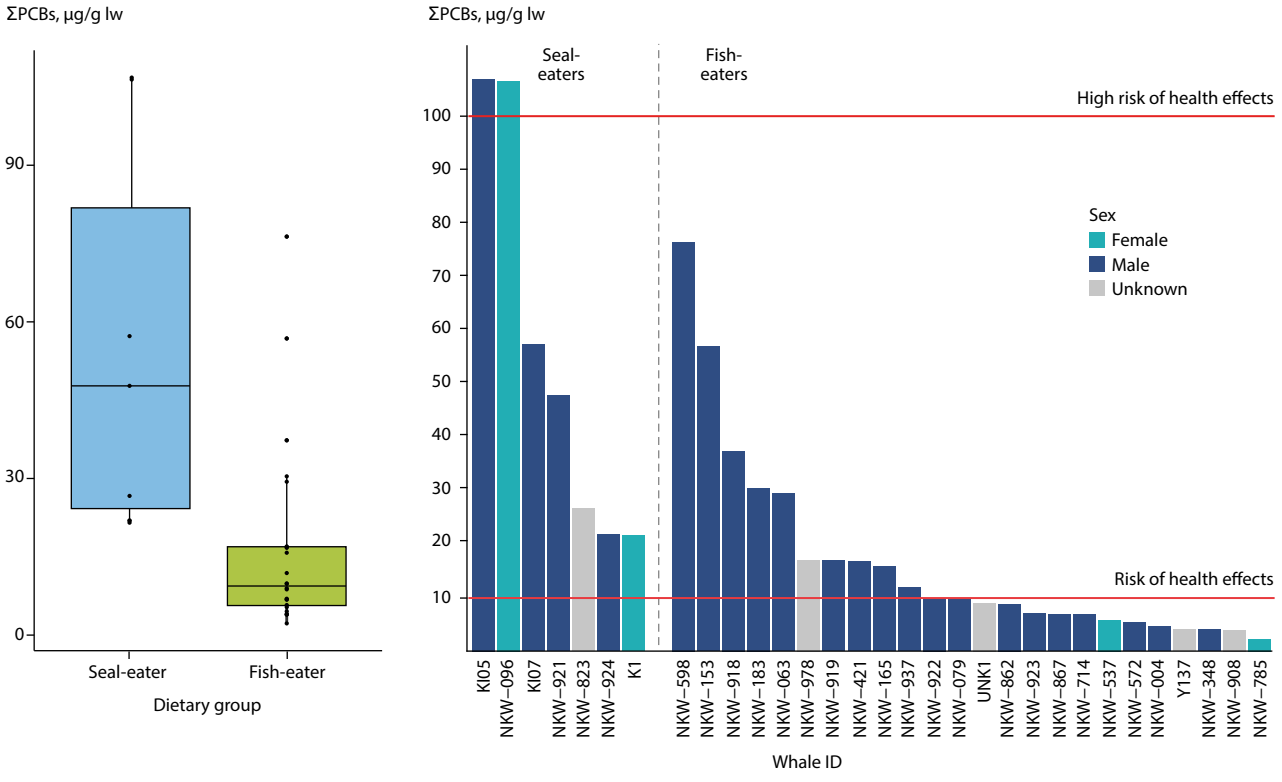

Figure 2.3.9 ΣPCB levels in blubber of seal-eating (n=7) and fish-eating (n=24) killer whales from Norway (Andvik et al., 2020). (Left) Box plot of ΣPCB levels in killer whales by dietary group. Horizontal lines represent the median concentration, whiskers represent the lower (first) and upper (fourth) quartiles, and dots represent individual whales, with points outside the box and whiskers being outliers. (Right) ΣPCB levels of individual whales shown in decreasing order by dietary group. Red horizontal lines indicate thresholds for risk (10 µg/g lw) and high risk (100 µg/g lw) of health effects as reported by Dietz et al. (2019).

been shifting since the 1970s and 1980s due to changing ice conditions. The diets of these low-Arctic breeding murres have changed to contain less ice-associated, cold-water Arctic cod and more of the sub-Arctic species, capelin and sand lance (Gaston et al., 1993, 2003, 2012). Conversely, high-Arctic breeding murres have continued to consume predominantly Arctic cod (Mallory et al., 2010, Provencher et al., 2012). These dietary patterns may explain inter- and intra-species differences in POPs of Canadian Arctic thick-billed murres and northern fulmars measured in 2008 (Braune et al., 2014). Similarly, temporal increases in hepatic organochlorine concentrations in murres from the High Arctic measured in 1998, 2003 and 2008, may be partially related to concurrent changes in diet and trophic position (Braune et al., 2014). In East Greenland between 2004 and 2015, the diet of little auks (*Alle alle*) contained less lipid-rich, ice-associated prey when sea ice coverage decreased, with adult birds consuming zooplankton at a higher trophic position, as reflected by an increase in blood $\delta^{15}N$ values (Amélineau et al., 2019). Concurrently, feather mercury (Hg) concentrations of these auks increased by 3.4% each year, notably occurring during the summer and not when overwintering (Fort et al., 2016; Amélineau et al., 2019). Surprisingly, additional research suggests that interannual variation in little auk feather Hg concentrations reflects changes in food contaminant levels, notably increased Hg levels in zooplankton during summertime, and not the reorganization of the food web or modification of seabird trophic ecology, based on $\delta^{15}N$ and $\delta^{13}C$ values in little auk whole blood (Fort et al., 2016). Similar patterns may occur for lipid-soluble POPs. For further studies of Hg, we refer the reader to the recent AMAP report on Hg in the Arctic in relation to climate change (AMAP, 2021).

Climate change is altering the structure of some Arctic food webs, and in turn, influencing trophic positions of predatory seabirds and potentially influencing long-term trends in environmental contaminants. After adjusting for the reduced trophic position of thick-billed murres in Hudson Bay, Canada (based on egg $\delta^{15}N$ values), declining trends of organochlorine concentrations (HCB, heptachlor epoxide, oxychlordane, dieldrin, *p,p′*-DDE and ΣPCB_{69}) measured in eggs between 1993–2003 were more moderate than originally thought (Braune et al., 2015a). In contrast, adjusting for the increased trophic position of High Arctic breeding murres resulted in increased rates of organochlorine concentration declines, although *p,p′*-DDE and ΣPCB_{69} remained relatively unchanged (Braune et al., 2015a). A direct link between climate change and contaminant trends was not investigated in this study.

Also in the Canadian sub-Arctic region of western Hudson Bay, dietary and/or food web shifts were linked to changes in POP trends in polar bears. The diet change was associated with faster increases in adipose concentrations of total polybrominated diphenyl ethers (ΣPBDEs) over time, and a switch from slowly decreasing, to slowly increasing concentrations of ΣPCBs from 1991–2007 (McKinney et al., 2009). Based on the decline of $\delta^{13}C$ values and particular shift in fatty acid profiles observed, the authors speculated that the change in contaminant trends may have been related to changes in the proportion of bearded seal (*Erignathus barbatus*), sub-Arctic harbor seal (*Phoca vitulina*) and harp seal (*Pagophilus groenlandicus*) in the bears' diet, although other food web changes could also have contributed. A similar change was observed in East Greenland polar bears between 1984–2011; a shift towards feeding on sub-Arctic hooded seal (*Cystophora cristata*) and/or harp seal was directly estimated based on quantitative fatty acid signature analysis, and was associated with NAO conditions (McKinney et al., 2013). However, at least over the time period studied, the shift did not significantly alter trends in adipose concentrations of any of the POPs examined (PCBs, OCPs, and PBDEs) (Figure 2.3.10; McKinney et al., 2013). A re-evaluation of this relationship including recent years through 2016 in East Greenland polar bears suggested that hooded and/or harp seals now represent the majority of the polar bears' diet, and that a concurrent rise in concentrations of certain POPs, including PCBs, occurred at least until 2013 (Dietz et al., 2021). The effect of this ecological change on POP concentrations may be due to harp seals, and particularly, hooded seals occupying a higher trophic position than the bears' historical main prey, ringed seals, and/or due to these migratory sub-Arctic seals acting as biovectors transporting contaminants from further south into Arctic marine food webs (McKinney et al., 2012).

In the southern Beaufort Sea, some individual polar bears are spending extended periods of time onshore during the reduced ice season and consuming onshore foods, namely the remains of subsistence-harvested bowhead whales (*Balaena mysticetus*) (Atwood et al., 2016; McKinney et al., 2017a). These onshore polar bears have shown lower concentrations of ΣCHLs, but not other legacy POPs, compared to individuals remaining on the sea ice year-round, likely because bowhead whales occupy a lower trophic position than the bears' main ice-associated prey, the ringed seal (Atwood et al., 2017). Recent observations of polar bears foraging on terrestrial species, including colonial nesting seabirds, have also been reported in the Beaufort Sea and for other subpopulations of polar bears (Iverson et al., 2014; Divoky et al., 2015; Bourque et al., 2020), which all else being equal, would serve to lower their POP exposures.

Temporal trends of lipophilic POPs and PFAS in relation to changes in feeding habits and body condition have also been investigated in adult female polar bears sampled in spring from the Barents Sea (Routti et al., 2017; Lippold et al., 2019). Temporal changes in $\delta^{15}N$ and $\delta^{13}C$ values indicate that the winter diet of Barents Sea female polar bears has shifted towards less marine, less ice-associated and lower trophic level prey items (Routti et al., 2017; Lippold et al., 2019), which is likely related to the rapid decline of sea ice in the area (Stern and Laidre, 2016). Although not statistically significant, the same polar bears also tended to get thinner over the period 1997–2006, whereas from 2006–2017 they got fatter (Lippold et al., 2019). Contaminant concentrations were strongly related to polar bear diet (lipophilic POPs and PFAS) and body condition (lipophilic POPs only), however, temporal changes in diet and body condition did not significantly affect trends of the measured pollutants (Routti et al., 2017; Lippold et al., 2019).

Climate change responses also affect POPs in terrestrial communities. The Arctic fox is an opportunistic predator and scavenger, that is among the most polluted of Arctic mammals. Although a terrestrial species, it also feeds on marine foods at coastal tundra areas, such as Svalbard. Studies investigating the influence of feeding habits and food availability on contaminant temporal trends in Arctic foxes from Svalbard reported that concentrations of all measured lipophilic POPs

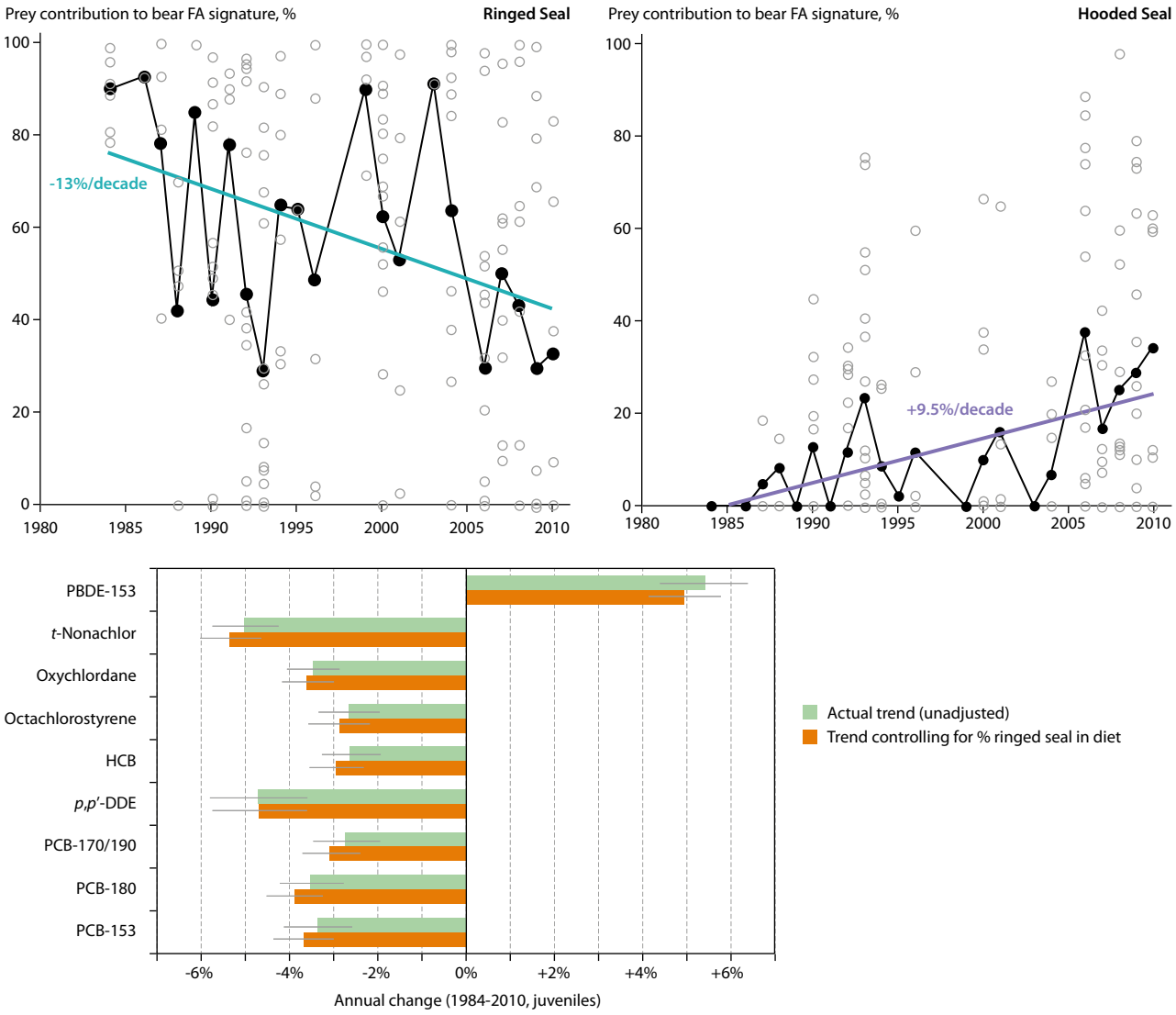

Figure 2.3.10 Dietary trends and POP concentrations in East Greenland polar bears. (top) Contribution of ringed seal and hooded seal to polar bear diets in spring-summer from 1984–2011, based on estimates from quantitative fatty acid (FA) signature analysis. Open circles represent individual bears, while black circles represent annual means. Non-significant trends for bearded seal, harp seal, narwhal, and walrus not shown. (bottom) Annual change of POP concentrations (mean +/- SE) in these polar bears, both unadjusted and adjusted (controlled) for changes in percentage of ringed seal in the diet over this same time period. Note that controlling for dietary shift did not significantly alter trends. Adapted from McKinney et al. (2013).

and PFAS increased with a higher intake of marine food items. Additionally, concentrations of β-HCH, perfluorooctane sulfonic acid (PFOS), perfluoroheptane sulfonic acid (PFHpS) and perfluorotridecanoic acid (PFTrDA) increased with an increasing availability of sea ice (Andersen et al., 2015a; Routti et al., 2017). A similar, but non-significant tendency was also observed for *p,p'*-DDE, HCB, CHLs, PBDEs and for perfluorohexane sulfonic acid (PFHxS) and the C_9–C_{11} perfluorocarboxylic acids (PFCAs). Arctic foxes use sea ice to scavenge the remains of seals killed by polar bears and to hunt newborn ringed seal pups (Gjertz and Lydersen, 1986). The increased availability of sea ice thus increased Arctic foxes' access to seals, which have higher levels of pollutants than terrestrial prey. Additionally, decreasing concentrations of HCB and PFAS in fox tissues were related to the increasing mortality of reindeer (*Rangifer tarandus platyrhynchus*) (Andersen et al., 2015a; Routti et al., 2017). Reindeer mortality has been connected to rain-on-snow events (Hansen et al., 2013), which entirely encapsulate short-growing vegetation in ice across large areas of High Arctic tundra (Peeters et al., 2019). The frequency of extreme weather events, such as warm spells and rain-on-snow events, has been increasing in Arctic terrestrial regions (Bintanja and Andry, 2017). Icing of the tundra prevents winter foraging opportunities for Arctic tundra herbivores, which affects their vital rates and has further consequences for the abundance of Arctic carnivores (Hansen et al., 2013). In conclusion, climate change may decrease POP exposures in Arctic foxes as 1) decreases in sea ice will reduce opportunities for Arctic foxes to eat more-contaminated seals and 2) increased numbers of reindeer carcasses due to more frequent rain-on-snow events will provide foxes with less-contaminated reindeer as a food source.

Behavioral changes and alternative uses of landscapes and food resources may, however, buffer the impacts of environmental change in some species (Loe et al., 2016; Hansen et al., 2019). For example, the proportion of Svalbard reindeer feeding along the shoreline has increased along with icier winters, and stable isotope values indicate that these reindeer are likely feeding on washed-ashore kelp (Hansen et al., 2019). Although not yet investigated, this may have further implications for the pollutant

intake of reindeer and their predators. In addition to increasing reindeer mortality, rain-on-snow events also promote the expansion of boreal predators into Arctic tundra. In Yamal, Russia, a high mortality event of domestic reindeer due to icing of the snow layer was followed by the increased presence of red foxes (*Vulpes vulpes*) and hooded crows (*Corvus cornix*) (Sokolov et al., 2016). Other changes, such as the 'shrubification' of the Arctic tundra, are also underway on land (Sturm et al., 2001, Tape et al., 2006), and like their marine counterparts, terrestrial fauna are also undergoing northward range shifts (Post et al., 2009). These shifts in terrestrial plant and animal communities have the potential to alter both contaminant movement and fate within terrestrial Arctic food webs.

2.3.3.3 Lipid dynamics and energy allocation

Organisms store energy as lipids for mobilization in times of food shortage. This adaptation, along with a trade-off to store energy, rather than use it for faster growth or reproduction, is favored in polar regions due to the strong seasonality in primary productivity and food availability in cold climates. In addition to influencing energy transfer within the food web, lipid dynamics directly affect the distribution of lipophilic POPs within the ecosystem.

Neutral storage lipids represent the main organic phase that non-ionic chemicals, such as most POPs, partition into (Jørgensen et al., 1997). The relationship between lipid content and POPs has rarely been studied over time in organisms with very large seasonal changes in lipid content. Arctic marine zooplankton adapted to fasting during winter hibernation, such as Calanoid copepods, store energy as energy-dense wax esters (Hagen and Auel, 2001; Scott et al., 2002; Lee et al., 2006), and not only as triacylglycerols, which act as the dominant neutral storage lipid class in vertebrates and winter-active zooplankton. As POPs partition into wax esters more than into triacylglycerols (Ruus et al., 2021), a change in the overwintering strategy of Arctic zooplankton to include more active feeding, combined with increased abundances of winter-active southern zooplankton in the Arctic, will change (and likely decrease) POP accumulation at the base of the aquatic food web.

Borealization is also expected to contribute to changes in lipid-dynamics within food webs by altering low- and mid-trophic level species compositions. As climate warms, the Arctic will likely experience an increasing abundance of species with lower lipid content, smaller size, faster growth and an adaptation for income breeding rather than capital reproduction based on stored energy (Figure 2.3.11) (Varpe et al., 2009; Fossheim et al., 2015; Renaud et al., 2018). As a result, the lipid mass at the base of the Arctic food web is predicted to decrease with a greater influx of southern secondary producers such as *Calanus finmarchicus*, and reduced influence of large, lipid-rich Arctic species, such as *Calanus glacialis* and *Calanus hyperboreus* (Renaud et al., 2018; Møller and Nielsen, 2020). However, at an ecosystem level, the overall lipid mass is expected to increase due to a more rapid turnover of copepods and a higher general biomass of zooplankton (Renaud et al., 2018).

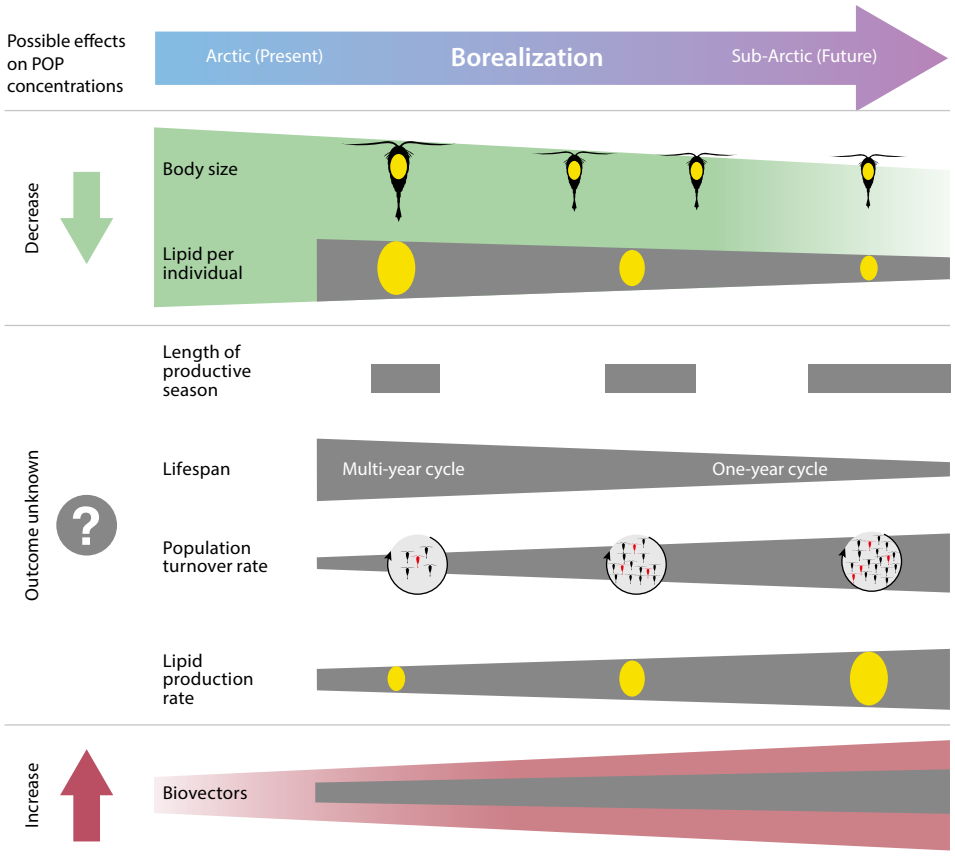

Figure 2.3.11 Projected effects of borealization on Arctic marine zooplankton and associated changes in organism POP concentrations. Adapted from Renauld et al. (2018).

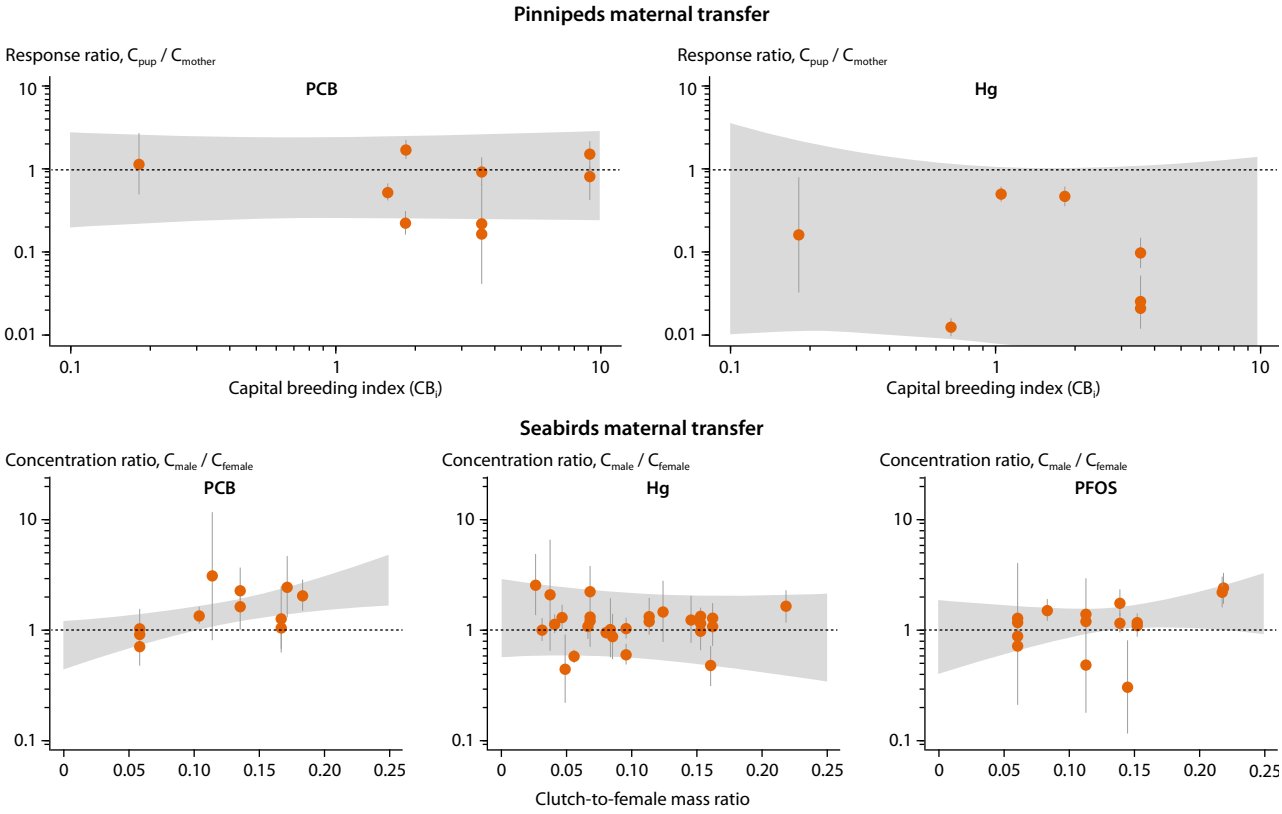

Figure 2.3.12 Relationship between reproductive strategy and maternal transfer of mercury (Hg) and POPs, including lipid-soluble PCBs and protein-associated PFOS, in marine mammal and seabird species. Response ratios in upper panels indicate the ratio between concentration in offspring (C_{pup}) and mother (C_{mother}), with values above one indicating higher concentrations in offspring, and values below one indicating higher levels in mother. Gray areas depict confidence intervals. Association of the estimated capital breeding index value and mother-to-pup contaminant transfer for various pinniped species. Capital breeding index values indicate the species' relative reproductive strategy (i.e. the degree to which the pup is dependent on income (newly acquired) or capital (stored body lipids) energy from the mother, with higher values indicating greater reliance on a capital breeding strategy. Response ratios in lower panels indicate the ratio between concentration in adult males and females, with values above one indicating higher concentrations in males, and values below one indicating higher levels in females (mothers). Clutch-to-female mass ratio represents the energy invested by the mother into the offspring. Figures from Hitchcock et al. (2017, 2019b).

An adaptation to income breeding (i.e. being less dependent on stored lipid reserves) may in turn result in lower maternal transfer of POPs to offspring due to growth dilution, and lower POP storage capacity due to lower lipid content (see Section 2.3.2.6). However, a meta-analysis of maternal transfer in marine mammals and seabirds, did not show any relationship between reproductive strategy and maternal transfer of POPs to offspring for existing mother-pup data in pinnipeds (Hitchcock et al., 2017) and maternal investment in birds (Hitchcock et al., 2019b). These studies also identified significant knowledge gaps as most results to date come from capital breeders and not income breeders (Figure 2.3.12).

In the freshwater environment, wet weight PCB concentrations and lipid content in Arctic char from a lake in southwest Greenland decreased from 1994–2008 and were negatively correlated with summer air temperatures; however, the relationship between lipid-normalized PCB concentrations and air temperatures was less clear, and did not show a significant trend (Rigét et al., 2010). As there was no indication of changes in trophic morph or trophic level, air temperature was suggested as the influential factor impacting PCB levels in the fish by indirectly altering processes that resulted in reduced condition (i.e. lower lipid content) of the fish.

The common eider (*Somateria mollissima*) is a large-bodied marine duck which undergoes large seasonal changes in lipid mass. Prior to breeding, females accumulate large lipid reserves and may subsequently lose up to 45% of their body mass during the egg laying and incubation fasting period due to lipid metabolism (Parker and Holm, 1990). Bustnes et al. (2012) investigated blood concentrations of PCB-153, *p,p'*-DDE and HCB in incubating common eiders on Svalbard and the sub-Arctic northern Norwegian mainland. As a result of their higher metabolism, Arctic birds experienced higher blood-level exposures to POPs during the incubation period than sub-Arctic birds, as their use of energy reserves led to a greater re-mobilization of lipid stores, and thus movement of associated lipophilic POPs from fat into the blood. Moreover, increases in blood POP levels were more pronounced in females with low body condition compared to females with high body condition. Based on these findings, increasing temperature in the Arctic can be expected to lead to reduced metabolism and mobilization of lipids in breeding birds and consequently lower blood-level POP exposure during breeding (Bustnes et al., 2012).

Arctic marine mammals also undergo seasonal changes in body fatness. For example, polar bears generally feed extensively in spring and early summer when ringed seals are pupping and molting on the sea ice (Atkinson and Ramsay, 1995). During sea ice-free periods, polar bears have reduced, if any, access to food, and therefore largely fast. Pregnant females spend winters in dens, fasting up to eight months in a row (Atkinson and Ramsay, 1995). The

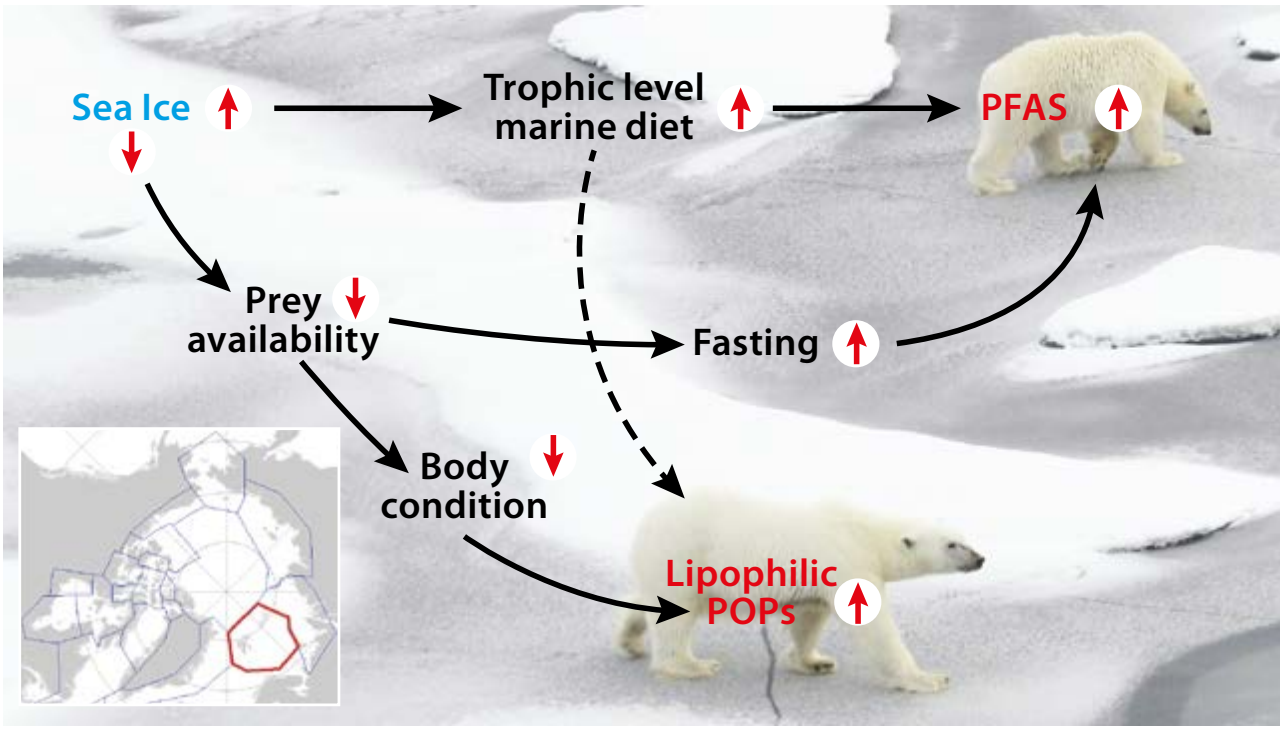

Figure 2.3.13 Diagram illustrating the relationship between sea ice variations at seasonal and regional scales and concentrations of lipophilic persistent organic pollutants (POPs) and proteinophilic per- and polyfluoroalkyl substances (PFAS) in polar bears from the Barents Sea. In seasons and areas with reduced sea ice extent, polar bears were thinner, and consequently had higher tissue concentrations of lipophilic POPs. Sea ice decline may decrease or increase PFAS concentrations in polar bears by either reducing the intake of high trophic level marine prey or promoting fasting, respectively. Location of study shown in red on inset map. Upward (↑) and downward (↓) arrows indicate increases and decreases, respectively. Modified from Routti et al., 2018.

dynamics of lipophilic pollutants in polar bears are tightly connected to annual changes in lipid accumulation, as well as overall body condition, which has been documented to have decreased over time in several polar bear management areas (Rode et al., 2012, 2018; Obbard et al., 2016; Sciullo et al., 2016;). For example, studies from the Barents Sea have shown that concentrations of PCBs are several times higher in both plasma and adipose tissue of thin female polar bears compared to fat ones, and that body condition is a more important predictor for lipophilic POP concentrations than diet as inferred from stable isotopes (Tartu et al., 2017b). In seasons and areas with reduced sea ice extent, polar bears were thinner, and consequently had higher tissue concentrations of lipophilic contaminants (Figure 2.3.13). Although the relationship between reduced sea ice, body condition, and increased levels of pollutants has been shown at seasonal and inter-annual scales, long-term changes in springtime polar bear body condition, which are likely related to sea ice conditions, did not seem to affect POP trends in polar bears (Lippold et al., 2019). Plasma concentrations of proteinophilic POPs (i.e. PFAS) were not related to changes in body fatness of polar bears from the Barents Sea (Routti et al., 2017; Tartu et al., 2017a). However, PFAS concentrations were higher in fasting polar bears than in non-fasting bears, which may be related to higher protein concentrations or reduced metabolic rates, and thus lower contaminant excretion in fasting animals (Figure 2.3.13; Tartu et al., 2017a). Concentrations of the proteinophilic Hg (measured as total Hg) were inversely related to the body mass index (BMI) of polar bears from the southern Beaufort Sea (McKinney et al., 2017b).

2.3.3.4 Behavioral changes related to sea ice cover

Climate change alters the behavior of wildlife reliant on ice, including Arctic marine mammals and seabirds, which likely has consequences for their diet and subsequent contaminant exposure. Altered foraging behaviors of little auks in East Greenland were observed between 2004–2015 as sea ice concentrations decreased; the birds spent more time flying, less time underwater, and took deeper and longer dives (Amélineau et al., 2019). Similarly, early results suggest that higher circulating Hg levels in thick-billed murres breeding in Hudson Bay, Canada are associated with lower levels of circulating triiodothyronine (the biologically active thyroid hormone), that in turn, are associated with an increased amount of time spent foraging under water. Moreover, these relationships are only evident under less optimal (i.e. poor) ice conditions (e.g. 2016, 2017) (Elliott and Fernie, 2019). Similar patterns for POP levels may also be anticipated and are currently under investigation (Elliott and Fernie, unpubl. data).

Behavioral changes have also been observed in marine mammal species, but their potential links with contaminants remain to be investigated. For example, following a period of drastic sea ice cover decline, beluga whales around Svalbard spent less time near glacier fronts and more time in fjords, which likely shifted their diet consisting only of Arctic fish species to one that includes more Atlantic species (Vacquié-Garcia et al., 2018). Adult ringed seals, on the other hand, now spend more time near tidal glaciers, where they presumably still find Arctic prey (Hamilton et al., 2019). Most young ringed seals do, however, undergo annual migrations to marginal ice zones during the

summer/autumn, where they preferentially use areas with 50% sea ice concentrations for foraging and resting (Freitas et al., 2008; Hamilton et al., 2015; Lone et al., 2019). As the marginal ice zone of the Barents Sea has shifted northward in past decades, ringed seals are now more likely to select a less optimal habitat, which increases their foraging effort and likely their energetic costs (Hamilton et al., 2015; Lone et al., 2019).

The sea ice habitat of polar bears has undergone dramatic change over the last few decades (Durner et al., 2009; Stern and Laidre, 2016; Lone et al., 2017). In several parts of the Arctic, the optimal sea ice habitat for polar bears has moved further from coastal denning areas, forcing them to move longer distances or spend longer portions of the year in lower-quality habitats with reduced access to high-quality prey (Lone et al., 2017; Ware et al., 2017; Laidre et al., 2018a). Reaching sea ice habitats far from coastal areas is energetically demanding, as energy requirements are tightly related to movement patterns (Ware et al., 2017; Pagano et al., 2018), and swimming is particularly costly for polar bears (Griffen, 2018). Studies from the Barents Sea indicate that 'offshore' polar bears, which migrate over long distances to reach optimal sea ice habitat, have higher energetic costs than bears that remain within coastal areas as the sea ice retreats (Blanchet et al., 2020; Blévin et al., 2020). Long-distance movements and energy requirements as factors influencing diet and contaminant exposure in coastal and offshore polar bears from the Barents Sea were investigated by Blévin et al. (2020). A comparison of polar bears with similar body conditions showed that offshore polar bears were exposed to higher concentrations of pollutants than coastal bears. This was found to be related to differences in feeding habits, energy expenditure, and geographical distribution. Offshore bears fed on a marine-based and higher trophic level diet, whereas coastal bears relied on terrestrial and lower trophic level prey. Offshore bears also had higher energy requirements, and thus, higher food and contaminant intake than coastal bears. Furthermore, offshore polar bears were distributed farther north in marginal ice zones, where the uptake of pollutants is more efficient than in open water areas, and further east where seal prey are more polluted, likely as a result of closer proximity pollutant emission sources and transport pathways. However, offshore bears were, on average, fatter than then coastal ones (Tartu et al., 2018; Blanchet et al., 2020), and consequently measured plasma concentrations of lipophilic POPs were similar in bears with different strategies, and only proteinophilic PFAS were higher in pelagic bears (Tartu et al., 2018).

Conclusively, these studies indicate that the fate of pollutants in polar bears from the Barents Sea is related to the climate-driven shifts in their sea ice habitat. If the costs of migration override the benefits of the energy-rich, high trophic-level prey in the future, offshore bears may exhibit higher concentrations of both lipophilic POPs and PFAS than coastal bears. Also, any change in movement strategies is likely to affect the fate of pollutants in polar bears.

2.3.3.5 Ecological food web changes due to increased temperature

As water temperatures increase, the metabolism and food demands of fish and other ectotherms also increase, leading to an overgrazing of lower trophic levels (Piatt et al., 2020). Moreover, the elevated metabolic demands associated with higher water temperatures can lead to reduced prey fish quality and quantity, with cascading effects for fish, seabirds, and marine mammal predators. This may be of particular significance for seabirds with high energy demands, such as the thick-billed murre that eats more than 50% of its body mass in fish per day. A decrease in prey quality and quantity, in addition to increased competition for available prey, has previously led to mass die offs of as many as one million murres based on estimates of those washed ashore (Piatt et al., 2020). This increased mortality, combined with failed reproduction, leads to significant ecological changes in marine food webs of both pelagic and benthic communities due to subsequent the sinking of carcasses at sea. Previous studies have shown high levels of various contaminants and enantiomer-selective accumulation in Arctic benthic scavenging amphipods (Borgå and Bidleman, 2005; Bidleman et al., 2013). Contaminant levels in these benthic amphipods were as high as those in marine mammals and seabirds (Svendsen et al., 2007). Thus, climate change and extreme events resulting in mass mortalities may lead to a shift in energy and contaminants within and between food webs, including from pelagic to benthic food webs as a result of the sinking of carcasses at sea, and from marine to terrestrial food webs in the case of seabird mortalities occurring at or near land-based nesting colonies (Evenset et al., 2007; Kristiansen et al., 2019).

2.3.4 How will climate change, in combination with other stressors, affect contaminant toxicity?

Measuring and predicting the effects of climate change on contaminant exposure in Arctic ecosystems is challenging because such interactions do not occur in isolation; climate change and environmental pollutants may interact with numerous other environmental and health stressors that may lessen, enhance, or produce unexpected impacts. Concurrent environmental stressors including ocean acidification, the presence of litter and plastics, increased fishing and hunting pressure, changes in food availability, oxygen depletion, harmful algal blooms, competition from invasive species, habitat destruction and biodiversity loss, along with a range of social, economic, and political factors, such as increasing human migration, resource exploration and extraction, local development, ship traffic, and recreation and tourism activities, may interact with the stress of climate change and contamination, resulting in both direct and indirect responses in ecosystems and organisms.

Novel infectious diseases, changes in pathogen distribution, and an increasing presence of zoonotic pathogens are likely to occur with climate change (Dietz et al., 2019 and references therein). The physiological stress induced by increased temperature might impair the immune system in cold-blooded organisms such as fish, rendering them more susceptible to infection and disease (Mariana et al., 2019). The rate of phocine distemper virus (PDV) infection in pinnipeds from the North Pacific Ocean was recently found to be significantly higher in years following the presence of an open water route along the northern coast of Russia (VanWormer et al., 2019). The authors concluded that reduced sea ice removed a barrier to pinniped movement, thus allowing viral transmission

between the North Pacific and eastern North Atlantic Oceans. Although the role of contaminants on disease transmission or animal health was not investigated in this study, some POPs have been shown to suppress immune responses, specifically in marine mammals (AMAP, 2018b). Taken together, it is conceivable that the combined effects of altered environmental conditions (i.e. temperature-stress), contaminant exposure (i.e. immunosuppression) and ecological changes (e.g. ice loss leading to greater disease transmission), could have greater impacts to animal health than any one factor alone.

Adding another layer of complexity, organisms are simultaneously exposed to multiple types of environmental contaminants with varying toxicities, including but not limited to POPs, CEACs, Hg and other potentially unrecognized chemicals or substances. More recently, plastics, including microplastics and nanoplastics, have been identifed as substances that may interact with contaminants, or regardless, affect organisms directly (AMAP, 2017a). Contaminants have been shown to bind to plastic particles with high affinity, and thus plastics may act as vectors transporting chemicals to the Arctic, within the Arctic, and facilitate their uptake by organisms. Moreover, plastics contain chemical additives, and therefore may serve as a source of new contaminants to Arctic ecosystems. Plastics are also substrates for microbiota which may play a role in ecosystem functioning. However, as there are currently no studies addressing how climate change directly or indirectly interacts with microplastics to affect contaminants within Arctic food webs, this topic is not addressed further here. See Chapter 2.2 (Box 2.2.2) for additional details on microplastics in the abiotic environment.

Climate change may also affect an individual's sensitivity to toxicants (i.e. climate-induced toxicant sensitivity; Moe et al., 2013; Noyes and Lema, 2015; Hooper et al., 2013) by altering the toxicokinetics (i.e uptake, distribution, metabolism and elimination of toxicants) and toxicodynamics (i.e. toxicant interactions with biological receptors and enzymes) of contaminants (see Section 2.3.2.1). Such changes may also alter a species' sensitivity and thresholds for effects, including lowering effect thresholds such that impairments are experienced with lower levels of contaminant exposure, as seen in developing children (Stein et al., 2002).

Most studies investigating environmental pollution in combination with other stressors usually focus on the impacts of climate-related environmental parameters (e.g. temperature increase or drought). Most, if not all species have an optimal range of temperatures for which they can maintain homeostasis, outside of which, their ability to function may be impaired. However, the effects of environmental temperatures changes are especially pronounced in ectothermic organisms which heavily rely on external sources of heat to control body temperature. Ectotherms display a reaction norm (general performance curve) in response to temperature, whereby they have an optimal temperature window, as well as upper- and lower- critical limits (Figure 2.3.14). The response variable (i.e. performance) reflects important determinants of fitness, such as life history traits (e.g. age at maturity), that directly affect the vital rates of a life table (e.g. survival, fecundity, growth). The ability of an organism to cope with other stressors, such as toxicants, depends on where within this temperature range they are (Sinclair et al., 2016).

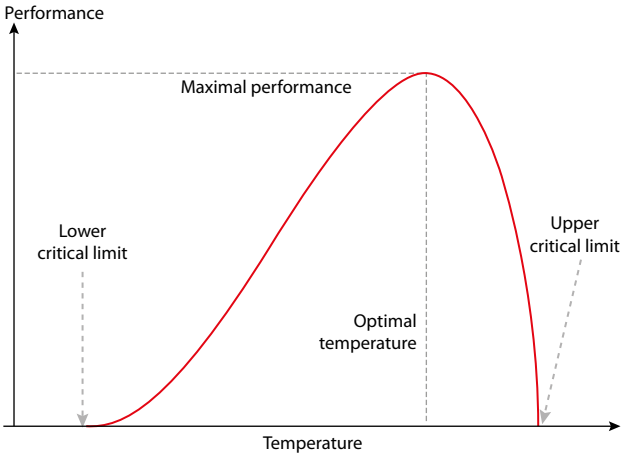

Figure 2.3.14 General performance curve (reaction norm) demonstrating the effect of temperature variation on fitness for ectotherms.

Understanding the effects of multiple stressors, compared to a single stressor such as temperature, can be more challenging. Usually, exposures to multiple stressors are predicted to have a synergistic outcome, in other words, a combined effect that is greater than the sum of the two. However, most laboratory-based studies are designed such that synergistic effects are also the most likely outcome; for example many studies investigate the effects resulting from two stressors under continuous exposure at the same time (Gunderson et al., 2016), when real exposure regimes most likely might differ from this scenario. Organisms do not always experience constant and simultaneous exposure to all stressors; rather, exposures can be pulsed, or experienced out of phase with one another, depending on habitat, season or other variables. Exposure to stressors such as climate change and toxicants may be on small or large spatiotemporal scales, static or dynamic, and the timing of responses and their duration might differ depending on the stressor. Taking these more realistic exposure scenarios into account by recreating pulsed and/or staggered exposures, and including an organisms' time for a compensatory physiological response to maintain homeostasis, might result in combined effects that are additive or antagonistic, rather than synergistic, and provide more ecologically-relevant outcomes.

For example, most studies consider climate change in terms of mean temperatures. However, recent attention has been given to the magnitude of daily temperature fluctuations, because not only the daily mean, but also the magnitude of daily fluctuations around the mean, are predicted to increase. Recent non-Arctic studies suggest that pesticide toxicity increases with daily temperature fluctuations as illustrated by the increased mortality and growth rate of aquatic damselfly *(Ischnura elegans*) larvae after chlorpyrifos exposure under conditions of fluctuating daily temperatures (Verheyen and Stoks, 2019). Chlorpyrifos exposure under constant temperature did not cause mortality nor changes in growth rates. Chlorpyrifos exposure under fluctuating daily temperatures also resulted in reduced energy storage (fat content) and increased enzyme activity compared to exposures under constant temperature conditions. These findings demonstrate that natural variations and fluctuations can influence responses to multiple stressors, and therefore are important considerations for both laboratory- and field-based studies.

Correspondingly, the health effects of multiple stressors, including those related to climate change, are increasingly being identified as knowledge gaps for the Arctic (Dietz et al., 2019; UNEP/AMAP, 2011). Multi-stressor laboratory experiments can aid in understanding the combined effects of climate change and pollution, however, experiments specific to Arctic biota are still scarce, as organisms can be difficult to obtain due to the inaccessibility, harsh conditions, and high costs of research in the region. Although relevant POP multi-stressor laboratory studies are currently lacking, studies on polyaromatic hydrocarbons (PAHs) can provide insights as these petroleum-related substances share some properties of POPs and therefore behave similarly in some species.

Laboratory studies on copepods have explored life stage-, gender- and species-specific responses to combined exposures of elevated temperatures and a crude oil-associated PAH, pyrene (Grenvald, et al., 2013; Hjorth and Nielsen, 2011; Van Dinh et al., 2019). By transferring energy from primary producers to higher trophic levels, copepods are an essential link in the Arctic marine food web. When exposed to elevated temperatures and pyrene, survival of the naupliar stages of the Atlantic copepod *C. finmarchicus* and the true Arctic copepod *C. glacialis* decreased with increasing temperature and pyrene concentration, but more so for *C. finmarchicus* than for *C. glacialis* (Grenvald et al., 2013). Pyrene negatively impacted older naupliar stages of both species, as it delayed development into the next naupliar stage.

A separate study examining the combined influences of elevated temperature, pyrene exposure, and food concentration on the grazing rates of adult *C. finmarchicus* found males and females displayed different sensitivities to multiple stressors. Reduced grazing rates were only observed at higher food concentrations in both sexes, however, females, but not males, exhibited lower grazing rates when exposed to higher temperatures and pyrene. The interactive effect of temperature and pyrene was found to be insignificant, suggesting the observed differences were more likely related to the interaction of gender, food availability and pyrene exposure (Van Dinh et al., 2019). Mortality rates of adult *C. finmarchicus* were not reported, making comparisons to results from the naupliar stages in other studies difficult, however, the sex-specific responses to the combined stress of elevated temperature and PAH exposure illustrate the importance of intra-specific variation in the susceptibility to multiple stressors (Van Dinh et al., 2019).

In recent years, Arctic cod has been the subject of an increasing number of laboratory studies. This fish has a pan-Arctic distribution and is a keystone species in the Arctic food web, connecting lower and higher trophic levels (Hop and Gjøsæter, 2013). Arctic cod is directly affected by climate change as warming temperatures constrain their distribution and reduce under-ice spawning habitat (Dahlke et al., 2018; Marsh and Mueter, 2019). Heat stress caused by increased temperatures close to the upper critical limit of optimal homeostasis (Figure 2.3.14) might lead to elevated oxidative stress through an increase in reactive oxygen species. Overproduction of reactive oxygen species may impair normal functioning by affecting cellular metabolism, immune system responses, detoxification processes, DNA damage, expression of heat shock proteins, and cellular apoptosis (Iwama et al., 1999; Mariana and Badr, 2019; Petitjean et al., 2019). These responses are also affected by environmental contaminants. Enhanced growth and stronger year classes have been associated with warmer temperatures and reduced sea ice (Bouchard and Fortier, 2011; Bouchard et al., 2017). With warmer temperatures, metabolic rates also increase; for example, oxygen consumption almost doubles in Arctic cod embryos with a temperature shift from 0°C to 3°C (Dahlke et al., 2018). Rapid biotransformation responses after exposure to crude oil and elevated temperatures suggest adult Arctic cod may have the ability to cope with acute PAH exposure (Andersen et al., 2015b). However, it remains difficult to project the interaction effect of higher temperatures and contaminant exposure to Arctic cod and subsequent cascading effects up the food chain in a warming Arctic.

Modeling was used to evaluate the effect of multiple stressors including climate change, pollution, and egg predation on population dynamics and viability in the sub-Arctic common eider duck (Bårdsen et al., 2018). Eider ducks feed low in the food web and generally have low contaminants levels, apart from periods in the breeding season, such as late in the egg incubation period, when the female has fasted for approximately 20 days. During incubation the use of lipids for energy results in the re-mobilization of contaminants from lipid stores and a corresponding increase of contaminants in blood (Bustnes et al., 2010). Thus, when body condition is poor (i.e. low lipid stores), circulating contaminant levels are high. Projections of eider duck population growth and abundances using the Leslie matrix population model showed that egg predation alone was a strong enough factor to cause population extinctions, whereas an increase in sea surface temperature and reduced clutch size due to elevated contaminant exposure alone did not (Bårdsen et al., 2018). However, any combination of the three stressors reduced the population growth rate, and the effect of pollution on clutch size was increasingly negative when co-occurring with a warming climate and increased egg predation. Population viability was lowest when all stressors occurred simultaneously. For pollution alone to be a significant stressor on the population through effects on clutch size, the level of exposure needed to be unrealistically high, however, when co-occurring with increased predation pressure, the threshold level needed for pollution to cause extinctions was lower (Bårdsen et al., 2018).

Field-collected data on wildlife populations have also been used to investigate the impact of multiple stressors. The great skua (*Stercorarius skua*), a seabird that breeds in the North Atlantic from Iceland and the Shetland Islands, Scotland to Svalbard, showed organochlorine and organobromine contaminants had negative effects on return rate (i.e. survival), and that breeding colonies had different sensitivities to contaminant exposure (Bustnes et al., 2015). Effects occurred at lower contaminant exposure levels in breeding colonies that were in poorer body condition, probably as a result of food limitations. The impact of multiple stressors on contaminant levels in free-ranging wildlife has also been shown for polar bears in the marginal ice zone of the Barents Sea (see Box 2.3.1).

Several studies suggest that POP exposure has adverse effects on lipid metabolism, immune function, circulating thyroid

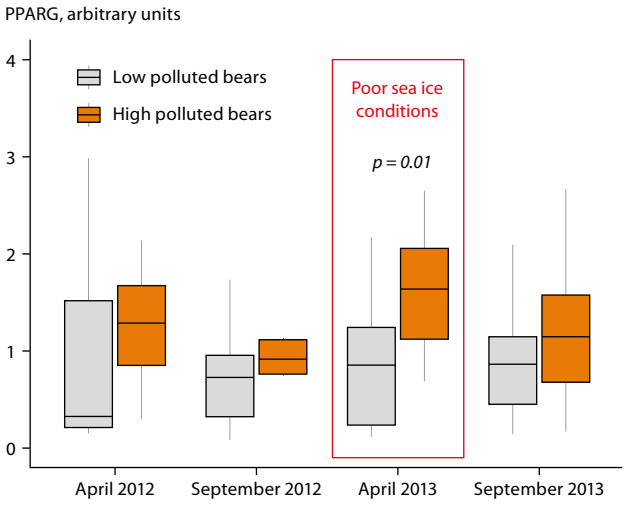

Figure 2.3.15 Transcript levels of a key regulator of lipid metabolism, peroxisome proliferator-activated receptor gamma (*PPARG*), in adipose tissue of low- and highly-polluted female polar bears from the Barents Sea are more contrasted during a period with poor sea ice conditions. Data from Tartu et al. (2017c).

hormones and neurochemistry of polar bears (Routti et al., 2019a) and climate change is likely to affect the sensitivity of polar bears to these adverse effects of pollutants. A wide range of biomarkers for energy metabolism, specifically gene transcript levels in fat, physiological parameters, and metabolomic and lipidomic markers in plasma, were investigated for associations with pollutant exposure in female polar bears from Svalbard (Tartu et al., 2017c). Several biomarkers involved in lipid metabolism were related to POP exposure. Furthermore, the differences between the biomarker responses of low- and highly- polluted bears were more contrasted during a period with poor sea ice conditions (Figure 2.3.15), suggesting that contaminant exposure and sea ice decline may have compound adverse effects on polar bears.

The decline of sea ice and increased pollutant exposure may also have additive or synergistic effects on polar bear susceptibility to disease. In vitro and correlative field studies indicate that the polar bear immune system is compromised by pollutant exposure, as well as by climate change. For example, polar bears from the southern Beaufort Sea staying for extended time periods on shore had heightened immune responses and were more exposed to pathogens than bears remaining on the sea ice year-round (Atwood et al., 2017; Whiteman et al., 2019). Also, concentrations of a stress hormone, cortisol, were higher in polar bears with low body condition, indicating that thinner polar bears were more stressed, and thus potentially more prone to adverse effects of pollutants, than fatter bears (Mislan et al., 2016).

The many overlapping changes occurring in the Arctic will challenge the ability of ecological communities to adapt and maintain resilience (AMAP, 2017c, 2017d, 2018c). The interaction between toxicants, climate change and other environmental changes is complex and adds to the difficulty in predicting effects in wildlife, however the impacts observed in the Arctic environment may be able to function as early warning signals for other regions (Borgå, 2019).

2.3.5 What can we learn from modeling climate change effects on food web accumulation?

The use of mechanistic models to understand Arctic environmental distribution, food web accumulation, and human exposure to contaminants is summarized in Wania et al., (2017). Various models focused on predicting exposure to Arctic indigenous populations are reviewed, including those that estimate human exposure directly stemming from consumption of specific traditional food items, as well as those that include the bioaccumulation of contaminants through the Arctic food web in calculations of human exposure. Further, Wania et al., (2017) show how models can be used to identify the range of physicochemical properties typical of chemicals that undergo LRT to the Arctic and accumulate in the food web. Models were also used to identify drivers of the contaminant temporal trends measured in indigenous populations and to investigate the link between estimated contaminant exposure in early life stages and adult human health parameters (Wania et al., 2017).

The published work on mechanistic modeling of food web POP accumulation in response to climate change over the past decade is scarce and does not always apply to Arctic or polar regions. Nevertheless, the task of evaluating how climate change will affect food web POP bioaccumulation can be divided into the efforts of i) combining abiotic contaminant transport models with biotic energetics and bioaccumulation models, and ii) isolating the climate dimension and its contribution to bioaccumulation. The combined environmental fate and transport model, CoZMo-POP (Wania et al., 2006), and the bioaccumulation model, ACC-Human to CoZMoMAN (Czub and McLachlan 2004; Breivik et al., 2010), are good examples of models combining abiotic transport, biotic energetics, and bioaccumulation models that are built on similar fugacity-based frameworks. To identify the influence of climate change on POPs using these or similar models, a climatic mode is included in the abiotic transport model to estimate changes to contaminant transport and exposure at the base of the food web (e.g. Lamon et al., 2009). This modeling framework has been used to assess the potential implications of various climate change scenarios for long-term human exposure to POPs in the Arctic (Armitage et al., 2011). When modeling the most important processes contributing to future Arctic human exposure to POPs (i.e. emissions, transport, abiotic vs. biotic changes), Armitage et al. (2011) concluded that climate-induced changes to food web biomagnification would be most influential. Details on climate-related impacts to the physical environment that would influence contaminant exposure are found in Chapter 2.2.

The AQUAWEB bioaccumulation food web model (Arnot and Gobas, 2004) was used to explore how future climate scenarios of increased ambient temperatures (+2°C and +4°C), increased primary production (+50% and +100%), and decreased organism lipid content (-10%), would affect contaminant levels through the Arctic marine food web from copepods to seabirds (Borgå et al., 2010). Due to higher predicted levels of primary production, and therefore an increase in organic particles in the water, the net overall bioaccumulation in the food web was reduced. This reduction was greatest for the most hydrophobic

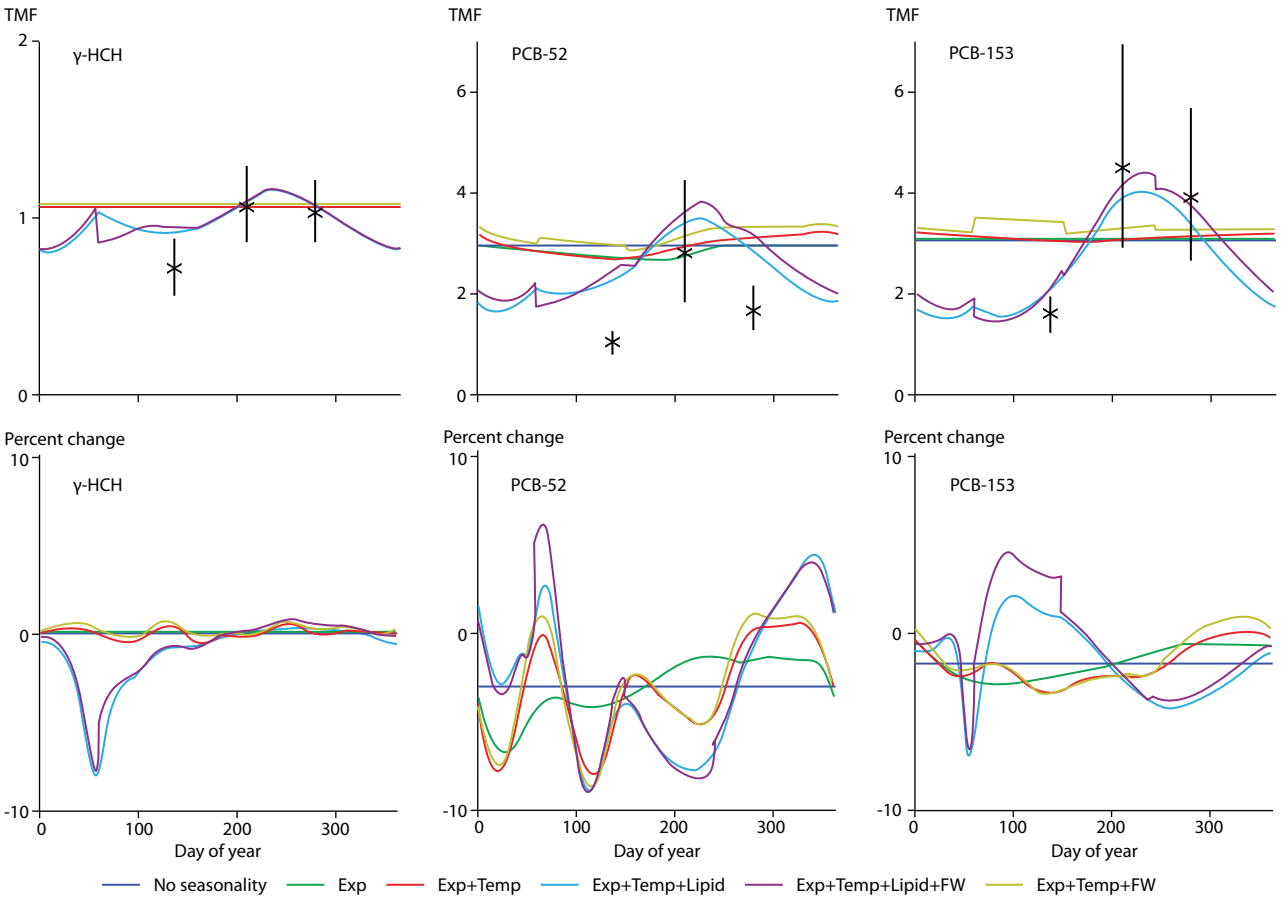

Figure 2.3.16 Seasonal changes in empirical and model-derived trophic magnification factors (TMFs) for various POPs in an Arctic marine food web. (upper panels): Empirical TMFs compared to model projections for the year 2007 estimated under various scenarios, including combinations of no seasonal variation (no seasonality), and seasonal variations in abiotic exposure (Exp), temperature (Temp), lipid content (Lipid) and food web structure (FW). Empirical TMFs from Kongsfjorden in 2007 (Hallanger et al., 2011c) are depicted with the mean and error bars. (lower panels): Percent change in model-derived seasonal TMFs in a future climate state in the year 2107 compared to 2007.

compounds subjected to the largest temperature increase used in the model. The reduced bioavailability of POPs predicted by the model resulted from the projected increases in primary production. However, the actual effect of climate change on primary production is uncertain, and the potential for both increased and decreased levels of primary production under future climate change scenarios have been reported.

To approximate a more realistic climate scenario, one study modeled how seasonal changes in abiotic exposure, water temperature, lipid content, and food web structure affected contaminant biomagnification within an Arctic marine food web (AMAP, 2016a) (Figure 2.3.16). The annual cycle of temperature and baseline water contaminant exposure was estimated using the Danish Eulerian Hemispheric Model (DEHM) in climate mode to compare two years a century apart (2007 and 2107) (AMAP, 2016a). The AQUAWEB bioaccumulation food web model (Arnot and Gobas, 2004), parameterized for an Arctic marine food web (Borgå et al., 2010), was used to model seasonally-dynamic, temperature-sensitive parameters affecting contaminant uptake and elimination, seasonal lipid content, and food web structure. The best fit between present day (2007) empirical and modeled seasonal food web biomagnification, as assessed by trophic magnification factors (TMFs), resulted primarily from the inclusion of seasonal variations in lipid content; seasonal changes in temperature, abiotic exposure, and dietary relations had only minor effects on food web biomagnification. Since food web structure did not contribute to explaining variation in seasonal food web biomagnification, it was not altered in the model scenario of future Arctic climate. Modeled food web biomagnification for the year 2107 differed only marginally from that of 2007, with magnitude and direction depending on the contaminant modeled; the mean change from 2007 to 2107 was negative for PCB-52 (-3.1%) and PCB-153 (-1.7%), and positive for δ-HCH (0.4%). Thus, the seasonal variation in food web biomagnification observed over a single year is greater than the change expected in response to a predicted future climate state, at least for the three POPs modeled. These results also emphasize the importance of understanding the different dimensions of environmental and physiological variation for interpreting the future impacts of climate change. Additionally, the model's best-fit seasonal scenario was that which included annual variations in lipid content, indicating this physiological factor is also crucial for making sound predictions of contaminant food web bioaccumulation for the future. Changes in abiotic drivers alone are not sufficient to explain food web bioaccumulation on a temporal scale. As the focus of this study was the effect of climate change on food web

accumulation, the DEHM model scenarios for each year were run using the same contaminant emission levels, however, it is expected that changes in contaminant emissions will occur in the future and will have a greater influence than climate change on exposure (see Chapters 2.1 and 2.2).

Models have also been used to identify the effect of climate change on food web accumulation by comparing the outcomes of different ecosystem conditions under IPCC-based future climate scenarios. In the Laurentian Great Lakes of Lake Erie and Lake Superior, four climate scenarios were contrasted by combining model parameters on bioenergetics (i.e. temperature and body-size effects on consumption and respiration) and bioaccumulation of PCB-153 for various fish species with different temperature regimes (Arnot and Gobas, 2004; Ng and Gray, 2009, 2011). The authors predicted that future climate conditions would lead to higher growth and feeding rates, and thus increased bioaccumulation. However, the specific conditions leading to higher growth differed by species; high temperatures hampered growth in cold-water species such as the native forage fish, mottled sculpin (*Cottus bairdii*) and the predator species, lake trout (*Salvelinus namaycush*), whereas low temperatures lowered the growth of warm-water species, such as the invasive forage fish, the round goby (Ng and Gray, 2011). If we speculate on how this translates to Arctic ecosystems, expected temperature increases would result in reduced growth and lower bioaccumulation in Arctic-resident, cold-water species, and increased growth and higher bioaccumulation in warm-water species moving north due to borealization. Note that changes in diet due to climate change-induced alterations in predator-prey interactions were not taken into account, and thus the effects of feeding and dietary uptake on growth rate and accumulation due to climate change are not known.

2.3.6 What research tools can be used to assess the effects of multiple stressors on wildlife and ecosystem health?

Herein we've mostly provided examples of research reporting on the changes in exposure, bioaccumulation, and toxicity of single chemical or chemical groups associated with global climate change, although the exposure to multiple contaminants, in addition to multiple stressors, is a more realistic scenario (see Section 2.3.4). Assessing the potential effects from these complex exposures, however, is challenging. A recent review of the biological effects of contaminants in Arctic wildlife and fish (Dietz et al., 2019) discussed different toolboxes that could potentially be used to assess the effects of toxicant mixtures and additional environmental factors on wildlife health. For example, risk quotient analyses, which assess links between contaminant exposures and various endocrine, reproductive or immune system effects could be used (Dietz et al., 2019 and references therein).

More commonly used indicators of risk, such as biomarkers of exposure or effects, are usually tied to a specific toxicant or specific modes and mechanisms of action. There has been a shift, however, towards realizing the need for a broader perspective to address the combined effect of multiple stressors, where integrated responses at a higher level of biological organization are addressed (Segner et al., 2014). One such response is the overall allocation and use of energy by an organism, which can be assessed using dynamic energy budgets (Jager et al., 2006; Jager and Zimmer, 2012; Jager, 2019). Typically, such integrated responses can be linked to the fitness of organisms and are easier to extrapolate to population-level effects using, for example, individual-based population models, as shown for eider ducks in Section 2.3.4 (Bårdsen et al., 2018). Ideally, climate change and exposure models (see Section 2.3.5) should be combined with effect models that assess the combined effects of multiple stressors on population levels. These may include models investigating changes in vital rates in response to pollutants and environmental parameters, as suggested for polar bears (Routti et al., 2019b). Still, empirical studies of climate change-sensitive toxicity on population-relevant parameters, such as survival and reproduction, are needed to support and ground-truth model projections.

With the large and growing number of contaminants present in the Arctic, there is still a need to investigate their interrelationships and the underlying structure in their variance. Although not new, models are still very useful tools for condensing and understanding the multidimensional space represented by contaminants and their drivers in the ecosystem.

Various omics methods are increasingly being used to identify changes in individual gene expression and regulation due to contaminant exposure and other factors affected by climate change (e.g. sea ice extent, body condition). These studies are transitioning from initial screening phases ('stamp collection') to more developed phases of process understanding. Combined with adverse outcome pathways that identify the sequence of cellular events leading to an adverse effect (e.g. Leist et al., 2017), these molecular-based methods hold promise for a more holistic understanding of individual and ecosystem health under multiple stressors by reflecting approaches used in human toxicology, and having the exposome and an integrated 'one health' view as goals on the horizon.

2.3.7 Research conclusions, recommendations, and future perspectives

Current research suggests that climate change will impact the LRT of pollutants to the Arctic and within the Arctic by altering atmospheric, environmental, and ecological processes, and thus will influence the exposure and accumulation of POPs in Arctic wildlife. Studies have reported changes in POP tissue concentrations in relation to climate-induced alterations of species trophic interactions that are driven by changes in abundance, habitat range, accessibility of prey species, and changes in sea ice coverage and/or temperatures, as shown for polar bears, ringed seals, and Arctic seabirds (McKinney et al., 2009, 2010, 2015; Braune et al., 2014, 2015a) (Figure 2.3.17).

A summary of the research findings, conclusions, and knowledge gaps identified in this review are provided in Table 2.3.1. Conclusions regarding climate-driven alterations to the physical environment and ecosystems in the Arctic are provided below for questions derived from the headings of Sections 2.3.2 through 2.3.5 of this chapter. Recommendations in relation to each conclusion are shown in italics directly below the relevant conclusion.

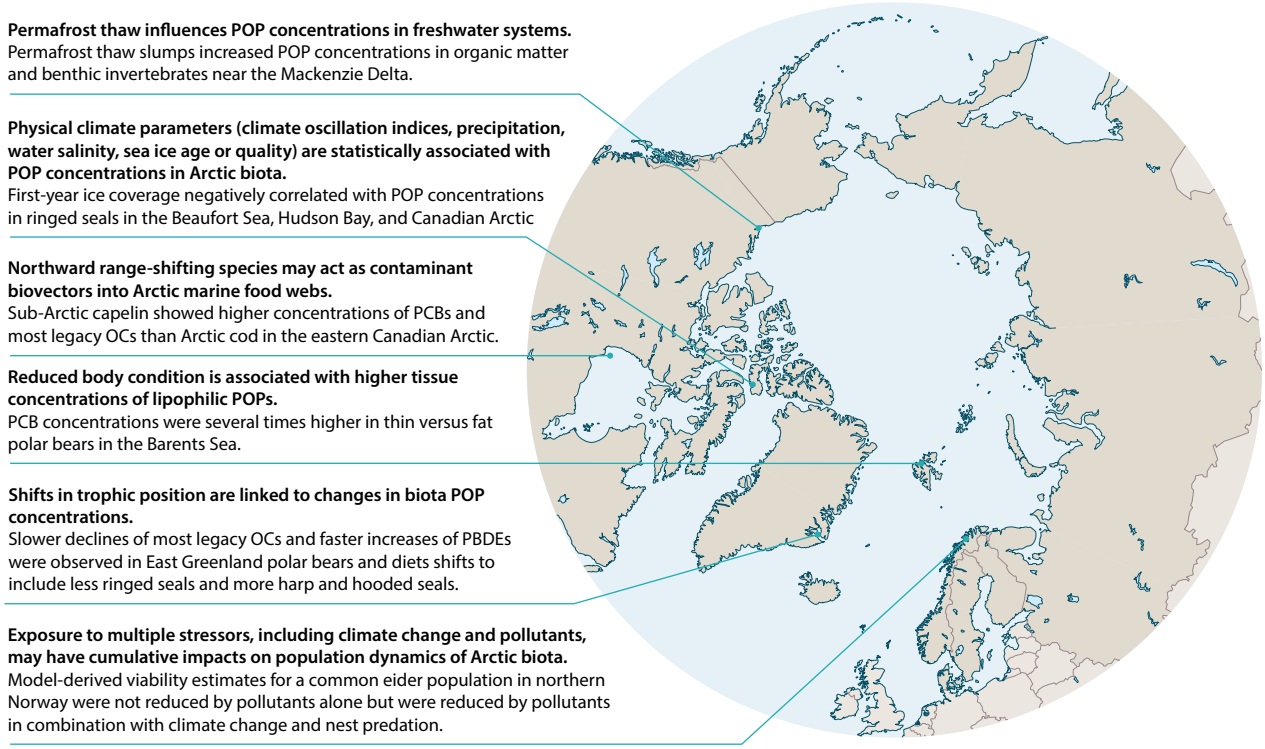

Figure 2.3.17 Major observations of climate change influences on POPs in Arctic ecosystems and food webs. Note that the highlighted processes are likely occurring across the Arctic but are listed according to where the research findings included in this review originated.

2.3.7.1 Conclusions and recommendations

How do climate-driven changes in temperature affect POP exposure and bioaccumulation in Arctic biota?

Conclusion: Temperature increases can affect the LRT of POPs to the Arctic and re-mobilization of POPs from secondary sources within the Arctic (see Chapters 2.1 and 2.2), thus potentially increasing exposure of Arctic biota to POPs. In combination with compound-specific physical-chemical properties, and species-specific biotransformation capacities, environmental temperature changes could affect the bioavailability of POPs for direct uptake, as well as temperature-sensitive uptake and elimination processes, especially for cold-blooded organisms. However, current findings from models suggest that climate-related changes in temperature may have minimal effects on bioaccumulation. Yet, some evidence suggests seasonal fluctuations in temperature could contribute to intra-annual variations in contaminant accumulation.

Recommendation: The net effect of temperature on POP distribution and fate in the physical environment has been investigated (see Chapters 2.1 and 2.2*) and will likely increase LRT and mobilization of contaminants to and within the Arctic. Therefore, knowledge of how sources and emissions are changing will be important for interpreting contaminant changes in Arctic wildlife. However, the effect of temperature on contaminant bioavailability, uptake, and elimination rates in biota that may ultimately define POP levels in an organism is less clear and requires further evaluation. There is a need for empirical data demonstrating how the effect of temperature change on the uptake and elimination rates of POPs and CEACs with various physiochemical properties and recalcitrance depend on species-specific temperature performance curves.*

Do variations in large-scale climate patterns affect POP exposure and bioaccumulation in Arctic biota, and if so, how?

Conclusion: Currently, only a small fraction of the vast collections of climate data and POPs monitoring data that are available have been studied together. Of those that have, several studies have reported higher concentrations of POPs in Arctic biota following NAO+ and AO+ conditions compared to time periods following NAO- and AO- conditions. Additionally, higher concentrations were observed in Arctic biota in relation to a greater influx of Atlantic ocean currents into the Arctic. These findings suggest higher POP exposure in Arctic biota following time periods with enhanced transport of air and water masses from North America and Europe toward the Arctic, but other underlying mechanisms may also be involved.

Recommendation: Additional studies that include more species, time points, and locations (including freshwater and terrestrial ecosystems), are required for examining the relationship of meteorological parameters (i.e. wind and precipitation patterns) to biota POP concentrations; however, the use of climate indices to investigate the effects of climate change on pollutant exposure in Arctic ecosystems is not straightforward (Hermanson et al.*, 2005, 2010; Stock* et al.*, 2014). The use of meta-analysis in ecotoxicology is still in its infancy, but could be applied in these studies if both monitoring programmes and individual research studies make raw data publicly available, or at least report metadata properly beyond its specific use in a particular study (Bechshoft* et al.*, 2018; Hitchcock* et al.*, 2018). Future studies should include ancillary biological and ecological data (e.g. body condition, lipid content,*

fatty acid profiles, and stable carbon, nitrogen, and sulphur isotopes) to disentangle the impacts of climate-driven physical changes from those resulting from ecological or physiological effects on exposures in biota. Furthermore, temporal trend studies on the impact of a changing climate on the ecotoxicology and environmental chemistry of POPs in the Arctic have typically used climate variables averaged over large spatial and temporal scales (e.g. month, season or year; see Chapter 2.2 on abiotic temporal trends). However, potentially more relevant, higher-resolution spatial and temporal data and variability (e.g. local sea ice conditions, daily and seasonal temperature variations) should be considered, if possible. Considering the potential effects of intermittent and/or extreme weather events (e.g. rain-on-snow events) is also important as these anomalies are predicted to increase in response to a warming climate.

How do climate-driven decreases in sea ice impact POP exposure and bioaccumulation in Arctic biota?

Conclusion: The melting of multi-year sea ice, just like the melting of permafrost and glaciers, may release stored contaminants into the Arctic marine food web (see Chapter 2.2). Although this could cause short-term increases in POP levels in biota, the eventual complete replacement of multi-year ice with annual ice, which may be less contaminated due to more frequent freeze-thaw cycles, should lead to reduced POP exposures via this pathway. Research findings available to date show generally, but not consistently, increasing associations between sea ice condition and contaminant levels in biota, likely resulting from the influences of other species-, food web-, and ecosystem-specific factors. Currently, it is difficult to separate the direct effects of declining sea ice cover on POP levels in biota from the indirect effects resulting from changes in food web structure and function, which in turn may impact accumulation of POPs in Arctic biota.

Recommendation: As making robust conclusions by comparing single climate- and ecological-parameters (e.g. sea ice extent, primary productivity, dietary composition) to POP concentrations in biota can be challenging or impossible, on-going studies should strive to obtain both abiotic and biotic parameters to simultaneously compare the strength of their associations with POP levels in Arctic biota. Use of models and other research tools that account for multiple environmental and biotic variables could be useful in determining the contribution of single variables to changes in contaminant levels observed in wildlife.

How does increasing terrestrial runoff affect POP exposure and bioaccumulation in Arctic biota?

Conclusion: Increased terrestrial runoff is predicted to affect both exposure and food web biomagnification of POPs, but to date, there are few data to test or support this prediction. A pilot study from Lake Hazen suggests that increased glacial discharge had no effect on POP trends in Arctic char.

Recommendation: Additional experimental evidence for effects of terrestrial runoff on biota POP levels in aquatic and coastal systems is required. Terrestrial runoff affects nutrients, organic matter quality and quantity, and contaminants in a complex process that influences both food web ecology and the availability and uptake of POPs and CEACs in biota.

How does permafrost degradation and associated increases in freshwater turbidity alter POP exposure and bioaccumulation in Arctic biota?

Conclusion: Recent studies suggest that permafrost degradation and thaw can increase the amount of POPs bound to sedimentary organic matter and increase levels in amphipods in small lakes.

Recommendation: There are knowledge gaps which require additional research to better understand the impacts of changing water quality (i.e. increasing nutrient concentrations and turbidity) on contaminant dynamics in freshwater food webs. These gaps include impacts on fish physiological conditions in lakes and rivers impacted by permafrost RTS, links between water quality and POP trends in fish from Arctic freshwater environments, and changes in water quality due to permafrost degradation and RTS. Additional studies are also needed from a wider range of lakes across the circumpolar Arctic.

What does a shift in the timing of seasons mean for Arctic organisms in terms of POP exposure and bioaccumulation?

Conclusion: Changes in seasonal timing and species phenology, in terms of migration, reproduction, and food availability, may have potential impacts on levels of POPs in Arctic biota. Many of these changes are dependent on species- and ecosystem-specific characteristics and may result in specific windows of elevated vulnerability during the year and during the organisms' life span.

Recommendation: As currently the effects of phenological changes on contaminant bioaccumulation are not known, empirical studies are required to assess whether further investigation of these factors is warranted. The cascading effects of changes in seasonal timing and potential mismatches in Arctic communities are unknown, warranting interdisciplinary collaborations with biologists / ecologists in future contaminant studies. At the local- and regional- levels, Indigenous Knowledge from hunters and fishers may be able to provide useful data in this regard (see Chapter 2.5).

What impact does increasing Arctic primary production have on exposures to POPs in Arctic biota and food webs?

Conclusion: The net effect of increasing primary production on contaminant dynamics in Arctic food webs is unknown as predictions of future primary production and their downstream impacts on dissolved- versus particulate-POP concentrations are uncertain. However, increased primary production in combination with temporal mismatches with grazers could lead to increased sedimentation and contaminant delivery to the benthic ecosystem, although changes in benthic-pelagic coupling due to the northward-range shifts of pelagic fishes may influence this.

Recommendation: Given the potentially counteracting effects of increased primary production on contaminant dynamics (i.e. lower food web exposures via biodilution versus higher food web exposures via increased dietary intake of particulate-bound POPs), additional research on these processes should be undertaken.

How do climate-driven changes in species interactions influence POP exposure in Arctic biota and food webs?

Conclusion: With many unknowns and variable findings from different species and ecosystems, the *net* effect of climate change-driven alterations in species interactions on food web contaminant accumulation is not known. Studies of some polar bear populations indicate that sea ice decline may reduce the dietary importance of marine foods, leading to lower intakes of POPs. A foraging switch from Arctic- to boreal-prey species may lead to higher POP concentrations in predators if their new prey are seasonal migrants that carry POP levels reflective of more contaminated regions at lower latitudes. Climate-driven migrants may therefore act as contaminant biovectors into Arctic marine food webs, although the importance of this phenomenon is not fully understood. For instance, invading Atlantic species also differ in size and physiology, and it is not known if these characteristics will counteract the potentially higher exposure to POPs.

Recommendation: Monitoring of dietary tracers and other ancillary biological and ecological data should be included in biomonitoring programs to track changes and interpret ongoing contaminant datasets. This may assist in evaluating the role of changing species interactions on contaminant levels if/when trends deviate from expected patterns. Adjusting for some of these parameters will aid the interpretation of trend data; for example, controlling for changes in wildlife diets could be helpful in discerning whether declining contaminant levels are the result of changing global emission, or related to climate-driven ecological changes. Modeling future scenarios of shifting food webs and their impacts on POP exposure in top predators should be pursued in subsequent studies once a better understanding of how climate change affects food web structure and function is developed. Without further action on climate change, alterations in the Arctic ecosystem are expected to continue and may impact the effectiveness of policies seeking to reduce POP exposures in Arctic people and wildlife through primary source reductions alone.

In what ways might POP burdens in Arctic biota and food webs be affected by changing lipid dynamics and energy allocation?

Conclusion: Lipid amount, quality, and dynamics within organisms and ecosystems are expected to change with climate change, and thus have potential consequences for contaminants, and lipophilic POPs in particular. Decreased body condition has been related to increased blood levels of POPs in some polar bear and eider duck populations. In many areas, polar bear body condition has declined along with sea ice, which all else being equal, should lead to increased tissue concentrations of lipophilic POPs, and specifically, elevated levels of contaminants within blood. However, temporal changes in diet and body condition did not significantly affect trends of pollutants measured in Barents Sea polar bears.

Recommendation: When feasible, lipid content and body condition metrics should be routinely measured in contaminant trend studies and be incorporated into data interpretations as changing lipid dynamics could introduce variability into POP temporal trends and cross-comparisons between individuals or populations. Investigation of the behavior of proteinophilic contaminants (e.g. PFAS and Hg) with changes in body condition (i.e. lean body mass) may also be warranted.

Do climate-driven changes in animal behavior affect their exposures to POPs and food web bioaccumulation, and if so, how?

Conclusion: Few studies have examined the consequences of behavioral changes on POP exposures. Offshore polar bears from the Barents Sea with a high energetic cost/high energy diet were exposed to higher levels of pollutants than coastal polar bears. Concentrations of lipophilic POPs were, however, similar in both bear types, as offshore bears were fatter than coastal ones. Changes in seabird foraging patterns have potential to impact their POPs levels, but so far, have not been thoroughly examined.

Recommendation: Additional research examining the impact of behavioral changes on POP exposure may be warranted. This should be done by studying relationships between pollutant exposure, movement ecology, diet, food web structure and energetics, and would benefit from multidisciplinary research teams and approaches to data collection. As behavioral and dietary changes may affect contaminant levels in biota, they may also introduce variation into long-term temporal trends that should be identified and accounted for.

How does climate change, in combination with other stressors, affect contaminant toxicity to Arctic biota?

Conclusion: Few studies have examined this topic to date, and there is a general lack of understanding of how climate change and other stressors affect POP toxicity in the Arctic or elsewhere. Studies on polar bears, however, suggest that contaminant exposure and sea ice decline have synergistic adverse effects on lipid metabolism, as biomarker responses between less- and more-polluted bears showed greater contrast during a period with poor sea ice conditions.

Recommendation: An improved understanding of the effects of multiple stressors, inclusive of climate change impacts, on contaminant toxicity in Arctic biota is crucial. Knowledge of the effects of contaminants on the abundance, development and health of wildlife populations, and the mechanisms and processes underlying these effects, is needed for toxicants individually and in combination with other stressors (i.e. climate change, pathogen and parasite exposure, etc.), as effect thresholds under these more realistic scenarios may be different from those based on exposure to a single contaminant or stressor in isolation. Protecting Arctic biota and people from the effects of POPs requires that contaminant researchers expand their scope of research and collaboration to consider POP exposures and effects within the true context of many co-occurring and potentially interacting anthropogenic stressors facing populations, communities, and ecosystems in the Arctic.

What can be learned from models of climate change effects on food web accumulation of POPs?

Conclusion: Models are important tools for evaluating the net result of contrasting processes, as climate change can lead to both increased and decreased exposures and enhanced or reduced bioaccumulation processes, depending on the processes, areas, chemicals and species interactions affected. Mechanistic process-oriented models combining physicochemical transport models with bioenergetic and food web models, can help to understand how the contribution and variation of different

parameters, such emission sources, transport processes, temperature, lipid dynamics, and food web structure, affect exposure, uptake, elimination and overall bioaccumulation of contaminants. Nonetheless, for some of these parameters (e.g. food web changes), there remains insufficient understanding or empirical data to populate models.

Recommendation: Models combining climate change impacts on abiotic and biotic processes affecting POPs and CEACs should be generated and evaluated with measured data. In some cases, and related to other recommendations, additional research is required to constrain model parameters. In addition to the need for more information on processes and physicochemical parameters tied to chemical emissions, transport and partitioning, more ecological realism, inclusive of variation, is needed for parameters related to ecosystem structure and functioning, and especially for those that may affect bioaccumulation and magnification.

2.3.7.2 Perspectives on future research and monitoring

Temporal trends of contaminants are used by the Global Monitoring Plan as a measure of the effectiveness of the Stockholm Convention on Persistent Organic Pollutants. In addition to global regulations, variations in climate and trophic interactions can also influence contaminant temporal trends. Therefore, to inform future regulations and policies, it will be increasingly important to identify the underlying source(s) of environmental contaminant changes.

This review has demonstrated that there is vast diversity of direct and indirect mechanisms by which climate change can influence contaminant exposure, accumulation, and effects in ecosystems, and that ecosystem responses might vary substantially as well. This means that a broad range of habitats, species, and processes must be considered for a thorough understanding and interpretation of resulting consequences for the distribution, accumulation, and effects of environmental contaminants.

Given the complex interactions between climate change, contaminants, and ecosystems, it is important to plan for long-term, integrated pan-Arctic monitoring of key biota and ecosystems, and to collect ancillary data, including information on climate-related parameters, local meteorology, ecology, and physiology, in addition to contaminant data. To be operative, this requires a stable institutional background and the ability to integrate short-term, local efforts and resources into a coherent service.

Although biological and ecological diversity should be reflected in research, it is essential that studies are also designed in a way to ensure they also possess adequate statistical power in addition to data richness.

We recommend that future research identifies and characterizes the direct linkages between climate-induced changes in LRT of contaminants to the Arctic, and the exposure, accumulation, and toxicity of POPs and metals to wildlife. It will be challenging to separately identify the effects of climate change and LRT on Arctic wildlife that are also forced to cope with many other stressors that also influence the accumulation and toxicity of these chemicals and metals. However, forward progress may be made with comprehensive monitoring of Arctic ecosystems and wildlife in combination with lab-based mechanistic studies, new research methods and models that reflect the complexity of natural variation and species interactions, and risk assessments that holistically consider the effects of multiple exposures and stressors on individual and population health.

Changes in the structure and function of Arctic ecosystems are not currently predictable. To project the effects of climate-driven changes in food webs on POPs in Arctic wildlife, we recommend that the contaminants community continue to work closely with other research disciplines.

2.4 Associations between climate change and temporal trends of contaminants in Arctic biota

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2.4.1 Introduction

In response to increased awareness of contaminants in the Arctic, several Arctic countries established national monitoring programmes for persistent organic pollutants (POPs) in the region's biota in the 1990s. Some of these programmes have now been in operation for more than 20 years and have generated long-term time series data of POP concentrations for selected Arctic species. These temporal datasets can serve multiple purposes. Long-term trends can be used to evaluate the effectiveness of global and regional regulations intended to decrease concentrations of certain contaminants in the environment. Similarly, time series data can also be used to detect increasing trends of unregulated compounds in the Arctic environment and provide support for new regulatory actions. Finally, long-term monitoring data can be included in research-oriented studies on the long-range transport (LRT) and accumulation of POPs in the Arctic or used to advance the understanding of exposure levels for specific animals in the context of environmental health studies. As many Arctic species are part of indigenous peoples' traditional diets, monitoring of contaminant levels in biota provides information important for food security and contaminant exposure in humans.

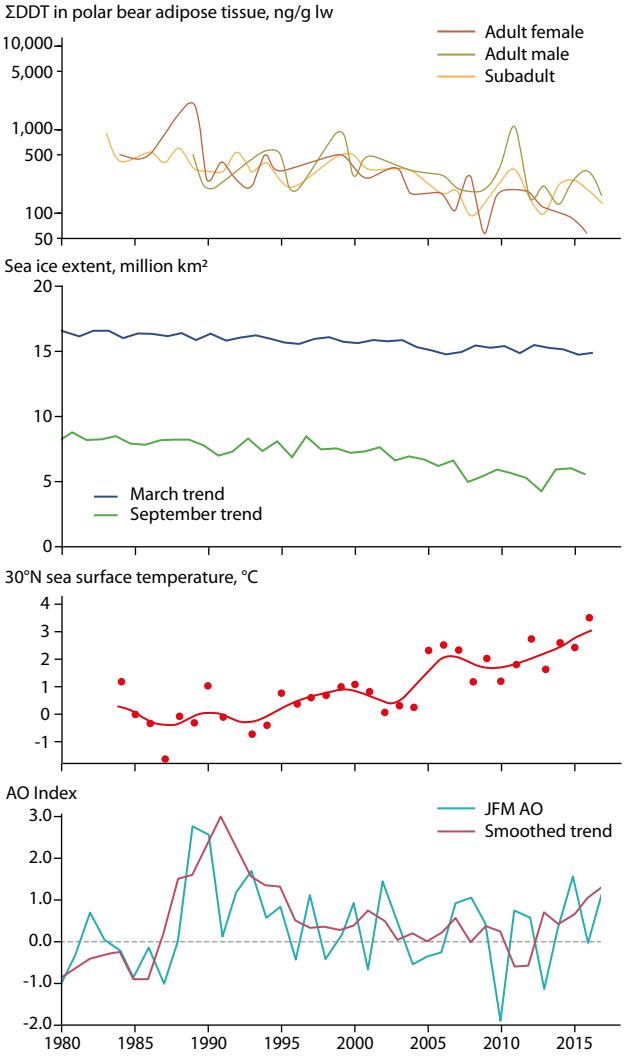


Figure 2.4.1 Examples of temporal changes of contaminants and climate parameters. (top panel) ΣDDTs (sum *p,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD) in East Greenland polar bears. Updated from Dietz et al., 2013. (second panel) Sea ice extent in the Greenland Sea. Source: National Snow and Ice Data Center (NSIDC), USA, https://nsidc.org/data/G02135/versions/3. (third panel) Sea surface temperature north of 80°N. Source: Danish Meteorological Institute (DMI), Denmark. http://ocean.dmi.dk/arctic/meant80n_anomaly.uk.php. (fourth panel) Arctic Oscillation (AO) index. Source: National Weather Service, National Oceanic and Atmospheric Administration (NOAA), USA, https://www.cpc.ncep.noaa.gov/products/precip/CWlink/daily_ao_index/ao_index.html.

The use of contaminant time series data to inform chemical management decisions is based on the underlying premise that concentrations in biota are mainly determined by the amount of the compound released during production and use, and therefore, can be controlled through regulations. Thus, increasing or decreasing concentrations of contaminants in fish and wildlife tissues are mainly assumed to reflect changes in their emissions. This relationship generally holds true; for example, a rapid decrease in perfluorooctane sulfonic acid (PFOS) concentrations was observed in Arctic biota following the discontinuation of PFOS production (Butt et al., 2007; Rigét et al., 2013b). However, contaminant levels in biota also integrate changes in the physical environment and ecosystems (see Chapters 2.2 and 2.3). For example, changes in environmental transport processes due to meteorological conditions, increases in secondary emission sources, and changes in predator/prey relationships can affect the contaminant exposure experienced by a target species. Thus, time series of POPs in Arctic biota could also be influenced by the direct and indirect environmental changes caused by a warming climate (Ma et al., 2016).

All time series for POPs in Arctic biota were assessed in a recent AMAP report (AMAP, 2016b; Rigét et al., 2019). The report generally found that temporal trends differed between POPs depending on the date of their regulation. Those POPs that were subject to regulation by national initiatives in the 1970s–1990s, such as polychlorinated biphenyls (PCBs) or the insecticide dichlorodiphenyltrichloroethane (DDT), showed decreasing trends, generally beginning prior to the implementation of the 2004 Stockholm Convention on POPs, as exemplified for ΣDDTs in East Greenland polar bears (Figure 2.4.1). Compounds that came under regulation

later, such as the polybrominated diphenyl ethers (PBDEs) or PFOS, which were added to the Stockholm Convention in 2009, showed mixed trends, including shifts from increasing to decreasing concentrations. The physical-chemical properties of PFOS, which differ from those of legacy POPs, might further influence the PFOS time trend. A few time series for chemicals under regulation, including hexachlorobenzene (HCB) and β-hexachlorocyclohexane (HCH), were still increasing in concentration, possibly indicating local sources or unintentional production of these contaminants.

In addition to the trend data collected by national monitoring programmes, time series have also been derived from Arctic and Antarctic sample collections in Environmental Specimen Banks (ESBs), such as the Biorepository of the National Institute of Standards and Technology (NIST) (Becker et al., 1993; Becker and Wise, 2006) or the ESB at the Swedish Museum of Natural History (Odsjö, 2006). For example, the analysis of northern fur seal (*Callorhinus ursinus*) blubber samples collected from Alaska between 1987–2007 and stored in the NIST Biorepository showed that concentrations of PCBs, DDT, and other legacy POPs decreased over the 20 year time period examined, while concentrations of PBDEs and perfluorinated alkylated acids increased (Reiner et al., 2016). ESBs in other countries have banked Arctic and Antarctic samples as well, and polar monitoring programmes often include sample collections that have been used for retrospective time trends.

According to Macdonald (2005), the largest barrier to studies investigating the influence of climate on POP temporal trends is the limited availability of consistent, long-term monitoring data for pollutants. The monitoring of contaminants in Arctic biota that has taken place over years and decades enables such investigations. This chapter reviews and assesses studies from different locations in the Arctic that have examined associations between POP concentrations and climate parameters (Table A2.4.1). Physical parameters affected by climate change include, but are not limited to, temperature, sea ice extent or thickness, water salinity, pH, and climate oscillation indices, such as the North Atlantic Oscillation (NAO) or Arctic Oscillation (AO) indices, that reflect large-scale regional climate patterns, including fluctuations in atmospheric pressure gradients, as explained in detail in Box 1.1 of Chapter 1. Examples of some of these parameters are shown in Figure 2.4.1.

Changes in the physical environment can have direct effects on contaminant availability (see Chapter 2.2), and climate change can affect ecosystem structures, with indirect consequences on contaminant exposure (see Chapter 2.3). Both of these factors are reflected in POP time series in biota. For example, increased ocean temperature can affect species distributions (Kortsch et al., 2012; Renaud et al., 2012), and receding sea ice can shrink the living space of diverse pagophilic species, spanning invertebrates to marine mammals (Gross, 2005; Kovacs et al., 2011). If predator/prey relations change, contaminant exposure can change as well. These relationships are usually assessed by the analysis of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotopes, which are indicative of diet sources and trophic level.

This chapter will thus address the overall question: Can we link changes in temporal trends of POPs with climate parameters and/or food web changes? These associations are often based on correlational analyses, which do not necessarily reflect causal relationships between variables, and identifying the

Box 2.4.1 Statistical approaches to assess climate-related influences on POP temporal trends

Time series of POP data from the Arctic now span several decades. The availability of long-term datasets enables relationships between climate-related and POP time series to be examined, including associations between interannual variations in climate indicators and changes in POP concentrations. A summary of the statistical and model approaches that may be used for these analyses is given by Ma et al. (2016).

Climate indicators can reflect local (e.g. sea ice extent) or regional (e.g. AO, NAO, PNA) changes, therefore the selection of climate parameters for an analysis depends on their relevance to the hypothesis under study. Additionally, in some cases, changes in climate may not have an immediate influence on POP concentrations in the Arctic biota, and may only be observable after months or years (e.g. effects of climate on food web structure and POP bioaccumulation), thus analyses may need to consider the potential for a time lag between changes in climate indicators and POP concentrations. In studies of POP trends in Arctic wildlife, biological variables should be included as control variables to minimize the effects of variation related to animal age, sex, and diet (e.g. stable isotopes or fatty acid signatures). It may be necessary to transform biological data to meet normality and variance criteria of the statistical models applied.

One approach to studying links between POP concentrations in biota and climate indicators is to apply plausible linear models to time series data and use a model selection criterion to find the most parsimonous model(s) (Burnham and Anderson, 2002). Akaike's Information Criterion corrected for small sample size (AICc) is one selection criterion that can be used to identify the best fit model for a dataset. In this approach, models are ranked according to their AICc and the model with lowest AICc value is considered the most parsimonious. As a rule of thumb, there is only weak or no evidence for selecting between the models within a value of two from the most parsimonious AICc value. In this case, model averaging can be applied. Using this approach, an AICc weight is calculated for each model as the relative likelihood divided by the sum of all model likelihoods. AICc weights can then be used for calculating the importance of all the variables included in one or more of the models, as well as a weighted average of the parameter estimates for the variables included (see Burnham and Anderson (2002) for detailed calculation procedures). The R software (R Development Core Team, 2019) library *MuMIn* can be used for model selection and model inference, and the *car* library can be used for component-residual plots. These are useful for evaluating the effect of each variable in a model when the other variables are accounted for.

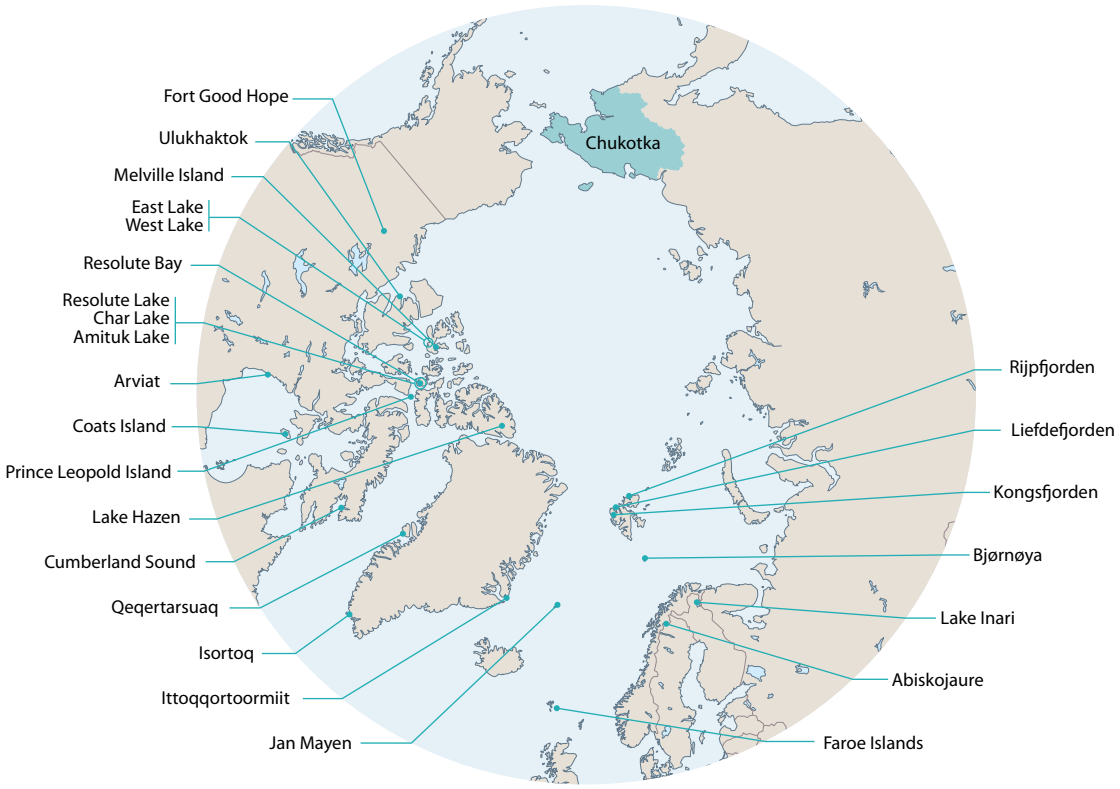


Figure 2.4.2 Arctic locations referenced in this chapter.

mechanisms underlying such relationships is often part of ongoing research. Limitations and perspectives such as these are included in the assessment as far as this information was available to the authors. In relation to the use of contaminant temporal trends to inform chemical regulations such as the Stockholm Convention, this chapter will also attempt to answer the question: Do the findings of this chapter have implications for national and international regulations of chemicals?

This chapter focuses on associations between climate parameters and POP temporal trends in Arctic biota. Brief descriptions of existing POP time trends in biota will be given for each of the Arctic countries, however, the contaminant trends themselves are not the focus of this assessment. A summary of the studies addressing links between time trends of POPs in Arctic biota and climate change is provided in Table A2.4.1. Examples of statistical approaches used to analyze associations between POP concentrations in Arctic biota and climate parameters are given in Box 2.4.1. Figure 2.4.2 shows the locations of Arctic sites mentioned in this chapter. This chapter will also include relevant information on climate-related effects on POP time trends in Antarctic species to complement findings from the Arctic.

2.4.2 Links between POP time trends and climate change in North America

2.4.2.1 United States (Alaska)

POP monitoring and time series

Since 1999, the Seabird Tissue Archival and Monitoring Project (STAMP) has collected and banked seabird eggs from Alaska including thick-billed murre (*Uria lomvia*) and common murre (*Uria aalge*) eggs, and starting in 2004, glaucous gull (*Larus hyperboreus*) and glaucous-winged gull (*Larus glaucescens*) eggs as well (AMAP, 2016b). The project has been a collaboration between the U.S. Fish and Wildlife Service Alaska Maritime National Wildlife Refuge, the U.S. Geological Survey Biological Resources Division, and NIST. The egg contents are banked at the NIST Biorepository.

Terrestrial environment

There are currently no known studies of associations between climate parameters and POP temporal trends in Arctic terrestrial biota available from the United States.

Freshwater environment

There are currently no known studies of associations between climate parameters and POP temporal trends in Arctic freshwater biota available from the United States.

Marine environment

Contaminant data for thick-billed and common murres from STAMP suggest complex relationships exist between the Pacific Decadal Oscillation (PDO) and POP concentrations in seabird eggs collected from coastal areas of the Bering Sea and the Gulf of Alaska between 1999–2010 (Kalia et al., 2021). The influences of climate variability on POP levels are possibly related to shifts in bird diet. In thick-billed murre eggs, the highest levels of PCB congeners and chlorinated pesticides were observed when the PDO index was approximately zero. Lower levels were observed when the PDO index was at an extreme positive (i.e. warm phase) or extreme negative (i.e. cool phase) value, corresponding to the patterns of $\delta^{13}C$ and $\delta^{15}N$ values in

the thick-billed murre eggs. In contrast, in sympatric common murres there were weak or null associations between the PDO index value and POP levels in eggs. Changes in sea surface temperature are associated with altered primary production and the biochemistry of phytoplankton (Bermudez et al., 2015; Racault et al., 2017). These changes could be propagated through the food web and affect higher trophic level animals (Walther, 2010), as discussed in Chapter 2.3. The different patterns of association observed between the PDO index and POP levels for the two bird species might reflect foraging differences between the species; common murres tend to feed in the meso-pelagic zone closer to the colony, while thick-billed murres dive deeper and forage further from shore (Gaston and Hipfner, 2020; Ainley et al., 2020).

2.4.2.2 Canada

POP monitoring and time series

In Canada, POPs are monitored annually in Arctic biota as an index of contamination of Arctic ecosystems. This monitoring is coordinated and funded mainly through the Northern Contaminants Program (NCP). Since 1991, data generated under the NCP have been used to monitor contaminant concentrations in traditionally harvested foods and provide information on the state of the Arctic environment, both of which are used to inform decision-making by regulatory agencies, individuals, and indigenous communities relying on country foods for subsistence. Monitoring efforts mostly target freshwater and marine species, but data collection from terrestrial animals (e.g. reindeer) is also ongoing.

Several POP time series from Canadian Arctic biota have been published in recent years. Trends of legacy compounds, flame retardants (FRs), polychlorinated naphthalenes (PCNs) and per- and polyfluoroalkyl substances (PFAS) have been reported in seabird eggs (Braune and Letcher, 2013; Braune et al., 2015b; Braune and Mallory, 2017; Braune and Muir, 2017, Braune et al., 2019). Additional trends of legacy compounds, FRs and/or PFAS have been published for ringed seals (*Pusa hispida*), beluga whales (*Delphinapterus leucas*) and polar bears (*Ursus maritimus*) (Addison et al., 2014; Houde et al., 2017a; Brown et al., 2018; Letcher et al., 2018; Noël et al., 2018; Smythe et al., 2018). Trends of contaminants in terrestrial biota such as moose (*Alces alces*), reindeer (*Rangifer tarandus*), and Arctic fox (*Vulpes lagopus*) have also been summarized (Gamberg et al., 2005). Moreover, time trends of current-use pesticides, FRs and PFAS in biota from multiple Arctic locations, including Canada, have been recently reviewed (Balmer et al., 2019b; Muir et al., 2019; Vorkamp et al., 2019). The general declining trends of legacy POPs in biota have been attributed by many authors to the past national and international restrictions on uses and emissions of legacy POPs in circumpolar and neighboring countries.

Terrestrial environment

There are currently no known studies of associations between climate parameters and POP temporal trends in Arctic terrestrial biota available from Canada.

Freshwater environment

Recent acceleration of glacier retreat, sea ice loss and thinning, and permafrost thaw and degradation due to climate warming may have an impact on the temporal trends of POPs in freshwater lakes. In remote Arctic lakes, POPs enter the environment primarily through river runoff and atmospheric deposition, including both wet and dry deposition. Over the last century, legacy pollutants have accumulated in lake sediments and catchment areas, both of which contain large amounts of organic carbon, a major reservoir for POPs. Since warming favors POP partitioning from particle- and liquid-phases to the gas phase, it is suggested that under global warming conditions, POPs would have a greater tendency to volatilize from lake water into air (Ma et al., 2016). Changes in air-water exchange will in turn alter the mass balance in water-sediment exchange (Ma et al., 2016). These processes associated with permafrost-related disturbances may contribute to inputs of POPs to the water column, making POPs more available for fish, although the magnitude of this process is still unknown.

Potential associations between increasing PCB and DDT concentrations in burbot (*Lota lota*) and increased primary productivity in the Mackenzie River (Fort Good Hope, Northwest Territories) linked to warming temperatures were hypothesized by Carrie et al. (2010). They found two-fold increases in concentrations of total hexachlorinated PCBs and three-fold increases in ΣDDTs in burbot liver over the period 2000–2008. As increased algal primary productivity was observed over this time period, Carrie et al. (2010) suggested that increasing POPs in burbot could be related to increased primary productivity due to climate warming, resulting in higher concentrations of organic matter, and thus potentially higher contaminant availability to the fish. However, subsequent annual sampling at the same site showed declining concentrations of ΣPCBs (two-fold) and ΣDDTs (one-and-a-half-fold) in burbot from 2008–2012 (Stern et al., 2014).

Moreover, associations between climate parameters and temporal trends of POPs in Arctic char (*Salvelinus alpinus*) were investigated at four Canadian High Arctic lakes (Amituk, Hazen, Char and Resolute) using data collected over 12 to 17 years (Cabrerizo et al., 2018b). A multiple regression approach was used to study the relationship between temporal trends of legacy POPs in fish and other variables, including sampling year, fish weight, chlorophyll a concentrations (as a proxy for primary productivity), and climate parameters such as temperature, annual precipitation (rain and snow) amounts, and interannual atmospheric climate fluctuations, such as those represented by the NAO index. Temperature was found not to be a significant influence on Arctic char POP concentrations, likely because ambient air temperatures, rather than lake water temperatures, were used in the analysis, and air temperatures fluctuate considerably more than profundal lake water temperatures at the depths where Arctic char are primarily found. Significant negative correlations were detected between primary productivity and PCB concentrations in the Arctic char of Amituk Lake and Resolute Lake, suggesting a dilution effect may be occurring. This is in contrast to the findings for burbot in the Mackenzie River, where positive associations between POPs and primary production were suggested (Carrie et al., 2010).

Relationships between the NAO index of the preceding spring/summer and concentrations of ΣPCBs and ΣDDTs in landlocked char from Lake Hazen in northern Ellesmere

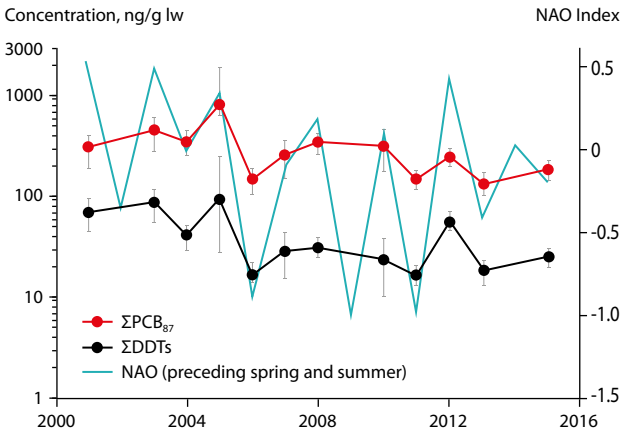


Figure 2.4.3 Trends of ΣPCBs and ΣDDTs in landlocked Arctic char from Lake Hazen (Northern Ellesmere Island, Canada), and the North Atlantic Oscillation (NAO) index from spring of preceding year. ΣPCBs represents the sum of 87 congeners; ΣDDTs represents the sum of *p,p'*- and *o,p'*-DDT isomers. Redrawn from Cabrerizo et al., 2018b.

Island are shown in Figure 2.4.3 (Cabrerizo et al., 2018b). Concentrations of ΣPCBs, ΣDDTs, and ΣHCHs in Arctic char were positively associated with interannual variations of the NAO. Furthermore, concentrations of ΣHCHs in Arctic char were positively correlated with annual precipitation suggesting the importance of wet deposition pathways in delivering HCHs to the Arctic lakes. The inclusion of climate parameters enhanced the explained variability of POP temporal trends in fish by up to 57% in comparison to regression models that did not include the same suite of parameters (Cabrerizo et al., 2018b), adding to the evidence that climate change may be influencing POP levels in freshwater fish.

In a recent study of two lakes, East Lake and West Lake on Melville Island in the Canadian High Arctic, the temporal trends of PCBs, legacy organochlorine pesticides and PBDEs were investigated in zooplankton, landlocked Arctic char muscle and char stomach contents (see Box 2.3.1 of Chapter 2.3). Compared to East Lake, West Lake is receiving greater inputs of terrestrial carbon from thawing permafrost and has higher turbidity as a consequence of the subaqueous slumps that have formed (Cabrerizo et al., 2019a; see Chapter 2.2). Greater particulate inputs from the West Lake catchment and subaqueous slumps have led to higher particulate-associated contaminant concentrations in the lake. Correspondingly, higher lipid-based concentrations of PCBs, organochlorine pesticides (e.g. DDT, chlordanes) and PBDEs have been observed in char muscle and stomach contents from West Lake, and these concentrations have increased over time (Figure 2.4.4). The higher concentrations in suspended particulates in West Lake have also led to higher PCB concentrations in zooplankton in addition to landlocked Arctic char (Cabrerizo et al., 2019a, 2019b; Muir, unpubl. data).

The effect of lakeshore permafrost thaw and slumping on POP concentrations in freshwater biota was also studied in lakes of the Mackenzie River Delta Uplands in the Northwest Territories of Canada (D'Onofrio, 2014). D'Onofrio (2014) found that amphipods (*Gammarus* sp.) in lakes affected by slumps had higher concentrations of PCBs and organochlorine pesticides than those from lakes not affected by slumping.

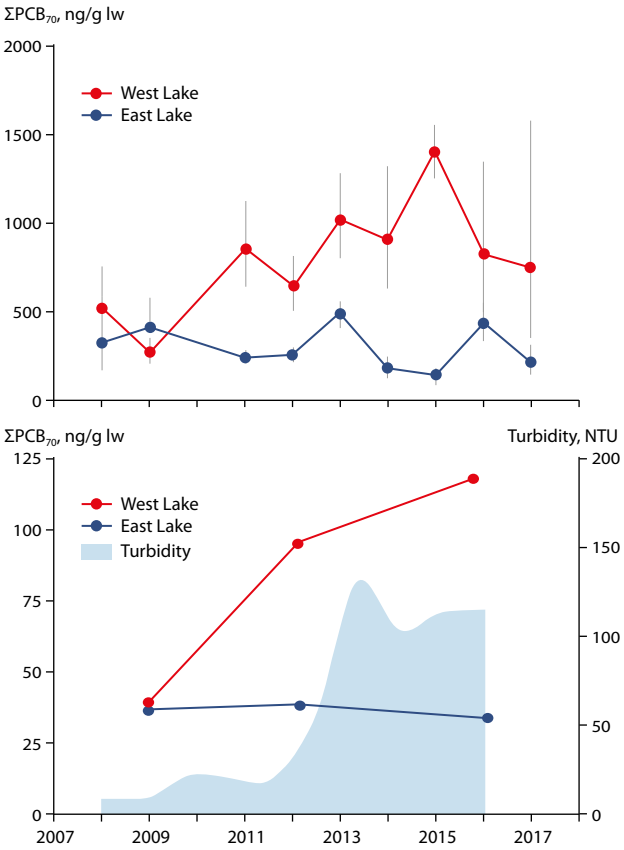


Figure 2.4.4 Long-term trends of ΣPCB concentrations in (upper) muscle and (lower) pooled stomach contents of landlocked Arctic char from East Lake and West Lake on Melville Island, Canada. Turbidity of West Lake is also shown in the lower panel. ΣPCBs represents the sum of 70 congeners. Sources: Cabrerizo et al., 2019b and Muir, unpubl. data.

Additionally, POP concentrations in amphipods were positively correlated with the percentage of the catchment slumped. While amphipods from lakes impacted by slumps generally had higher mean concentrations of ΣPCBs, ΣDDTs and ΣHCHs than those from undisturbed lakes, the differences for ΣHCHs were smaller than for the more hydrophobic POPs.

The studies by Cabrerizo et al. (2018b, 2019a, 2019b) and D'Onofrio (2014) all found higher concentrations of the more hydrophobic POPs (e.g. PCBs, DDT) in biota from freshwater systems impacted by slumping and permafrost thaw. However, the conditions differed greatly between the lakes under study. In the case of the Mackenzie River Delta Upland lakes, the slump-disturbed lakes were more oligotrophic, with reduced levels of dissolved and particulate organic carbon, while West Lake on Melville Island had greater turbidity and elevated levels of particulate matter. Similarly, Carrie et al. (2010) also pointed to an association between higher amounts of particulate organic carbon and enhanced bioavailability of POPs. All of these studies suggest that climate-related effects in Arctic freshwater environments have the potential to enhance POP bioaccumulation, but through different pathways. These mechanisms are not entirely clear, but include downstream effects leading to elevated POP concentrations in lake sediments and increased exposure of benthic organisms to POPs. The role of increased primary productivity is unclear as potential associations with both increased and decreased bioavailabilty have been discussed.

Marine environment

Seabirds

Changes in trophic position have been shown to influence the rate of exposure and uptake of contaminants in thick-billed murres breeding in the Canadian Arctic (Braune et al., 2015a). In murre eggs collected from Coats Island, a Northern Hudson Bay colony, between 1993 and 2013, positive relationships between organochlorine chemicals (i.e. ΣPCBs, *p,p′*-DDE, HCB, oxychlordane, dieldrin and heptachlor epoxide) and $\delta^{15}N$ were detected. As the birds changed their diet from Arctic cod (*Arctogadus glacialis*) to capelin (*Mallotus villosus*), resulting in lower $\delta^{15}N$ values, concentrations of all organochlorines concurrently declined due to lower exposure levels. On the other hand, an increase in the trophic position of murres at the high Arctic colony of Prince Leopold Island between 1975 and 2013 from predominantly feeding on Arctic cod, was negatively associated with key organochlorine compounds such as *p,p′*-DDE and ΣPCBs. This finding was attributed to large reductions in emissions during the 1970s and 1980s having a stronger influence on organochlorine temporal trends in murre eggs than changes in diet (Braune et al., 2015a).

Legacy POPs were examined in eggs of northern fulmars (*Fulmarus glacialis*) and thick-billed murres from Prince Leopold Island (1975–2014) to determine whether or not climate parameters had any discernible effects on temporal trends (Foster et al., 2019). The majority of variability in the data, particularly for the legacy organochlorines, was found to be related to changing emission patterns. However, correlations between contaminants and the NAO index were found for fulmar eggs, and between POPs and rainfall amounts for murre eggs, after a time-lag was accounted for. The data suggested that years with NAO+ conditions were followed by increased concentrations of dieldrin, chlorobenzenes, *cis-* and *trans-*nonachlor and mirex in fulmar eggs. In addition, years with increased rainfall were followed by higher concentrations of dieldrin, chlorobenzenes, octachlorostyrene, DDE and most PCBs, and decreased levels of oxychlordane in murre eggs (Foster et al., 2019).

More recently, Morris et al. (2021) reported on the comparative temporal trends of PCB-153, PBDE-47, *p,p′*-DDE, α-HCH and PFOS in polar bears and thick-billed murre eggs from Coats Island in Northern Hudson Bay, as well as relationships of the contaminants with climate parameters (Table 2.4.2). The relationships between annual mean POP concentrations, sampling year, and annual and seasonal climate parameters were studied using general linear models (GLMs). The concentrations of POPs in murre eggs were normalized to the mean $\delta^{15}N$ prior to modeling with climate factors to control for dietary differences. A number of climate parameters were significantly related to POP concentrations with or without year as a covariate (Table 2.4.2). Most of the multivariate models resulted in relatively subtle adjustments to the annual trends observed using simple linear regressions relating POP concentrations and sampling year, although the multivariate models were stronger and more explanatory. Models that incorporated sampling year with time-lagged AO or NAO index values were consistently among the best ranked for predicting contaminant concentrations in murre eggs. Generally, concentrations of POPs were higher in murre eggs when annually-compiled, time-lagged (1–3 years relative to sampling years) AO or NAO index values were also greater. On the other hand, there were a number of different seasonal relationships with oscillation indices that indicated that some POP concentrations decreased in murre eggs when one-year time-lagged June AO or summer NAO index values were greater.

More sea ice (i.e. greater seasonal coverage or earlier freeze-up dates) was related to higher concentrations of legacy POPs in thick-billed murre eggs (Table 2.4.2) (Morris et al., 2021). The study also showed that greater seasonal and time-lagged air and land surface temperatures were most often related to decreasing concentrations of legacy POPs and PBDE-47 in murre eggs. Higher concentrations of POPs in murre eggs were also related to greater time-lagged, seasonal wind speeds. However, inverse relationships were also observed depending on the season, but without wind direction information these results are difficult to interpret. Increased precipitation levels can result in increased atmospheric scavenging and deposition of contaminants (Macdonald et al., 2005), and this was consistent with observations for *p,p′*-DDE in murre eggs (Morris et al., 2021).

Ringed seals

Ice coverage data was integrated in a long-term study (1993-2008) of organochlorine contaminants in male ringed seals from Ulukhaktok in western Canada (Gaden et al., 2012). In that study, higher levels of *p,p′*-DDE and PCB-153 were reported in seal blubber during years with early ice breakup. Gaden et al. (2012) suggested that this association could be related to increased foraging of seals during years with longer ice-free conditions. In general, changes in environmental conditions may affect the feeding ecology, and consequently the levels of POPs in marine biota, as discussed in Chapter 2.3.

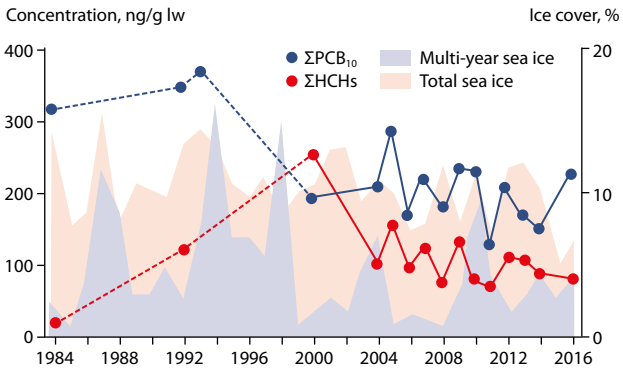
In another long-term study (1972–2016) investigating POPs in ringed seals and associations with climate-related variables in the Canadian Arctic, Houde et al. (2019) found site- and contaminant-specific relationships between concentrations in seal blubber, climatic patterns, and ice coverage. In Resolute Bay (Nunavut), positive correlations were observed between seal POP levels and the AO condition for the year preceding seal sampling. Relationships were opposite (i.e. negative) when using AO information for the year of sampling, and when using NAO data for this high Arctic site. Correlations were also reported between POP concentrations and the Pacific/North American pattern (PNA). In the Hudson Bay area (i.e. Arviat), mainly positive associations were found between levels of POPs (i.e. ΣPCBs, ΣDDTs, ΣHCHs and Σchlordanes) in seal blubber and the AO, NAO, and PNA indices for the year of sampling or the preceding one.

Results linking seal POP concentrations and ice coverage were dependent on the different types of ice analyzed and modeled (Houde et al., 2019). In Resolute Bay, Arviat and the Beaufort Sea seals, several POP concentrations were found to be positively correlated with total sea ice coverage, suggesting an increased accumulation of contaminants in years with greater ice extent (Figure 2.4.5). The type of ice (new vs. old ice) was also differently associated with POP accumulation in seals as observed for concentrations of ΣPCBs, ΣDDTs, ΣHCHs and Σchlordanes in seals from Resolute Bay and the Beaufort Sea, most of which were positively associated with multi-year ice coverage, and generally negatively correlated with new ice coverage.

Table 2.4.2 Relationships of POP concentrations (ng/g wet weight) in eggs of thick-billed murres (Coats Island, Nunavut) and tissues of polar bears from the Western Hudson Bay (WHB) and Southern Hudson Bay (SHB) subpopulations (Morris et al., 2021) with sampling year and climate-related parameters as determined using general linear models (GLMs). Arrows indicate the direction of the concentration trend with increasing values of the variable, and the numbers in parentheses indicate the time-lag in years (if no number is shown, there was no time-lag applied). Commas separate different seasonal or time-lagged models, with the variables compiled annually (black), in winter (blue), spring (green), summer (red) and fall (orange) or in June (purple, murres only). Two arrows side by side represent significant multivariate models with the first arrow representing the trend with year and the second arrow representing the trend with the climate variable of interest. Temporal trends are shown as '↔' when the rate of change was ±1% per year. Temporal trends marked with asterisks (*) were statistically significant, and only significant results are shown for climate/weather variables. If no arrows are shown (--) there were no significant climate/weather models with AIC_C values stronger than year alone detected.

Compound	Parameter(s)	Thick-billed murres[a]	Polar bears (WHB)[b]	Polar bears (SHB)[c]
α-HCH	Year only	↓*	↓*	↓
	Air/land surface temperatures (˚C)	↓(1),↓(1),↑(3)	--	↑(1)
	Wind speeds (km/h)	↓(3),(3)	↓	↓(3), ↑(2)
	Arctic Oscillation	↓↓(1)	↓(3)	↓↓(2)
	North Atlantic Oscillation	--	--	↓↓(2),↓↓(2)
	Sea ice coverage (TAC)[d]	↑(1)	--	↓,↓,↓
	Ordinal date of sea ice break-up[d]	--	--	↓
	Ordinal date of sea ice freeze-up[d]	↓(1)	--	↑(1)
	Number of sea ice-free days[d]	↓(1)	--	--
p,p'-DDE	Year only	↓*	↓*	↔
	Air/land surface temperatures (°C)	↓↓(1)	--	--
	Wind speeds (km/h)	--	--	↑(2), ↑(2)
	Arctic Oscillation	--	↓(3),↓(3),↓(1),↓↓(2),↓(3)	↑(2),↓(2)
	North Atlantic Oscillation	↓(2)	↓(3)	↓, ↑(2),↓(2),↓(2)
	Sea ice coverage (TAC)[d]	--	↑(2)	↓
	Ordinal date of sea ice break-up[d]	--	--	↓
	Ordinal date of sea ice freeze-up[d]	--	↓↓(2)	--
	Number of sea ice-free days[d]	--	↓↓(2),↓(3)	--
PCB-153	Year only	↓*	↑	↓
	Air/land surface temperatures (°C)	--	↑(2)	↓(2),↓(2),↓↓(2),↓(3)
	Arctic Oscillation	↓↑(2)	--	↓(1),↓(1)
	Precipitation (mm)	--	--	↓(3)
	Sea level pressure (hPa)	↓↓(2), ↓↓(3)	--	↑,↓↓
	Sea ice coverage (TAC)[d]	--	--	↑(2)
	Ordinal date of sea ice freeze-up[d]	↓↓(1)	--	--
PBDE-47	Year only	↓*	↑*	↔
	Air/land surface temperatures (°C)	--	--	↓(2),↓
	Wind speeds (km/h)	↓↓,↓↑(1)	--	--
	Arctic Oscillation	↓	--	--
	North Atlantic Oscillation	↓↓	↑↓	↓(2),↓(3)
	Precipitation (mm)	--	↑↓(1),↑↓(2)	↓(2)
	Sea level pressure (hPa)	--	↑↑(2)	↑(2),↓(1)
	Ordinal date of sea ice freeze-up[d]	--	↑↑	↑(1)
PFOS	Year only	nm	↑	↓
	Wind speeds (km/h)	nm	↑,↑(1)	↓,↑(1)
	Arctic Oscillation	nm	↑(1)	↓(1)
	North Atlantic Oscillation	nm	↑	↓
	Sea level pressure (hPa)	nm	--	↑
	Ordinal date of sea ice freeze-up[d]	nm	--	--

TAC: total accumulation coverage (proportion of a defined area with sea ice cover); AIC_c: Akaike's Information Criterion corrected for small sample size; nm: not measured; [a]Murre egg samples were collected from 1993 to 2015; [b]Southern Hudson Bay (SHB) samples were analyzed from 2007 to 2016 (fat, legacy POPs) or from 2008 to 2016 (liver, PFOS); [c]Western Hudson Bay (WHB) samples were analyzed from 1991 to 2015 (fat, legacy POPs) or from 2007 to 2015 (liver, PFOS); [d] Sea ice variables were compiled in the central or north western Hudson Bay for WHB bears, in the central or eastern Hudson Bay for SHB bears, and in the North Hudson Bay Narrows for murres.



	Total sea ice	Multi-year sea ice	AO preceding year
ΣPCB_{10}	-	0.26	0.27
ΣHCHs	-	0.50	0.28
ΣCHL	-	0.26	0.25
ΣCBz	0.36	0.52	0.45

Figure 2.4.5 Trends of ΣPCB and ΣHCH concentrations in ringed seal blubber (Resolute Bay, Western Lancaster Sound, Canada) in relation to total sea ice and multi-year ice coverage. The associated table provides correlation coefficients for relationships between POP concentrations, percent sea ice coverage and the Arctic Oscillation (AO) index value of the preceding year. ΣPCBs represents the sum of ten congeners; ΣHCHs represents the sum of α-, β- and γ-HCH; ΣCHL represents the sum of chlordanes; ΣCBz represents the sum of 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene, and hexachlorobenzene. Lines until 2000 are dashed to indicate uncertainty related to more irregular sampling. Source: Houde et al., 2019.

Beluga whales

In adult male beluga whales from the eastern Beaufort Sea (1989–2015), PCB concentrations were associated with $\delta^{13}C$ values, while levels of dieldrin and mirex were correlated with $\delta^{13}C$ and $\delta^{15}N$ values in the same animals (Noël et al., 2018). These results could be indicative of a shift in diet, and consequently, contaminant exposure over time. In the same population, associations have been observed between climate-related parameters and mercury concentrations, for which more data were available (Loseto et al., 2015; Noël et al., 2018). Smythe et al. (2018) highlighted that climate-driven processes could influence the exposure of beluga whales to contaminants of emerging concern, but they were not well-understood.

Polar bears

An early study by McKinney et al. (2009) was among the first to provide evidence of associations between climate change and contaminant accumulation in Arctic biota. Their study reported changes in the feeding ecology of Western Hudson Bay polar bears between 1991 and 2007. As inferred from stable isotope and fatty acid analyses, bears consumed less ice-associated bearded seals (*Erignathus barbatus*), and more open water-associated harbor seals (*Phoca vitulina*) and harp seals (*Pagophilus groenlandicus*) in years with shorter periods of ice coverage. This change in diet corresponded with higher concentrations of brominated and chlorinated contaminants in polar bear fat, compared to a theoretical situation without dietary changes. The dietary shift had a greater impact on highly biomagnifying chemicals (i.e. PCBs and PBDEs) compared to less biomagnifying chemicals (i.e. HCH isomers) in polar bears.

The study by Morris et al. (2021) described above for thick-billed murres also addressed comparative temporal trends of PCB-153, PBDE-47, *p,p'*-DDE, α-HCH and PFOS in polar bears from the Hudson Bay, together with changes in climate parameters (Table 2.4.2). Unlike POP concentrations in murre eggs, levels in polar bears could not be normalized to $\delta^{15}N$, as the extent of stable isotope data available for these animals was limited. As described for the murre eggs, relatively subtle adjustments to annual contaminant trends were observed when climate factors were included in the GLMs. However, models that incorporated sampling year with time-lagged AO or NAO index values were among the best ranked for describing contaminant concentrations in both species, and showed positive associations between POP concentrations and time-lagged AO or NAO indices.

The relationships observed for the legacy POPs in Western Hudson Bay bears were consistent with many of those for the murre eggs (Table 2.4.2), although results for polar bears varied depending on the degree of sea ice coverage and the time-lag periods used to calculate sea ice metrics. Later freeze-up dates (i.e. less sea ice) were related to greater concentrations of PBDE-47 in both subpopulations of polar bears, and greater PFOS concentrations in Southern Hudson Bay bears. In murre eggs, less sea ice was associated with lower concentrations of legacy POPs (Morris et al. 2021). Reasons for these differences are unknown.

As shown for thick-billed murres, inverse relationships were also found between seasonal and time-lagged air and land surface temperatures and concentrations of legacy POPs and PBDE-47 in polar bears (Table 2.4.2). Years with greater fall or winter precipitation (i.e. snow) amounts were associated with lower POP concentrations in polar bears. This inverse relationship between contaminant concentrations and precipitation amounts was not observed in murres, and may be related to the hunting behavior of polar bears. For example, greater snowfall in the fall/winter may impede seal predation by polar bears as has been observed in spring when bear predation on seal dens was negatively correlated to snow depth (Hammill and Smith, 1991; Ferguson et al., 2005).

2.4.2.3 Conclusions for North America

Few studies have investigated the effects of climatic factors on the accumulation of POPs in Canadian Arctic fish and wildlife. Present results indicate correlations between temporal trends of POPs in fish, seabirds, ringed seals and polar bears and diverse climate parameters. These associations were found to vary between sites, species, and tissues analyzed. As suggested by McKinney et al. (2015), the development of tissue-derived tracers (e.g., specific stable isotopes, lipid markers) of body condition, habitat, and trophic interactions could be of great help to better understand the ecological and biological changes that are related to climate change and have consequences for contaminant levels. Continuous long-term monitoring and assessment of contaminants, and systematic and standardized recording of environmental parameters are important. Collaborative work between experts in meteorology, hydrology, contaminants and climate change is also warranted to adequately evaluate associations between all environmental factors potentially affecting contaminant accumulation in biota. Broader time frames are also needed to account for time lags between changes in POP emissions, climate parameters, and contaminant accumulation in the Arctic environment (Foster et al., 2019).

2.4.3 Links between POP time trends and climate change in Greenland and the Faroe Islands

2.4.3.1 Faroe Islands

POP monitoring and time series

A monitoring programme for contaminants in the environment of the Faroe Islands was established in 1997 with funding from the Danish Environmental Protection Agency. Environmental monitoring is predominantly focused on marine species, including pilot whales (*Globicephala melas*), Atlantic cod (*Gadus morhua*), black guillemots (*Cepphus grylle*) and northern fulmars, as well as some freshwater species, such as Arctic char and brown trout (*Salmo trutta*), and, with less intensity, terrestrial species including mountain hares (*Lepus timidus*) until 2010, and sheep (*Ovis aries*) until 2017. Most species are sampled annually, but some are sampled biannually or with lower frequency (Figure 2.4.6). The longest-running time series dating back to 1986 involves samples of pilot whales collected before the establishment of the monitoring programme. Time series results have recently been reported in Andreasen et al. (2019). Additionally, published temporal trends for FRs, chlorinated and brominated dioxins and furans, and PFAS in pilot whales have shown decreasing concentrations of PBDEs, but increasing concentrations of long-chain perfluorocarboxylic acids (PFCAs) (Bjurlid et al., 2018; Dassuncao et al., 2017). Although associations between mercury in Arctic char and air temperature have been studied, linkages between POP concentrations and climate parameters in Faroese biota have not yet been investigated.

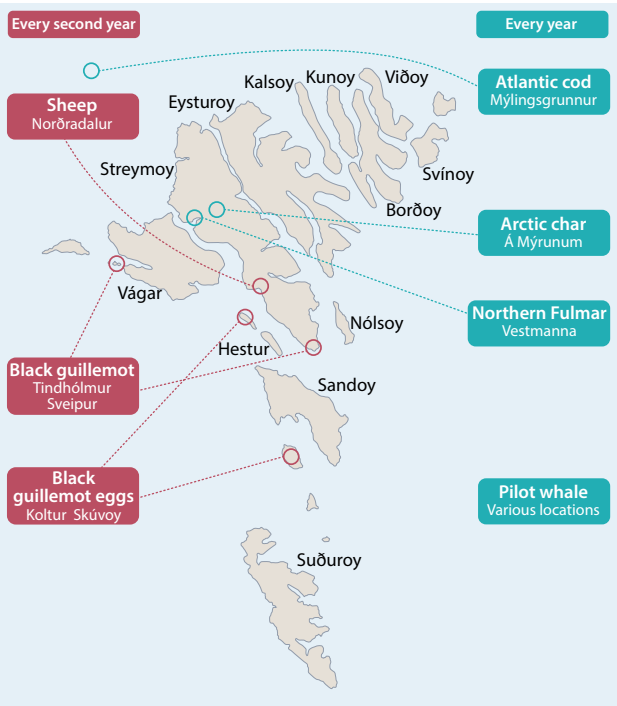


Figure 2.4.6 Sampling stations and frequency for POP monitoring of biota from the Faroe Islands.

2.4.3.2 Greenland

POP monitoring and time series

The contaminant monitoring programme in Greenland, the AMAP Core Programme, was established by the Danish Environmental Protection Agency in 1994. Originally designed with a five-year sampling interval (1994; 1999), it was later changed to annual (1999-2002) and eventually to biannual sampling (since 2002) (Rigét et al., 2000b). However, polar bears are sampled every year. The AMAP Core Programme has several sub-programmes, including temporal trends of POPs in key species (Rigét et al., 2016), trends of chemical elements, such as mercury, cadmium and selenium (Dietz et al., 2011), biological effects of POPs and heavy metals on polar bears (Sonne et al., 2012), and emerging issues, such as chemicals of emerging Arctic concern (Vorkamp et al., 2015a).

Figure 2.4.7 shows the programme's key species and collection locations. In 2005, the programme established an ESB in which all samples for analysis and excess samples are stored for potential other uses, such as screening studies within or outside the programme (Vorkamp et al., 2017), or studies run by external collaborators (Rotander et al., 2012).

Contaminant trends in Greenland biota have been published regularly, including those for legacy POPs, PBDEs, hexabromocyclododecane (HBCDD), PFAS and endosulfan in ringed seals (Rigét et al., 2006, 2013b; Vorkamp et al., 2008, 2011, 2012, 2015b, 2017), HBCDD in glaucous gulls (Vorkamp et al., 2012) and legacy POPs, octachlorostyrene, PBDEs and HBCDD in polar bears (Dietz et al., 2013a, 2013b, 2018). The POP time series for ringed seals (from Qeqertarsuaq) and Arctic char were previously included in studies investigating associations with biological variables and climate parameters (Rigét et al., 2010, 2013a), as further detailed below.

Figure 2.4.7 Greenland species and locations sampled as part of the AMAP Core Programme. Species listed in red are analyzed for POPs on a biannual basis, while species listed in blue are banked for potential other purposes. All samples are stored in the Environmental Specimen Bank at Aarhus University, Denmark.

Terrestrial environment

There are currently no known studies of associations between climate parameters and POP temporal trends in Arctic terrestrial biota available from Greenland.

Freshwater environment

Organochlorine time trends in Arctic char from a lake near Isortoq in Southwest Greenland (Figure 2.4.7) were analyzed in relation to biological variables (fish length, $\delta^{15}N$, lipid content) and summer air temperature (Rigét et al., 2010). The study spanned the period from 1994–2008, inclusive of five years with organochlorine data. Concentrations of ΣPCBs, ΣDDTs and *trans*-nonachlor were significantly correlated with fish length, while no association was found with $\delta^{15}N$ for any of the contaminants. When expressed on a wet-weight basis, all compounds except HCB declined significantly in Arctic char over time. However, no temporal decline was evident when contaminant concentrations were expressed on a lipid-weight basis, since fish lipid content decreased over this time period as well. The temperature index increased significantly over this time period, however it is unknown if any causal relationships exist between these variables, such as an effect of increasing temperatures on fish lipid content and/or exposure to contaminants (Rigét et al., 2010).

A recent update of this analysis spanning eight years of organochlorine data collected between 1999–2017 included fish length and $\delta^{15}N$ values, as well as air temperature for the preceding summer, and AO index of the preceding winter as variables (Rigét et al., 2020). Wet-weight and lipid-normalized concentrations of all compounds (ΣPCBs, PCB-153, ΣDDTs and α-HCH) except HCB decreased significantly over time in Arctic char muscle. Using linear regression models and lipid-normalized POP concentrations, air temperature was found to be an important predictor for ΣPCBs and α-HCH in Arctic char, and in both cases, air temperature was positively associated with POP concentrations in fish (Figure 2.4.8). No association was found between the AO index and contaminants in Arctic char, while $\delta^{15}N$ values were only correlated to HCB concentrations, and were positively related. A lack of decreasing HCB concentrations has also been observed in other time trend studies (Rigét et al., 2019) and may indicate the existence of secondary emissions, which to some extent may be driven by climate change (Ma et al., 2011). However, this remains speculative at this point.

The positive associations between summer air temperature and contaminants (ΣPCBs and α-HCH) were different from findings reported from the Canadian Arctic, where no correlations were found between air temperature and POP concentrations in fish (Cabrerizo et al., 2018b), as described in Section 2.4.2.2.

Marine environment

Ringed seals

Time trends of selected POPs (PCB-52, PCB-153, *p,p′*-DDE, HCB, α-HCH and β-HCH) in ringed seals from Qeqertarsuaq (West Greenland), spanning nine years of data collected between 1994–2010, were explored for associations with climate parameters and biological variables (Rigét et al., 2013a). Climate parameters used in the analyses included the number of days

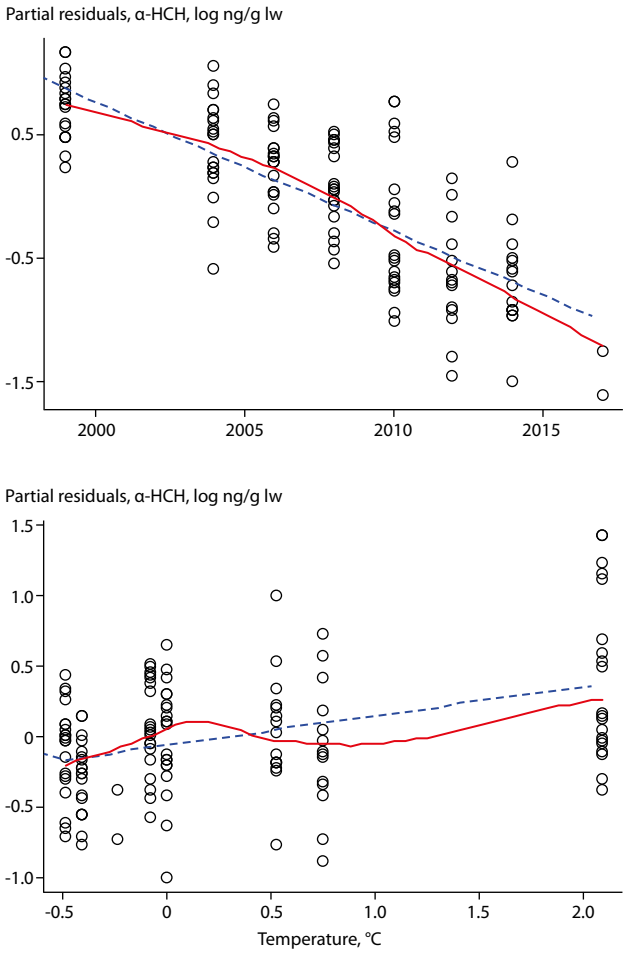


Figure 2.4.8 Relationships between α-HCH in Arctic char from Southwest Greenland, year and air temperature of the preceding summer (Rigét et al., 2020). Points indicate individual animals, the dashed line shows the least squares regression, and the solid red line shows the smoothed trend.

with >50% ice coverage (i.e. sea ice days; November–May), the AO index for the preceding winter, as well as water temperature and salinity values for the preceding summer.

Over the study period, sea ice cover decreased significantly, while the other climate parameters showed no significant change. In addition, all organochlorines showed decreasing trends, but associations with biological variables and climate parameters varied (Rigét et al., 2013a). Trophic position, as indicated by $\delta^{15}N$ values, had a strong predictive power for HCB concentrations in ringed seals, indicating a positive association with diet, but the strength of this relationship was less so for the other contaminants. Among the climate parameters, the most important associations observed were between sea ice days and PCB-153 (negative relationship), winter AO condition and α-HCH (negative relationship) and summer salinity and β-HCH (positive relationship). The mechanisms behind these associations are unknown and likely complex. Rigét et al. (2013a) suggested that the extent of sea ice may affect the type and availability of seal prey, with consequences for POP exposure and lipid stores. A stronger winter AO could transport more heat to the Arctic, possibly resulting in less sea ice coverage and enhanced volatilization of a relatively volatile compound like α-HCH. Higher salinity values could indicate a greater influx of the Irminger Current into the study area, potentially transporting compounds with relatively high water

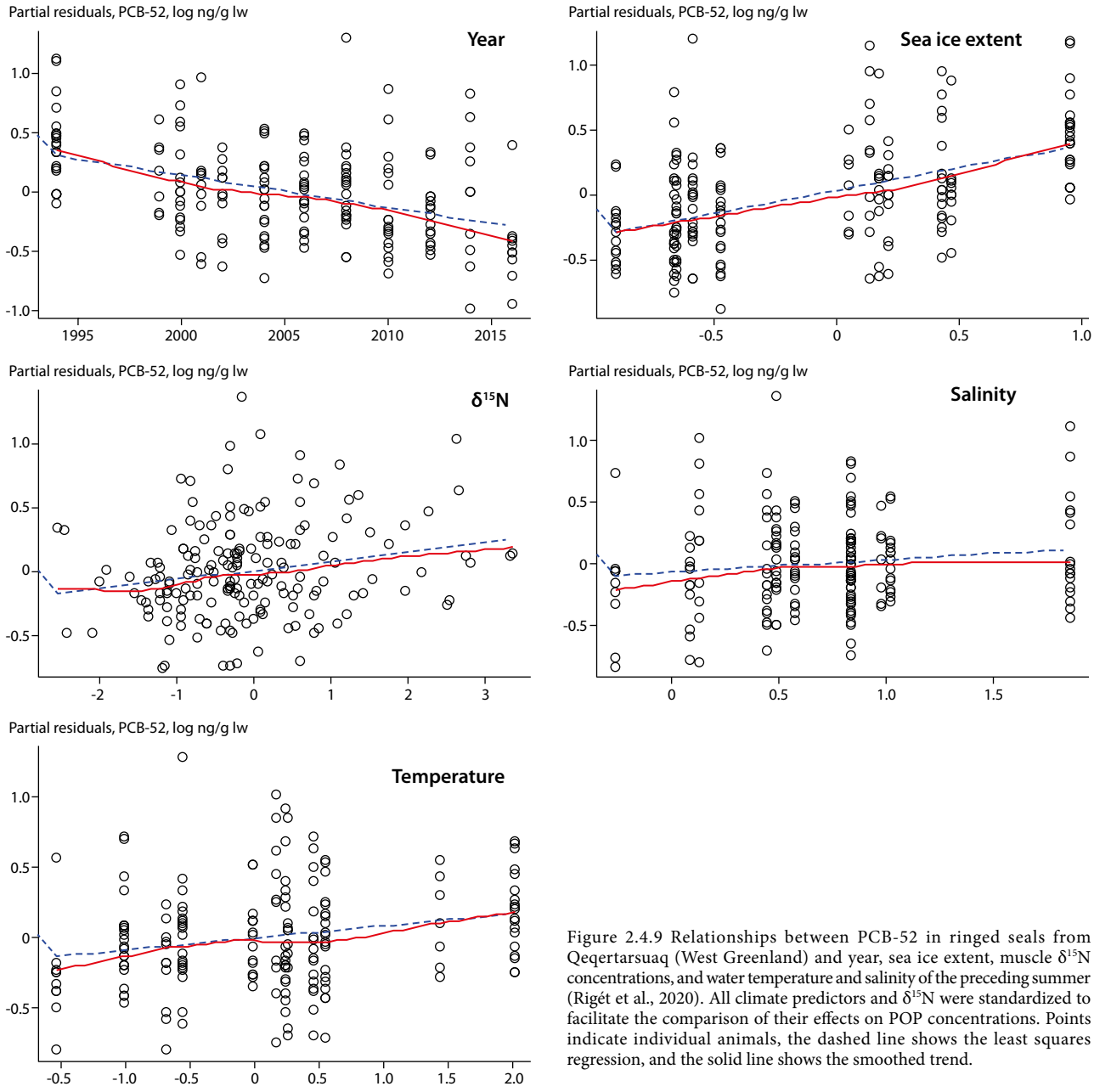


Figure 2.4.9 Relationships between PCB-52 in ringed seals from Qeqertarsuaq (West Greenland) and year, sea ice extent, muscle $\delta^{15}N$ concentrations, and water temperature and salinity of the preceding summer (Rigét et al., 2020). All climate predictors and $\delta^{15}N$ were standardized to facilitate the comparison of their effects on POP concentrations. Points indicate individual animals, the dashed line shows the least squares regression, and the solid line shows the smoothed trend.

solubility, such as β-HCH. An overall conclusion from this study was that biological and climate variables were important to include in temporal trend studies.

The study was recently updated and extended to include ringed seals from Ittoqqortoormiit (East Greenland; Figure 2.4.7) (Rigét et al., 2020). The update extended the study period to 1994–2016 inclusive of 12 years with data, and 1986–2016 inclusive of 14 years with data, for ringed seals from Qeqertarsuaq and Ittoqqortoormiit, respectively. It included the same compounds as analyzed previously (Rigét et al., 2013a), however, without β-HCH, and with ΣDDT replacing *p,p'*-DDE.

For Qeqertarsuaq (West Greenland), analyses included sex, age, and muscle $\delta^{15}N$ concentrations of ringed seals, as well as the water temperature and salinity values of the preceding summer, sea ice extent of the preceding year, and the AO index of the preceding winter. As described above for Arctic char, all compounds except HCB decreased in ringed seals over the study period. Sea ice extent was a major predictive variable for all compounds except α-HCH, indicating that concentrations of most POPs increased in seals with increasing sea ice extent (Figure 2.4.9). This was different from the findings of the previous study which covered a shorter time period (Rigét et al., 2013a). However, the negative association between winter AO index and α-HCH was confirmed in this updated study. Salinity in the preceding summer was positively correlated with ΣPCBs and ΣDDTs, as was water temperature with ΣPCBs, ΣDDTs, PCB-52 and PCB-153 (Rigét et al., 2020).

POPs in ringed seals from Ittoqqortoormiit (East Greenland) were studied in relation to seal age, sex, and muscle $\delta^{15}N$ values, the sea temperature index north of 80°N in the preceding year, the sea ice extent in September, and the mean AO index for the preceding winter (Rigét et al., 2020). The sea temperature anomaly north of 80°N and September sea ice extent showed significant increasing and decreasing trends, respectively, over the study period. All compounds, including HCB, decreased in seals over time. Both age and muscle $\delta^{15}N$ values were positively associated with POP concentrations, with the exception of

a negative association between α-HCH and age, and weak negative associations between HCB and age.

Unlike the results from West Greenland, sea ice extent was not an important predictor for any of the compounds. The winter AO index was positively correlated with HCB and α-HCH, also in contrast to the results for α-HCH in ringed seals from West Greenland. Seawater temperature was positively correlated with HCB, but not with any other compounds.

Various associations between sea ice and POPs in ringed seals have been reported previously, and these differences may be related to complex biological and physical processes. For example, Gaden et al. (2012) reported higher levels of POP concentrations in ringed seals of the Canadian Arctic during years with earlier sea ice breakup, suggesting the length of the ice-free season influenced the feeding behavior of ringed seals, as discussed in Section 2.4.2.2. Also in the Canadian Arctic, Houde et al. (2019) reported a positive relationship between sea ice coverage and POPs in ringed seals, suggesting increased accumulation of contaminants during years with greater ice extent, with a possible influence of ice type (i.e. first-year vs. multi-year ice coverage). Although sea ice extent was associated with POPs in ringed seals from West Greenland, it was not a relevant parameter for ringed seals from East Greenland. Therefore, the role of sea ice in influencing the POP exposure of ringed seals is not well understood. Less sea ice increases the air-water exchange of POPs, thus favoring volatilization and lowering the food web availability of volatile compounds (Bossi et al., 2013; Ma et al., 2016). At the same time, it also decreases the uptake of POPs from ice and by ice-associated organisms. Both processes are consistent with positive associations between extent of sea ice and concentrations in the seals.

The AO value was negatively associated with α-HCH in West Greenland seals, but positively associated with α-HCH, in addition to HCB, in East Greenland seals. An increasing AO index might reflect increased transport of air masses, but also heat, to the Arctic, which could serve to reduce sea ice, enhance volatilization of volatile compounds like α-HCH and HCB, and thus affect their availability for uptake into food webs. As discussed in Section 2.4.2.2, positive associations between oscillation indices and POPs in ringed seals were also reported from Canada (Houde et al., 2019). Positive associations shown between POPs and parameters such as salinity and seawater temperature in West Greenland might be reflective of a greater influx of warmer, higher salinity water, such as that of the Irminger Current which carries water from the Northeast Atlantic northward. A relatively larger influx of water from the Irminger Current than from the Arctic Ocean could have potential implications for the transport of contaminants to West Greenland. Sea temperature was also an important predictor for HCB in East Greenland seals, which could possibly be related to the air-water exchange processes of relatively volatile compounds like HCB.

Rigét et al. (2020) also discussed the effect of including biological and climate variables in calculations of annual POP concentration changes. Inclusion of these variables in calculations either reduced or enhanced the annual rate of POP concentration declines by factors ranging from 0.1 to 2.3. The influence of these variables appears minor at present, as emissions reductions are the main driver of POP temporal trends, however, this may change in the future. It is striking that these effects are smallest for ringed seals from East Greenland, for which the longest time series are available, and thus are most likely to be influenced by emission changes.

Polar bears

Similar to the findings reported for polar bears from Western Hudson Bay, Canada (Section 2.4.2.2), dietary changes over time were found for polar bears from East Greenland (McKinney et al., 2013). Based on analyses of fatty acid signatures and fatty acid $\delta^{13}C$ patterns in 310 subadult, adult male and adult female polar bears sampled between 1984–2011, and various prey species, Arctic ringed seal was originally identified as the main prey, accounting for 48% of the bears' diet on average over time. However, ringed seal consumption declined during the monitoring period from 90±2.5% of the diet in 1984 to 34±11% of the diet in 2011, while the consumption of subarctic hooded seals (*Cystophora cristata*) and potentially harp seals, increased from 0% of the bears' diet in 1984 to 26±9% of the bears' diet in 2011. Possible reasons for these shifts in diet are discussed in Chapter 2.3.

This dietary shift towards subarctic species with higher contaminant burdens likely influenced POP levels in the polar bears. POP concentrations in polar bear adipose tissue declined more slowly, paralleling changes in diet. Hooded seal consumption was negatively correlated with the NAO, indicating more hooded seals were consumed in years with lower NAO index values. Stronger negative NAO phase (NAO-) states were found to be associated with warmer temperatures and less sea ice in East Greenland, however, no statistical association was found between hooded seal consumption and the area of the East Greenland sea ice (McKinney et al., 2013).

This work has lately been extended through 2016 and expanded to include contaminant analyses of polar bear prey species (Dietz et al., 2021). After a long period of continuously declining POP concentrations in adipose tissue of East Greenland polar bears, a temporary shift to increasing POP concentrations was observed around 2007 (Dietz et al. 2013a, 2013b, 2018, 2021; Rigét et al., 2016). This short-term anomaly may be associated with changes in the bears' diet. The bears' main prey, ringed seals, did not show the same increase in POP concentrations around 2007. Furthermore, the proportion of ringed seal in the polar bear diet continued to decline while the proportion of hooded seal increased (Rigét et al., 2016; Dietz et al., 2021).

The changes in the POP time trends observed in East Greenland polar bears coincided with dietary changes identified by fatty acid signature analysis (McKinney et al., 2013; Dietz et al., 2021). The decline in ringed seal consumption and corresponding increase in harp and hooded seal consumption was most pronounced for juvenile polar bears, which represented the majority of the individuals included in the analysis (Figure 2.4.10). However, the diet of adult males had always consisted of a higher proportion of harp seal than ringed seal, offering one potential explanation for POP differences observed between age groups and sexes (Dietz et al., 2021).

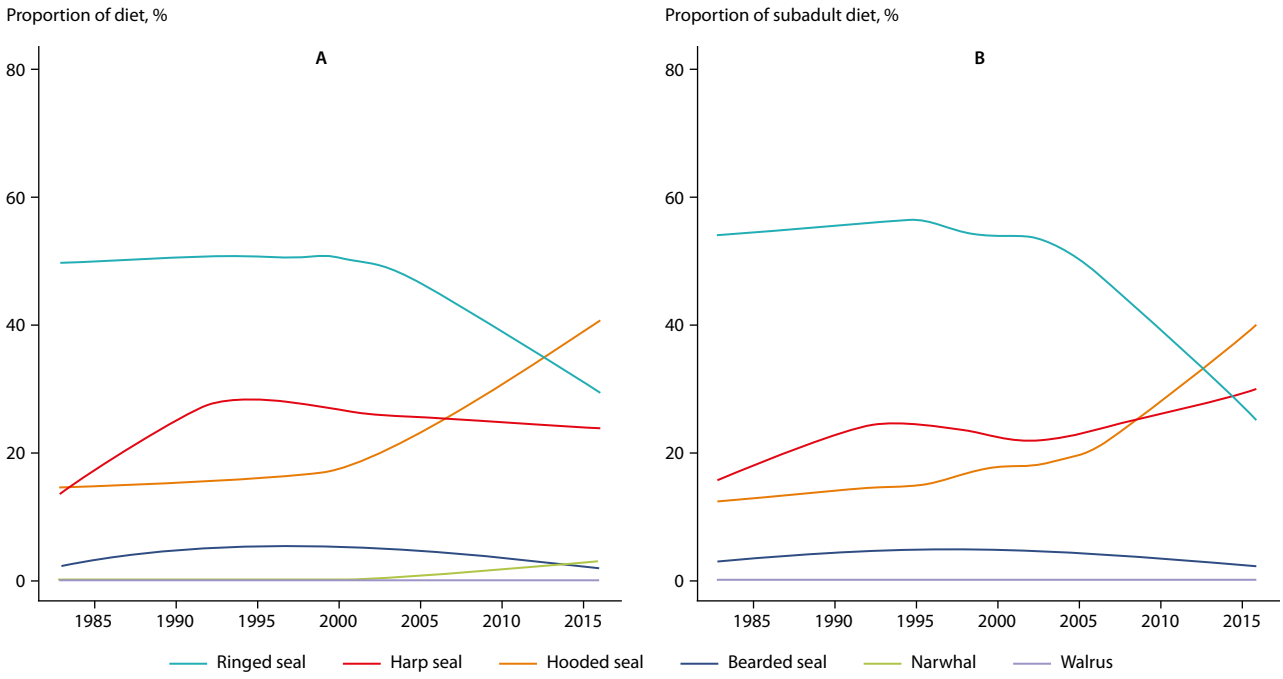


Figure 2.4.10 Percentage of various prey species comprising East Greenland polar bear diet over time (1983-2016) based on quantitative fatty acid signature analysis. A: all bears; B: juvenile bears (Dietz et al., 2021).

Selected POP concentrations for potential polar bear prey collected at Ittoqqortoormiit (East Greenland) in 2015 are shown in Figure 2.4.11 (Dietz et al., 2021). Most of these species have higher POP concentrations than ringed seals, which is consistent with the observed changes in diet and POP time trends in polar bears. The largest differences were those between ringed seals and hooded seals, as well as between ringed seals and narwhals (*Monodon monoceros*) (Figure 2.4.11). However, a different pattern was observed in PFAS concentrations; the highest median concentrations of both perfluorosulfonic acids (ΣPFSAs) and ΣPFCAs were found in ringed seals, followed by harp seals (ΣPFSAs) and narwhal (ΣPFCAs) (Dietz et al., 2021). Consequently, in years with low percentages of ringed seals as prey species, polar bears had relatively low PFAS levels and higher POP levels. The opposite was the case in years with a high percentage of ringed seals in their prey.

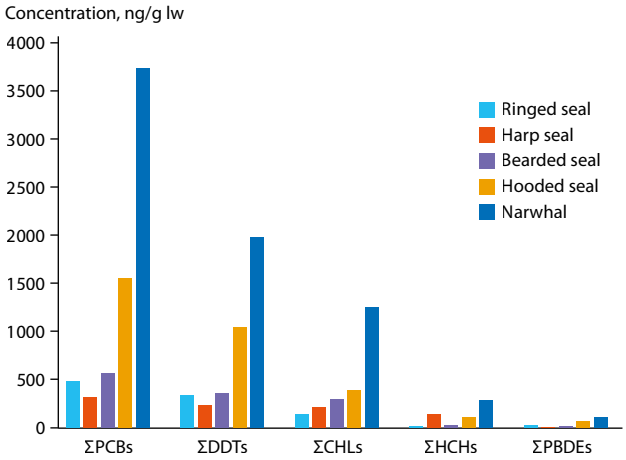


Figure 2.4.11 Median POP concentrations in blubber of East Greenland polar bear prey species collected in 2015 (Dietz et al., 2021). Ringed seal: n=20; harp seal: n=9; bearded seal: n=6; hooded seal: n=5; narwhal: n=19.

2.4.3.3 Conclusions for Greenland and the Faroe Islands

Current studies on the potential links between changes in climate parameters and POPs in Arctic biota have been limited to Arctic char, ringed seals and polar bears from Greenland, however, there is data available on temporal changes of POP concentrations in biota from the Faroe Islands and Greenland that would allow for additional studies of this kind, potentially validating the current findings.

A number of associations have been identified between POPs in Arctic biota and climate parameters. However, these correlations do not necessarily indicate that causal relationships exist and the mechanisms behind these observations are still largely unknown. Some plausible explanations exist, but require scientific evidence. Results from Greenland also show that both biological and climate change parameters are associated with POP temporal trends in Arctic char, ringed seals and polar bears, indicating complex interactions that warrant further study.

Although the same methodology was used, several contrasting results were found for ringed seals from West and East Greenland, respectively, including differences in the significance of sea ice extent, and whether associations between the AO and α-HCH concentrations in biota were positive or negative. In combination with differences from Canadian findings, these results suggest that the effects of climate change on contaminants in the Arctic may vary regionally. Most associations were found between climate parameters and relatively volatile compounds, such as HCB and HCH, suggesting that climate change might particularly affect those processes where volatility plays a role, such as air-water exchange or long-range transport.

The influence of climate change on POP time trends in biota, including effects mediated through ecosystem changes, appears to be limited based on the small variations in annual changes estimated by models with and without the inclusion

of climate parameters. The dominating influence on the overall development of POP concentrations in Arctic biota still seems to be that of reduced emissions. However, the effect of climate change seems to be larger on shorter time series that mainly consist of recent data, suggesting climate change may potentially be exerting an increasing influence on POP trends. Therefore, it will be important to continue POP monitoring, including attempts to quantify the current and future influence of changes in climate parameters.

2.4.4 Links between POP time trends and climate change in the other Nordic countries

2.4.4.1 Norway (including Svalbard)

POP monitoring and time series

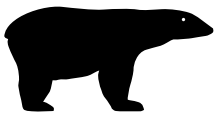
Monitoring of pollutants in the Norwegian Arctic is conducted by the government via the Environmental Monitoring of Svalbard and Jan Mayen (MOSJ) programme, as well as by other research and monitoring campaigns. MOSJ is an environmental monitoring system that utilizes a selection of key indicators collected by various thematic programmes. MOSJ obtains relevant information on pollutants from monitoring and research efforts of the Norwegian Institute for Air Research, the Norwegian Polar Institute, and the Institute for Marine Research, with biota studies being performed by the latter two. For example, polar bear samples from Svalbard are collected as part of annual monitoring operations by the Norwegian Polar Institute which predominantly focuses on population biology, but also conducts research on pollution, anthropogenic activities and climate change. Studies on the effects of climate change on temporal trends of POPs in Arctic biota from Norway have been built on MOSJ time series or conducted through independent research projects. The expected impacts of climate change on the Norwegian Arctic (Svalbard) were recently summarized in a report entitled *Climate in Svalbard 2100* (Hanssen-Bauer et al., 2019), however, there are only a few studies that have linked environmental pollution in biota with climate parameters in the region.

Terrestrial environment

Climate warming is particularly pronounced in the Svalbard area as evident by the reduced duration and extent of sea ice cover, reduced duration of terrestrial snow cover and increased frequency of rain-on-snow events (Descamps et al., 2016; Peeters et al., 2019). As Arctic foxes (*Vulpes lagopus*) from Svalbard feed on food items from both terrestrial and marine ecosystems (Eide et al., 2005; Ehrich et al., 2015), environmental changes may affect their feeding ecology and consequently, their contaminant exposure. Temporal trends of lipophilic POPs and PFAS were studied in young Arctic foxes from Svalbard in relation to their feeding habits and food availability (Andersen et al., 2015a; Routti et al., 2017). The POP study included 141 fox samples collected between 1997–2013, and the PFAS study included 113 fox tissue samples collected between 1997–2014. Feeding habits were studied using stable isotope values in fox muscle tissue, while reindeer mortality and sea ice cover were used as proxies for reindeer and seal availability, respectively.

POP concentrations in Arctic foxes were adjusted for variation in feeding habits and food availability (i.e. biological parameters were included as covariates in the models along with collection year), whereas unadjusted trends only included collection year as a predictor. In general, concentrations of PCBs, chlordanes, *p,p′*-DDE, mirex and PBDEs decreased in foxes by 4–11% per year, whereas HCB and β-HCH concentrations remained stable (Andersen et al., 2015a). The concentrations of all compounds in foxes increased along with the percentage of marine-based diet, HCB concentrations decreased with increasing reindeer mortality, and β-HCH concentrations increased with increasing sea ice cover. However, trends of lipophilic POPs, which were not adjusted for feeding habits or food availability, were similar to the adjusted trends (Figure 2.4.12).

Arctic foxes, juveniles
POPs: n=141; winters,1997-2013
PFAS: n=113; winters, 1997-2014



Polar bears, adult female
POPs: n=306; springs,1997-2017
PFAS: n=192; springs, 2000-2014

Compound	Measured trends	Trends corrected for covariates	Measured trends	Trends corrected for covariates
ΣPCBs	↓	↓	↓	↓
ΣOH-PCBs	↓	↓	↓	↓
ΣChlordanes	↓	↓	↓	↓
HCB	↔	↔	non-linear	non-linear
Mirex	↓	↓		
β-HCH	↓	↔	(↓)	↔
p,p'-DDE	↓	↓	non-linear	non-linear
ΣPBDEs	↓	↓		
PBDE-47			↓	↓
PBDE-153			↔	↑
PFOS	↓	↓ non-linear	↓ non-linear	↓ non-linear
PFNA	↔	(↑)	↑	↑
PFUnDA	↑ non-linear	↑	↑	↑

Figure 2.4.12 Comparison of unadjusted and adjusted POP trends in juvenile Arctic foxes and adult female polar bears from Svalbard. Adjusted trends were corrected for climate-related changes in feeding habits and/or food availability (Arctic foxes) and body condition (polar bears). Green, red and yellow colors indicate decreasing, increasing and no trends, respectively. Arrows in brackets indicate non-significant trends. The non-linear trends for HCB and *p,p′*-DDE in polar bears have shown increasing concentrations in recent years. Data are from Andersen et al. (2015a), Routti et al. (2017), and Lippold et al. (2019).

PFAS concentrations were also higher in foxes feeding on marine-based and higher trophic level food items. Concentrations of PFCAs with nine to twelve perfluorinated carbon atoms (C_9–C_{12} PFCAs) decreased in foxes in relation to increasing reindeer mortality, and some PFAS increased in foxes with increasing sea ice cover (Routti et al. 2017). PFAS concentrations in foxes adjusted for feeding habits and food availability were only slightly different from unadjusted concentrations. Adjusted PFOS concentrations decreased in Arctic foxes during the period 1997–2010 and were stable thereafter, whereas the unadjusted concentrations decreased throughout the study period (Figure 2.4.12). The difference in the trends may be related to the reductions in sea ice observed since 2010, as PFOS concentrations in foxes increased with sea ice extent. Concentrations of longer chain C_{12}–C_{13} PFCAs adjusted for fox feeding habits and food availability increased throughout the study period, whereas unadjusted concentrations increased only after 2003 (Figure 2.4.12).

In conclusion, concentrations of several lipophilic POPs and PFAS in Arctic foxes were associated with climate-induced variations in food availability and changes in feeding habits. However, temporal variations in feeding habits and food availability had only minor influences on temporal trends of pollutants.

Freshwater environment

Muscle tissue of Arctic char from Bjørnøya (Lake Ellasjøen) was collected from 1998–2017, spanning 13 different years and including 127 individuals in total. Time trends for concentrations of HCB and ΣPCBs (sum of PCB-101, PCB-118, PCB-153, PCB-138 and PCB-180) were corrected for biological covariates such as length, reproductive status, sex and body condition. While ΣPCB concentrations in char continuously decreased over the study period (Figure 2.4.13), HCB concentrations decreased only until 2006, whereafter concentrations began to stabilize (Figure 2.4.14). According to temperature records, the atmospheric temperature at Bjørnøya increased during the same time period (MOSJ, 2021).

The decrease of ΣPCB concentrations observed in Arctic char over time is most likely due to regulations targeting primary sources, but is also highly correlated with declining populations of glaucous gulls at Bjørnøya (Carlsson et al., 2018; Blévin, pers. comm.). It has been shown that gull guano is an important vector of contaminants into Lake Ellasjøen (Evenset et al., 2007); therefore, it is believed that a decrease in glaucous gull numbers, in combination with lower PCB emissions, is more influential than climate change in controlling ΣPCB concentrations of Arctic char in Lake Ellasjøen (Blévin, pers. comm.). The cause for the decrease of glaucous gull populations is subject to investigation and research.

It was suggested earlier that the stabilization, rather than a decrease of atmospheric HCB concentrations may be related to increased air temperatures in combination with continued emissions of HCB produced as a by-product of industrial processes (Hung et al., 2016). Warmer temperatures may cause higher emissions of HCB than PCBs from secondary sources as HCB is more volatile (Wöhrnschimmel et al., 2012a, 2013; Carlsson et al., 2018). As discussed in Chapter 2.2, sea ice acts as a barrier that reduces the transfer of POPs from the ocean into the atmosphere. However, the role of sea ice may be less relevant around Bjørnøya, where there is generally less ice than in Svalbard. It is therefore suggested that the declining population of glaucous gulls at Bjørnøya is a prominent factor for explaining the decreasing HCB concentration in Arctic char over time; they were found to be significantly correlated (Blévin, pers. comm.).

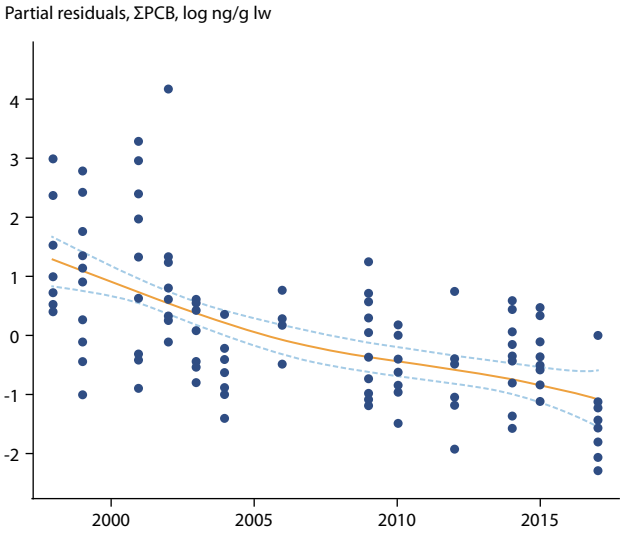


Figure 2.4.13 ΣPCB concentrations in Arctic char muscle collected from Lake Ellasjøen, Bjørnøya, Svalbard, between 1998-2017, adjusted for their influential biological drivers (condition, reproduction status and length). Points indicate individual fish concentrations, the solid line indicates the smoothed line of the partial residuals from the generalized additive model, and the dashed lines indicate the 95% confidence interval. ΣPCBs represents the sum of PCB-101, PCB-118, PCB-153, PCB-138 and PCB-180.

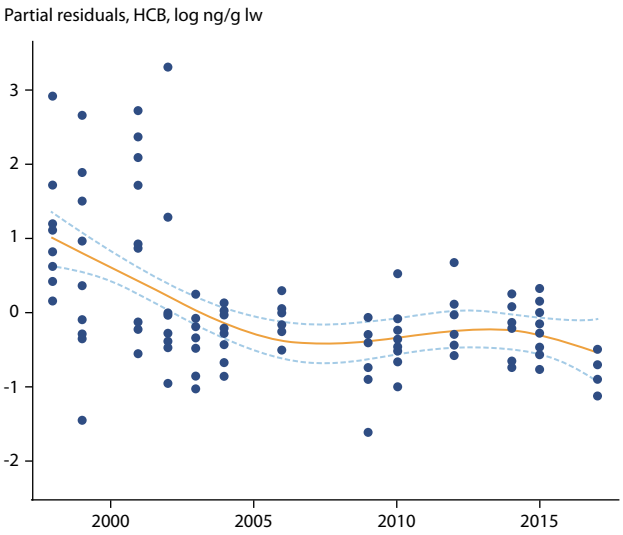


Figure 2.4.14 HCB concentrations in Arctic char muscle collected from Lake Ellasjøen, Bjørnøya, Svalbard, between 1998-2017, adjusted for their influential biological drivers (reproduction status and length). Points indicate individual fish concentrations, the solid line indicates the smoothed line of the partial residuals from the generalized additive model, and the dashed lines indicate the 95% confidence interval.

Marine environment

Zooplankton

Even though the contaminant exposure of apex predators is influenced by the changes and variations that occur in lower trophic level organisms, little attention has been given to the changes in contaminant concentrations of these organisms from a long-term perspective. The International Polar Year Contaminants in Polar Areas (COPOL) project has studied POP bioaccumulation in zooplankton (*Calanus* sp.) around Svalbard, especially near Kongsfjorden on the west coast of Spitsbergen (Table 2.4.3; Figure 2.4.15). Compared to time trends in other species (e.g. marine mammals), these data cover relatively short time spans (2007-2009, 2011), but are, to our knowledge, the longest time trends available for contaminants in Arctic zooplankton. Zooplankton were also sampled around Svalbard in 2018, but these data are not yet available. This dataset also includes information on climate parameters.

Seasonal differences in the biomagnification of POPs within a marine pelagic food web including zooplankton and multiple species of fish and seabirds were examined using trophic magnification factors (TMFs) determined at Kongsfjorden in May, July and October of 2007 (Hallanger et al., 2011a, 2011b, 2011c). TMFs within the food web showed seasonal variations; lower water concentrations of POPs in spring (May), and greater concentrations of POPs in high trophic level species (i.e. fish, birds) later in the season (July), resulted in the highest TMFs being observed during summer for most of the POPs investigated. Additionally, the biota sampled in July consisted of greater numbers of boreal zooplankton and fish species than those sampled in the spring and autumn. Together, the greater number of boreal species and higher TMFs observed in summer suggest that future warming of the Arctic and an increasing presence of subarctic species may result in increased contaminant biomagnification. However, other seasonal changes in the food web (e.g. lipid dynamics) can also impact biomagnification and could have a larger influence on TMFs than climate change (Carlsson et al., 2016).

The impact of abiotic factors, such as ice cover, on contaminant patterns in zooplankton was confirmed by Carlsson et al. (2014). The enantiomeric fractions of chiral pesticides (α-HCH, *trans*-chlordane, *cis*-chlordane and oxychlordane) were determined in zooplankton collected from pack ice located north of Svalbard and three Svalbard fjords that differ in their seasonal characteristics (e.g. extent and duration of ice coverage; influence of Arctic vs. North Atlantic water masses) (Table 2.4.3; Figure 2.4.15). Enantiomeric fractions in zooplankton were found to vary between fjords and between sampling years

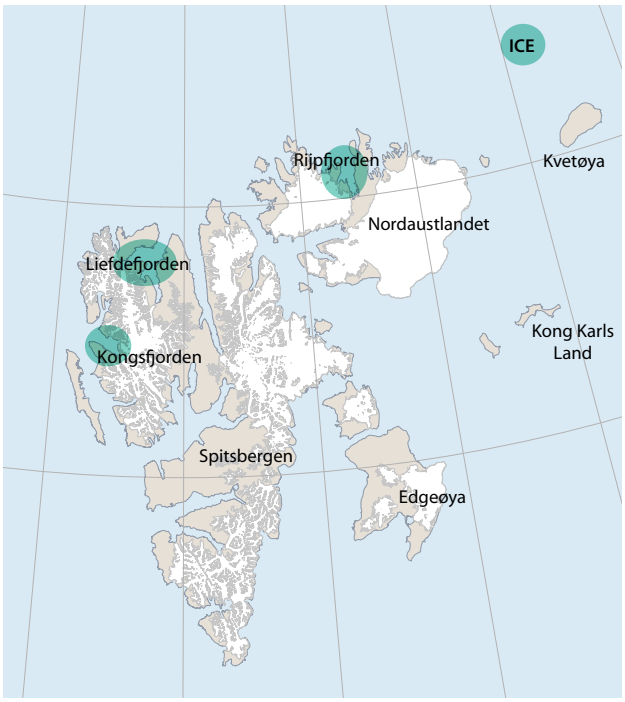


Figure 2.4.15 Locations of fjords and pack ice sampled for zooplankton around Svalbard. Source: Norwegian Polar Institute.

due to differences in the timing of seasonal events and the environmental conditions present at each location. For example, the extent of ice cover, timing of the spring algae bloom, and annual vertical migration from deep- to surface-waters and ice-covered areas were associated with differences in zooplankton POP concentrations and patterns. Additionally, ice coverage was related to the distribution of enantiomeric fractions of α-HCH, with lower enantiomeric fractions observed in zooplankton from ice-covered stations compared to those from ice-free areas. This difference may relate to the reduced air-ocean exchange of α-HCH and other pollutants that can occur when the ocean surface is covered by ice, while on-going biological processes degrade α-HCH present in the underlying waters in an enantiomer-selective way.

Seabirds

Temporal trends of POPs in relation to the AO index were studied in glaucous gulls from Bjørnøya between 1997–2006 (Bustnes et al., 2010). Concentrations of POPs (ΣPCBs, HCB and oxychlordane) decreased significantly, or tended to decrease non-significantly ($p>0.05$) in gull blood samples over the study period. However, gull POP concentrations increased in association with increasing AO index values for

Table 2.4.3 Published data regarding influence of climate on POP concentrations in low trophic level organisms of Arctic marine ecosystems around Svalbard.

Sample area	Sampling year	Dominating water mass*	Winter sea ice conditions*	Reference
Kongsfjorden	2007-2009, 2011	Atlantic	None/partly covered	Hallanger et al., 2011a; 2011c
Liefdefjorden	2008-2009	Atlantic/Arctic	March-July	Hallanger et al., 2011a; 2011b
Rijpfjorden	2009, 2011	Polar surface/Arctic	Ice cover until July	Carlsson et al., 2014
Pack ice	2010-2011	Meltwater/Atlantic	Annual ice cover	Carlsson et al., 2014

*As stated in Carlsson et al. (2014).

the preceding summer and preceding winter, indicating that POP levels were higher in gulls following years with greater transport of air masses from North America and Europe toward the Arctic. In contrast, POP concentrations in gulls decreased with increasing AO index values of the current winter, possibly as a result of changes in diet or winter areas. When AO variation was taken into account in temporal trend analyses, POP concentrations in gulls declined slightly faster than when AO variation was not taken into account. This suggests that climate variability determined by the AO during the period 1997-2006 slowed the decline of POP concentrations in glaucous gulls from Bjørnøya.

The temporal dynamics of POPs, including short-term variations related to egg laying, were studied in the blood of black-legged kittiwakes (*Rissa tridactyla*) from Svalbard over a five-year period (2007–2011) (Bustnes et al., 2017). POP concentrations in kittiwake blood varied greatly between years; PCB concentrations displayed a decreasing trend and HCB displayed an increasing trend, which coincided with higher air concentrations of HCB reported in the area. The temporal dynamics of POPs were also studied in the blood of common eiders (*Somateria mollissima*) nesting in Svalbard and a subarctic location in northern Norway over three- and five-year periods, respectively (Bustnes et al., 2012). Although assessing temporal trends was not the purpose of this study, it demonstrated that the annual variations observed in eider POP concentrations were greater in Svalbard than in northern Norway. The larger variation seen in High Arctic nesting eiders was likely related to changes in their body mass. As fasting eiders lost body mass during the egg incubation period, POPs remobilized from fat tissue into blood, and changes in body mass were particularly rapid during years when the temperature was low.

Polar bears

In the Barents Sea, the retreat of Arctic sea ice has been particularly fast. Since the late 1970s, the number of sea ice covered days has declined by over 40 days per decade (Stern and Laidre, 2016). Polar bears depend on sea ice for hunting their preferred prey of ringed seals and other ice-obligate seals. Two recent studies have investigated temporal trends of lipophilic POPs and PFAS in relation to changes in Barents Sea polar bear feeding habits and body condition (Routti et al., 2017; Lippold et al., 2019). These studies, undertaken in spring, included 306 and 192 adult female polar bear samples, respectively (Figure 2.4.12).

Stable isotope values for nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) determined in polar bear red blood cells declined 1‰ and 2‰, respectively, from 2000–2017 (Lippold et al., 2019). This suggests that over time, the winter diet of Barents Sea polar bears shifted to include less marine, less ice-associated, and more lower trophic level prey items, likely as a consequence of declining sea ice. Body condition also changed in the bears over time. From 1997–2006, bears tended, although not significantly, to get thinner, translating to a loss of 1.3 kg/year. In contrast, from 2006–2017, bears got fatter, with a gain of approximately 0.8 kg/year (Lippold et al., 2019). These changes in bear body condition are also likely connected to changes in sea ice and the availability of ice-obligate seals.

Contaminant concentrations in Barents Sea polar bears also changed over the time periods studied. Concentrations of PCBs, OH-PCBs, oxychlordane and PBDE-47 declined linearly over time in bears (Figure 2.4.16), whereas β-HCH and PBDE-153 concentrations were unchanged. HCB and *p,p′*-DDE declined in bears until 2009 and 2012, respectively, and increased thereafter (Figure 2.4.16) (Lippold et al., 2019). While body condition

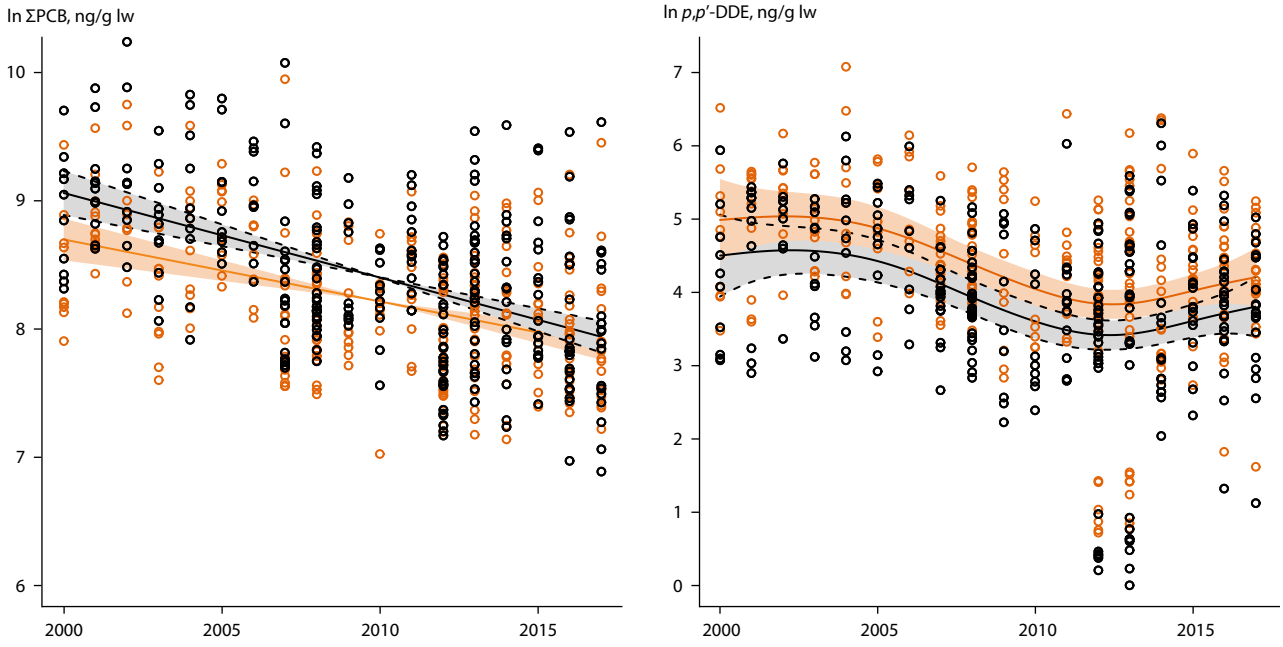


Figure 2.4.16 Temporal trends of ΣPCBs and *p,p′*-DDE in plasma of polar bears from Svalbard and the surrounding Barents Sea. Points represent partial residuals that have been rescaled to log(ng/g lw) concentrations by adding an average of the fitted values for the first year. Measured concentrations and trends are shown in grey, while concentrations and trends adjusted for climate-related variation in body condition and trophic level are shown in orange. Modified from Lippold et al. (2019).

and diet had a significant influence on POP concentrations in polar bears (Tartu et al. 2017b), the changes in these biological factors were not pronounced enough to significantly alter POP temporal trends (Lippold et al. 2019). Long-term POP trends were similar with or without including dietary tracers and body condition as covariates, except for PBDE-153, which increased when adjusted for body condition and $\delta^{13}C$. This suggests that long-term trends of lipophilic POPs in Barents Sea polar bears are still predominantly determined by contaminant emissions, and not climate-related variations in body condition and food webs.

A similar study also concluded that emission changes had a much larger influence than feeding habits on PFAS concentrations in Barents Sea polar bears (Routti et al., 2017). Previously declining PFSA concentrations in bears stabilized in 2009, and annual variations were not influenced by climate-related changes in their diet. PFCA concentrations adjusted for dietary changes generally increased, but the increase in C_{12}–C_{14} PFCAs tended to level-off after 2009 (Figure 2.4.12). The rate of increase was less steep or not significant for PFCA concentrations not adjusted for dietary changes. This is likely because the winter diet of the bears had changed to include more low trophic level prey and more terrestrial prey, such as geese and reindeer (Derocher et al., 2000; Stempniewicz et al., 2013; Tartu et al., 2016). In conclusion, current evidence suggests that temporal trends of POPs in Barents Sea polar bears are only affected by climate-related changes in body condition and food webs to a small degree, and that the main drivers of contaminant trends seem to be primary and secondary emissions.

2.4.4.2 Iceland

POP monitoring and time series

In Iceland, annual monitoring programmes and trace metals have focused on blue mussels (*Mytilus edulis*) and Atlantic cod (*Gadus morhua*). POP levels have generally been decreasing in both species (Sturludottir et al., 2013, 2014), although the time series have not been examined for links with climate parameters.

2.4.4.3 Finland

POP monitoring and time series

Perch (*Perca fluviatilis*) from Lake Inari in Northern Finland have been collected for POP analysis at three-year intervals since 2010 (Mannio et al., 2016), however, the data have not been examined for links with climate parameters. From 2019 onward, sampling has been conducted annually at Lake Inari, with additional sampling in the Tornionjoki and Kemijoki rivers being conducted at three-year intervals. This programme's dataset also includes dated sediment cores from Finnish lakes.

2.4.4.4 Sweden

POP monitoring and time series

Arctic char have been collected annually from Lake Abiskojaure in Northern Sweden every autumn from 1981–2017 and analyzed for trace metals and POPs (Faxneld et al., 2019), however, the POP data have not yet been examined for links with climate parameters or food web characteristics. Samples not used for chemical analyses are stored in the Swedish ESB. In addition to fish samples, specimens of reindeer (from Ammarnäs) and eggs of white-tailed sea eagles (*Haliaeetus albicilla*) (from North Bothnia) are also stored in the ESB.

2.4.4.5 Conclusions for the Nordic countries

Only a few studies from Norway have investigated the influence of climate change on temporal trends of POPs and their focus has generally been on the effects of changing dietary sources. Climate-related changes in food webs, food availability and/or body condition only appear to have had a minor influence on long-term temporal trends of POPs in polar bears and Arctic foxes from Svalbard thus far. The main drivers of temporal changes in chlorinated, brominated and fluorinated POPs for these species are likely still levels of primary and secondary emissions. However, climate variability was identified as a potential factor slowing the decline of POP concentrations in glaucous gulls from Bjørnøya. Additionally, decreasing PCB concentrations in Arctic char of Lake Ellasjøen, Bjørnøya, occurred in parallel to increasing atmospheric temperatures. However, the decline in the local glaucous gull population is a more likely explanation for this decreasing contaminant trend since bird guano is an important vector of POPs into the lake ecosystem. Although several studies have examined the temporal trends of POPs in Arctic zooplankton over short time periods, there are too few data to draw any firm conclusions with regards to the impacts of climate change.

Studies linking climate change parameters (e.g. pH, temperature, meltwater runoff and related terrestrial impacts on marine ecosystems) and time trends of POPs in biota in the Norwegian Arctic environment are largely lacking. Although time series of POPs in biota are available from Arctic areas of other Nordic countries, they have not been examined for links with regional climate parameters. As discussed for other regions, the availability of POP time series provides an opportunity for additional studies to explore the effects of climate parameters on the long-term trends of POP concentrations in biota from the Nordic countries.

2.4.5 Links between POP time trends and climate change in Russia

POP monitoring and time series

Studies on contaminants in the Russian Arctic have been referenced in earlier AMAP assessments (e.g. AMAP, 2010b, 2016b), More recently, temporal changes were addressed in a study of POP concentrations in locally-harvested foods from four coastal communities of Chukotka, Russia in the Bering Strait region (Dudarev et al., 2019a, 2019b). This study determined levels of PCBs, various organochlorine pesticides and chlorobenzenes in fish, reindeer and marine mammals. Although the majority of compound and species combinations indicated that the most recent samples had lower or unchanged contaminant concentrations compared to earlier samples collected in 2001–2002, HCB and mirex were generally detected at higher concentrations in more recent samples (Dudarev et al. 2019a). Yet, to our knowledge, these contaminant changes over time have not been examined for links with climate parameters.

2.4.6 Links between POP time trends and climate change in Antarctica

POP monitoring and time series

Comparisons between observations reported in the Arctic and Antarctic can provide a global perspective on the similarities and differences of climate change effects on contaminant trends in polar regions, especially as the two areas have different remote contaminant sources and transport pathways. Since the 1950s, surface air temperature has increased by 3–7°C in certain regions of Antarctica (Jacka et al., 2004; Turner et al., 2005; Chapman and Walsh, 2007; Ducklow et al., 2013), and glaciers have retreated by almost 87% from the Antarctic Peninsula (Cook et al., 2005). Increased DDT exposures in Antarctic freshwater lakes (Sun et al., 2005) and marine environments (Geisz et al., 2008) have been linked to increasing glacial meltwater inputs of contaminants. Warming-related increases in vegetation (Convey and Smith, 2006) and changes in soil chemistry (i.e. increased dissolved organic carbon) (Roberts et al., 2009) may increase the capacity of soil to sequester POPs (Cabrerizo et al., 2013). This in turn will likely result in greater inputs of contaminants into aquatic systems as contaminants present in Antarctic soils are re-mobilized (Bargagli, 2008) with the increasing glacial melt projected under future climate scenarios.

Unlike the Arctic, there are no systematic POP monitoring efforts ongoing in Antarctica. To our knowledge, there are no national monitoring programmes of POPs in Antarctic organisms, nor any systematic contributions to a joint pan-Antarctic program. However, Norway established a regular air monitoring program at Troll Station in 2007. Only a few studies have explored correlations between climate parameters and POP temporal trends in Antarctic abiotic compartments (Cabrerizo et al., 2013), and biota, such as fish (Cincinelli et al., 2016) and penguins (Geisz et al., 2008). Climate change may affect the structure of Antarctic trophic webs and the availability of prey in the same way as described for the Arctic (see Chapter 2.3), and thus could have similar consequences on POP exposure and bioaccumulation in Antarctic ecosystems. In order to compare with and complement observations in the Arctic, the following discussion focuses on POP temporal trends in marine fish and Adélie penguins (*Pygoscelis adeliae*). These species are native to Antarctic waters and thus suitable to study temporal changes in contaminant exposure caused by climate change in this region.

Terrestrial environment

There are currently no known studies of associations between climate parameters and POP temporal trends in terrestrial biota available from Antarctica.

Freshwater environment

There are currently no known studies of associations between climate parameters and POP temporal trends in freshwater biota available from Antarctica.

Marine environment

Fish

A study of POP temporal trends in the muscle tissue of emerald rockcod (*Trematomus bernacchii*) from the Ross Sea, Antarctica conducted over a 30-year period from the early 1980s to 2011 observed two elevated concentration peaks within generally decreasing POP trends (Cincinelli et al., 2016). These concentration peaks occurred in 2001 and 2005 for PCBs, and in 2005 for *p,p'*-DDE and PBDEs (Corsolini et al., 2006; Borghesi et al., 2008; Cincinelli et al., 2016).

The concentration peaks were ascribed to the release of POPs trapped in iceberg B15, which calved from the Ross Ice Shelf at the beginning of 2000. This iceberg, which broke into several pieces in 2000, 2002 and 2003 (NIC, 2014), was suggested as the possible source of POPs released into seawater that contributed to the elevated POP concentrations detected in fish (Cincinelli et al., 2016). These observations provide an example of the potential effects warming-induced polar ice melt and collapse can have on POP concentration trends in biota. In the Arctic, observations of relationships between climate and POP trends in fish are limited to freshwater species where associations have been found between POP concentrations and climate oscillation indices, temperature, precipitation and variables related to permafrost thaw (Sections 2.4.2.2 and 2.4.3.2). The direct effects of iceberg calving and melting on POP availability to marine fish have not been studied in the Arctic, although such effects are plausible.

Stable or slightly increasing POP levels have been reported in benthic-feeding Antarctic fish, such as the humped rockcod (*Gobionotothen gibberifrons*) and blackfin icefish (*Chaenocephalus aceratus*) (Corsolini, 2009), whereas stable or decreasing concentrations have been observed in the more pelagic, Antarctic silverfish (*Pleurogramma antarcticum*) (Corsolini et al., 2002, 2003) and mackerel icefish (*Champsocephalus gunarri*) (Goerke et al., 2004; Weber and Goerke, 2003). Data on PCBs and DDT in icefish (*C. aceratus and C. gunnari*) collected around the Antarctic Peninsula have shown an increasing trend since the 1990s, while other POPs (e.g. HCB) have shown a stable or slightly decreasing trend (Strobel et al., 2016). Benthic fish collected between 1988 and 1996 from Terra Nova Bay in the Ross Sea also showed increasing concentrations of PCBs, *p,p'*-DDE and HCB (Focardi et al., 1992; Corsolini et al., 2002). However, it is not known if these recently increasing trends are related to climate change in any direct or indirect way. Lana et al. (2014) analyzed POP data from seventeen fish species from four Antarctic regions (Antarctic Peninsula, Weddell Sea, Ross Sea, Adélie Land) and suggested that PCB burdens in the Antarctic region had not yet reached a steady state, probably because the environmental reservoirs of POPs that continue to exist after primary emissions have declined may be re-mobilized by warmer conditions related to climate change (Cabrerizo et al., 2013).

Seabirds

The decreasing concentrations of POPs observed in Antarctic pelagic fish over time generally correspond with declining trends observed in pelagic-feeding Antarctic seabirds, such as Adélie penguins (van den Brink et al., 2011). This is evident for the development of PCB concentrations since the 1990s, but not equally clear for DDTs, which increased in the 2000s (Corsolini et al., 2011). These contrasting trends can be ascribed to the transport of organic contaminants from the pelagic system to benthic environments, which is especially efficient in the Antarctic region because of a close association with sea ice dynamics. The organic matter transport from the water-ice

interface and pack ice may be affected by climate change; more ice will melt, contributing to the release of entrapped organic matter to the water column, from where it falls down to the benthic environment and community. Moreover, benthic organisms have limited capability to metabolize these contaminants (Brockington and Peck, 2001; Focardi et al., 1992).

Geisz et al. (2008) explored the potential effects of climate change on DDT levels in Adélie penguins from the Antarctic Peninsula. They found that ΣDDT concentrations in Adélie penguin eggs from the Palmer Archipelago did not decrease from the 1970s to the 2000s, which is in contrast to the observations reported for thick-billed murres, northern fulmars, and black-legged kittiwakes in the Arctic during the same time period (Braune et al., 2001; Braune, 2007; Geisz et al., 2008). The detection of *p,p′*-DDT in the penguins, despite the bans and restrictions on its use in place since the 1970s, suggests there was a current source of the chemical to the Antarctic marine environment. A previous study indicated that there had been very little recent DDT deposition in Antarctica, however levels in Antarctic glacial meltwater were measurable (Chiuchiolo et al., 2004). Thus, glacial meltwater was proposed as a possible source of DDT to the Antarctic marine food web. This hypothesis is supported by measurement-based estimates of the amount of DDT released annually from Antarctic glacial ablation (1–4 kg ΣDDT/y) (Geisz et al., 2008).

2.4.6.1 Conclusions for Antarctica

Contrary to the Arctic where the availability of long-term monitoring data allows for the investigation of correlations between climate parameters and POPs in biota over time, POP concentrations in the Antarctic have been collected less systematically. In some Antarctic fish, POP concentrations have declined over the last two decades, however, these temporal trends appear to be weak and variable. Additionally, concentration peaks of PCBs, DDTs, and PBDEs observed in Antarctic fish were potentially related to the release of POPs after iceberg B15 calved from the Ross Ice Shelf. Comparisons between Arctic and Antarctic data can provide insight into the influence of climate change on the distribution of POPs globally, and specifically in polar environments. In addition, Antarctic studies provide supplementary information that can aid assessments of climate-related impacts on contaminants in the Arctic.

2.4.7 Conclusions and recommendations

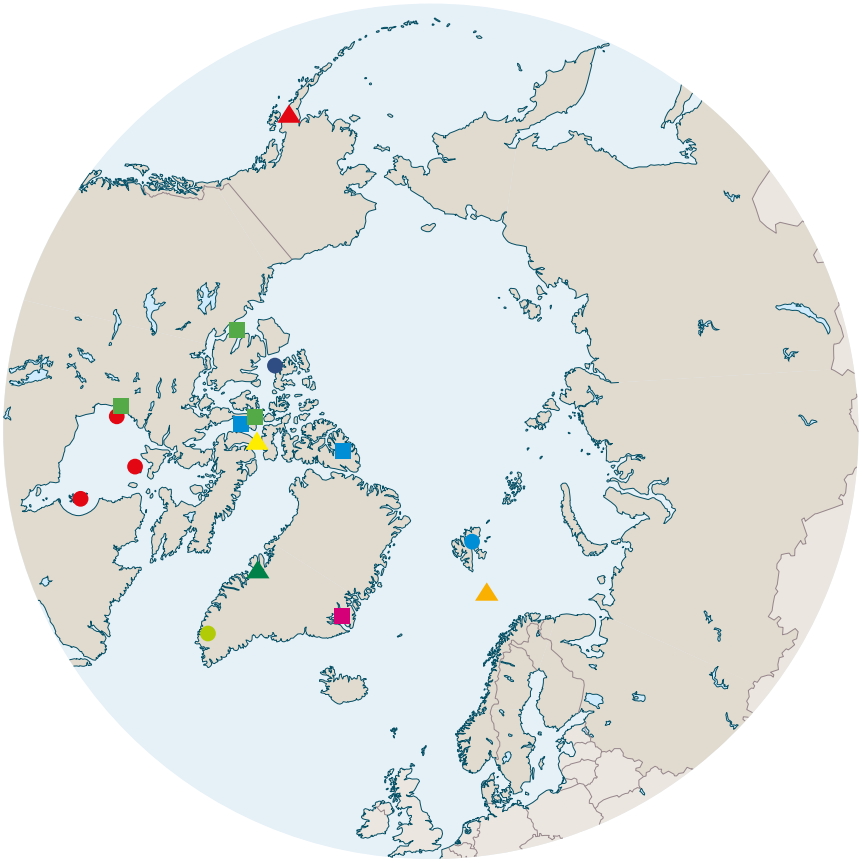
2.4.7.1 Conclusions

As documented in previous AMAP reports (e.g. AMAP, 2016b), and briefly summarized in this chapter, a number of long-term time series exist for POPs in Arctic biota including invertebrates, fish, birds and mammals representative of terrestrial, freshwater or marine environments. These datasets have now reached a length and statistical strength that allow for explorations of cross-linkages with climate parameters. However, so far, only a few studies, geographically limited to Arctic locations within the United States, Canada, Greenland and Norway, have linked contaminant monitoring data with climate parameters, leaving a great potential to extend this work and validate the findings reported so far. The majority of existing time series and related research focuses on POPs, while there is limited information on the temporal development of chemicals of emerging Arctic concern or other compounds of interest, such as POP transformation products. If time trends become available for other compounds in the future, it will be particularly relevant to assess them for associations with climate-related parameters, as their physical-chemical properties, sources and emissions may deviate from those of legacy POPs.

Using the information currently available, this chapter attempted to address the question: Can we link changes in temporal trends of POPs with climate parameters and/or food web changes? A number of studies have identified potential relationships between climate-related parameters and POP concentrations in Arctic biota. One of the first examples of this relationship was reported between the AO and annual changes in POP concentrations of glaucous gulls from Bjørnøya, Norway. Since then, a growing number of correlations have been identified between POP time trends in other Arctic biota and various climate-related parameters such as climate oscillation indices, sea ice extent, temperature and/or precipitation. However, whether these correlations really represent causal relationships between climate change and POP concentrations in biota, and the potential mechanisms underlying these correlations, are still largely unknown.

In some cases, the strength and/or direction of the relationships found between climate indicators and POP time trends in this chapter were not uniform, and varied between locations, species and compounds. For example, opposite associations between climate parameters and POPs in landlocked Arctic char were reported in studies from Canada and Greenland. In Canada, PCB levels in Arctic char were negatively linked with warming-associated increases in primary productivity, while in Greenland, PCB levels in Arctic char were positively associated with warming air temperatures. These differences highlight the potential for locally different phenomena to influence POP concentrations. In Canada, clear effects of thawing permafrost on POP levels in landlocked lakes have emerged (see also Chapter 2.2), presumably via increased inputs of particle-associated POPs, however, these have not yet been studied in other regions. On the other hand, positive associations between POP concentrations in the marine environment and climate oscillation indices (of preceding years) have been substantiated in several studies across species and locations. Reduced sea ice coverage was also associated with (higher) POP concentrations in Arctic wildlife in multiple studies from several locations, although exceptions existed.

There is some indication that shorter time series (i.e. those starting more recently), may be affected by climate change to a higher degree than longer time series. This was the case for POP trends in ringed seals from West Greenland, where monitoring started eight years after monitoring efforts in East Greenland. This observation suggests climate change could be exerting a greater influence on POPs in recent years, but this should be confirmed in other comparable sets of time series. Studies from Canada indicate a time-lag can exist between changes in climate parameters and changes in POP concentrations in Arctic biota.



- Arctic foxes and polar bears: Dietary shifts towards higher (Arctic foxes) and lower (polar bears) percentages of marine prey identified; associations between marine-based diet and higher POP concentrations observed in both species. Zooplankton: Indications of higher trophic magnification in summer because of changes in species composition.
- Glaucous gulls: Declining POP trends potentially slowed by the positive AO. Arctic char: Decreasing concentrations of PCBs seem to be associated with decreases in local glaucous gull populations as the main influencing factor.
- Ringed seals: Higher concentrations of HCB and α-HCH observed in years preceded by positive AO. Effects on temporal trends were minor. Polar bears: Dietary shifts towards more subarctic species observed, particularly in years with low NAO.
- Arctic char: Air temperature identified as an important predictor of ΣPCB and α-HCH concentrations.
- Ringed seals: Positive associations observed between sea ice extent and POPs, except α-HCH. Negative associations observed between AO and α-HCH. Effects on temporal trends were minor.
- Arctic char: Higher POP concentrations observed in years preceded by positive NAO.
- Thick-billed murres and polar bears: Associations observed between some POPs and wind speed, AO/NAO and sea ice. Trends were better described when the AO/NAO were included (with time-lags).
- Thick-billed murres: Dietary shifts increase trophic position, but have little influence on time trends. Higher POP levels observed following years with higher rainfall. Northern Fulmars: Higher POP concentrations observed in years preceded by positive NAO.
- Ringed seals: Higher POP levels observed in years preceded by positive AO. Positive associations between POPs and sea ice.
- Arctic char: Higher POP concentrations observed in lakes impacted by permafrost thawing.
- Thick-billed murres: Associations observed between Pacific Decadal Oscillation (PDO) and POP levels in eggs, potentially via changes in diet.

Figure 2.4.17 Locations of major observations linking climate-related changes to POP trends in Arctic biota.

Deviations from generally decreasing trends of POPs were seen in some species in association with ecosystem changes, and particularly with changes in predator-prey relationships. POP time trends in polar bears seemed to be influenced by dietary shifts that included more subarctic prey species with higher POP levels. Changes in trophic levels have also been observed for seabirds. Additionally, polar bear and seal movements and feeding ecologies appear to be influenced by climate-related changes to the physical environment (e.g. sea ice extent) suggesting a complexity that needs further study, as discussed in detail in Chapter 2.3.

In general, most associations between climate parameters and concentrations in biota were found for relatively volatile compounds such as HCB and α-HCH. While research biases have to be considered, as volatile compounds might be targeted specifically because of an expectation of associations with increasing temperatures, studies including both volatile and non-volatile compounds indicate that processes might be involved where volatility plays a role. Again, the underlying mechanisms linking climate change to the alterations in compound availability, uptake and/or wildlife exposure that are eventually observed as changes in long-term concentration trends in biota, are not yet completely understood. Several time trend studies reported stable, or even increasing, concentrations of HCB, in contrast to decreasing concentrations observed for other contaminants. Whether this is associated with ongoing primary emissions from the unintentional production of HCB, or due to increased emissions of HCB from secondary sources, is not known to date.

Additional data compiled from the Antarctic illustrates the potential effect of melting ice and glaciers on POP concentrations in biota. In addition to continuous ice melt, iceberg calving

events seem to be associated with POP concentrations in biota, as reflected by contaminant concentration peaks found within a generally decreasing POP trend. These effects have not been studied in the Arctic to any significant degree, but are likely to occur given the similarities in polar environments.

The results synthesized in this assessment can also be used to address a related question: Do the findings of this chapter have implications for national and international regulations of chemicals?

The long-term temporal changes of POP concentrations in the Arctic are important indicators used in effectiveness evaluations of the Stockholm Convention (AMAP, 2014). As the Stockholm Convention regulates primary emissions of POPs, the extent to which POP concentrations in Arctic biota reflect reduced primary emissions, or are influenced by secondary emissions and other biological, physical and chemical processes outside the scope of the Stockholm Convention, is important for assessing the success of chemical regulations. The results of this assessment suggest that primary emissions are still the main driver of the POP trends observed in Arctic biota. However, for some compounds in some species, climate was identified as an additional factor influencing the rate of concentration declines over time. The effects of climate on POP trends varied, resulting in either reduced or enhanced rates of change depending on the compound, species and climate variable examined.

Preliminary results suggest that changes in POP trends in biota lag behind climate-related changes to the Arctic environment and ecosystems. Consequently, we may now only be seeing the beginning of potentially larger climate influences on POP concentrations in Arctic biota. As specified in the recommendations offered below, it will therefore be essential to continue monitoring POPs, climate parameters and biological data, and improve attempts to link these variables together to obtain a better understanding of their complex interactions and also support Stockholm Convention effectiveness evaluations.

Other findings highlighted in this review are also relevant to national and international regulations. In general, POP concentrations have uniformly declined across many Arctic species and locations over the last few decades, while the effects of climate change on POP trends appear variable, and seem to differ between locations. These varied effects, observable at the regional or local level, potentially necessitate more species- and location-specific assessments. As discussed in Chapter 2.3, ecosystem responses to climate change may vary between and within species and regions, calling for approaches that can provide a nuanced understanding of climate change and POP interactions. Furthermore, there are indications that compound-specific differences in environmental fate and long-term accumulation in biota might become more pronounced over time. Climate-related effects have mainly been found for the more volatile compounds such as the HCHs and HCB, the latter being consistently observed as one of the few POPs not steadily decreasing in Arctic biota over time. This could be the result of ongoing primary emissions and/or climate-related effects that would need further study.

In summary, while the effects of climate change on long-term POP trends in biota may still be subtle compared to the effects of primary emissions reductions, there are indications that climate-related influences do exist and may become increasingly important over time. Associations have been identified between POP time trends in biota and physical parameters and ecosystem changes affected by climate change.

2.4.7.2 Recommendations

More time series for POPs in the Arctic should be analyzed for associations with climate-related parameters to extend current findings and achieve larger geographical coverage. The continuation of long-term monitoring efforts for POPs, and other relevent climate and biological data is essential, as the results of this assessment suggest that the observable influences of climate change on POP trends are recent and occur with lag-phases. This means the effects of climate that are currently subtle or non-existent may become more pronounced in the future.

The research examining associations between POP trends and climate parameters has so far been limited to a few legacy POPs, with a potential bias towards the most volatile compounds. Theses analyses should be expanded to include other contaminants, including chemicals of emerging Arctic concern, while ensuring that differences in physical-chemical properties are considered. Attempts should be made, perhaps in combination with modeling approaches, to study the importance of specific physical-chemical properties (e.g. volatility, water solubility) in mediating the effects of climate change on the development of POP concentrations in the Arctic environment over time.

A better use of existing data and research to elucidate associations between climate parameters and contaminant trends is recommended to improve our understanding of the processes and mechanisms underlying these relationships. Oscillation indices and sea ice coverage seem to be two parameters of recurring significance whose impact on POP trends in biota need further study.

Observations vary between locations, therefore, more knowledge of the influence of climate change on POP concentrations at different spatial scales is needed, including the identification of local phenomena. This can be achieved with coordinated approaches (e.g. comparing datasets between locations; duplicating study designs in new locations). Time trend assessments should recognize that climate change-related influences may occur locally and potentially introduce larger variations between time series from different locations.

Observations also vary between species, and the complex interactions between changes in the physical environment and changes in ecosystem that can occur are discussed in detail in Chapter 2.3. Markers of trophic level and dietary sources (e.g. stable isotope and fatty acid signatures) are two examples of biological factors that should be included in POP monitoring programme protocols, and POP time trends should be adjusted for these parameters.

Assessments of POPs in the Arctic should note the complementary information available from the Antarctic and take relevent findings into consideration when developing future research and monitoring initiatives for the Arctic.

Chapter 2.4 Appendix

Table A2.4.1: Studies investigating POP temporal trends in Arctic and Antarctic biota in relation to climate parameters.

Location	Species	Years	Contaminants	Climate-related parameters	Associations	Reference
Freshwater biota						
Canada	Burbot	2000-2012	PCBs, DDTs	Temperature (air)	Positive associations between primary productivity (driven by higher temperatures) and concentrations of ΣDDT and PCB congeners and homolog groups, but not over the entire timespan	Carrie et al., 2010
Canada	Arctic char	1989-2015[a]	ΣPCBs, ΣDDTs, HCB, ΣHCHs, toxaphene	Climate indices (NAO) Temperature (air), Precipitation	Positive association between ΣHCH and precipitation; positive associations between ΣPCBs, ΣDDTs, ΣHCHs and NAO in the preceding year	Cabrerizo et al., 2018
Canada	Zooplankton, Arctic char	2008-2017	PCBs, CHLs, DDTs	Turbidity (proxy for permafrost thaw)	Higher concentration of POPs in biota from lakes with higher carbon and particulate concentrations	Cabrerizo et al., 2019b; Muir, pers. comm.
Greenland	Arctic char	1999-2017	ΣPCBs, PCB-52, PCB-153, ΣDDTs, α-HCH, HCB	Biological variables ($\delta^{15}N$), Climate indices (AO), Temperature (air)	Positive association between ΣPCBs, α-HCH and air temperature	Rigét et al., 2020
Norway (Bjørnøya)	Arctic char	1998-2017	ΣPCBs, HCB	Temperature (air)	Possibly positive associations between HCB and air temperature, but indications of confounding factors	Blévin et al., pers. comm.
Terrestrial mammals						
Norway (Svalbard)	Arctic fox	1997-2013	PCBs, PBDEs, CHLs, *p,p′*-DDT, *p,p′*-DDE, β-HCH, HCB, mirex	Biological variables ($\delta^{13}C$, $\delta^{15}N$), Sea ice (coverage; proxy for seal availability), Reindeer mortality	Positive association between POPs and percentage marine-based diet; positive association between β-HCH and sea ice cover; negative association between HCB and reindeer mortality	Andersen et al., 2015
Norway (Svalbard)	Arctic fox	1997-2014	PFAS	Biological variables ($\delta^{13}C$, $\delta^{15}N$), Sea ice (coverage; proxy for seal availability), Reindeer mortality	Positive association between PFOS and sea ice cover; negative association between PFCAs and reindeer mortality	Routti et al., 2017
Marine fish						
Antarctica	Emerald rockcod	1981-2010[a]	PCBs, PBDEs, PCDD/Fs, CHLs, DDTs, HCHs, HCB	Iceberg calving event	Increase of concentrations after iceberg calving event	Cincinelli et al., 2016
Seabirds						
Antarctica	Adélie penguins	1964-2005	DDTs	Glacier melt timing	Stable concentrations of ΣDDT related to release from glacier meltwater	Geisz et al., 2008
Norway (Svalbard)	Glaucous gull	1997-2006	ΣPCBs, HCB, oxychlordane	Climate indices (AO)	Positive associations between ΣPCBs, HCB, oxychlordane and AO in preceding summer and winter; negative associations between ΣPCBs, HCB, oxychlordane and AO in current winter	Bustnes et al., 2010
Canada	Thick-billed murre	1975-2013[a]	ΣPCB, *p,p'*-DDE, HCB, oxychlordane, heptachlor epoxide, dieldrin	Biological variables ($\delta^{15}N$)	Positive association between trophic position and rate of POP decline	Braune et al., 2015a
Canada	Thick-billed murre	1975-2015[a]	PCBs, CHLs, CBz, DDTs, OCS, mirex, photomirex	Climate indices (AO, NAO), Precipitation, Sea ice (extent), Temperature (air, air temperature anomalies, sea surface)	Positive associations between some PCBs, *p,p′*-DDE, chlorobenzenes, OCS, dieldrin and precipitation; negative associations between PCB-170, PCB-180, oxychlordane, *cis*-nonachlor, heptachlor epoxide and precipitation	Foster et al., 2019
Canada	Thick-billed murre	1993-2015	PCB-153, PBDE-47, PFOS, *p,p′*-DDE, α-HCH	Climate patterns (AO, NAO), Precipitation, Sea ice (coverage, freeze-up/break-up dates, ice-free days)[a], Sea level pressure[a], Temperature (air, land surface), Wind speed	see Table 2.4.2	Morris et al., 2021

Location	Species	Years	Contaminants	Climate-related parameters	Associations	Reference
Canada	Northern fulmar	1975-2015[a]	PCBs, CHLs, CBz, DDTs, OCS, mirex, photomirex	Climate indices (AO, NAO), Precipitation, Sea ice (extent), Temperature (air, air temperature anomalies, sea surface)	Positive association between chlorobenzenes, *cis*-/*trans*-nonachlor, dieldrin, mirex, photomirex and NAO	Foster et al., 2019
United States (Alaska)	Thick-billed murre, Common murre	1999-2010	ΣPCBs, *p,p′*-DDE, HCB, α-HCH, *cis*-nonachlor, dieldrin, heptachlor epoxide, oxychlordane, mirex	Climate indices (PDO)	POP concentrations in thick-billed murres, but not common murres, were highest when PDO was near 0 and lowest during PDO extremes	Kalia et al., 2021
Marine mammals						
Canada	Beluga	1989-2015	PCBs, CHLs, DDTs, HCB, HCHs, dieldrin, mirex	Biological variables ($\delta^{13}C$, $\delta^{15}N$)	Associations between ΣPCBs and $\delta^{13}C$; associations between dieldrin, mirex and both $\delta^{13}C$ and $\delta^{15}N$	Noël et al., 2018
Canada	Polar bear	1991-2007	ΣPCBs, ΣPBDEs, ΣCHLs, ΣDDTs, α-HCH, β-HCH	Biological variables ($\delta^{13}C$, $\delta^{15}N$, fatty acid signatures), Sea ice (time of breakup)	Associations between POP concentrations and diet composition; associations between time of sea ice breakup and diet composition	McKinney et al., 2009
Canada (Hudson Bay)	Polar bear	WHB[c]: 1991-2015 SHB[d]: 2007 or 2008-2015	PCB-153, PBDE-47, PFOS, *p,p′*-DDE, α-HCH	Climate indices (AO, NAO), Precipitation, Sea ice (coverage, freeze-up/break-up dates, ice-free days), Sea level pressure[a], Temperature (air, land surface), Wind speed	see Table 2.4.2	Morris et al., 2021
Canada	Ringed seal	1993-2008	PCBs, CHLs, DDTs, HCB, HCHs, dieldrin, mirex, toxaphene	Sea ice (time of breakup)	Higher concentrations of some PCBs and *p,p′*-DDT in years with earlier sea ice breakup	Gaden et al., 2012
Canada (multiple locations)	Ringed seal	1972-2016[a]	ΣPCBs, ΣCBz/HCB ΣCHLs, ΣDDTs, ΣHCHs, oxychlordane, trans-nonachlor[b]	Climate indices (AO, NAO, PNA), Sea ice (coverage), Temperature (air)	Positive associations between POPs and AO/NAO of preceding year; negative associations between POPs and year of sampling for most locations, but with variation between locations; positive associations between ΣPCB and sea ice coverage; positive association between ΣCBz and sea ice coverage	Houde et al., 2019
Norway (Svalbard)	Polar bear	2000-2014	PFAS	Biological variables ($\delta^{13}C$, $\delta^{15}N$), Sea ice (habitat quality; proxy for seal availability)	Positive associations of most PFAS with trophic level / intake of marine diet; positive associations of most PFAS with higher quality habitat	Routti et al., 2017
Norway (Svalbard)	Polar bear	1997-2017	ΣPCBs, ΣOH-PCBs, PBDE-47, PBDE-153, *p,p′*-DDE, HCB, β-HCH, oxychlordane	Biological variables ($\delta^{13}C$, $\delta^{15}N$)	Positive associations of POPs with trophic level / intake of marine diet	Lippold et al., 2019
Greenland	Polar bear	1984-2011	ΣPCBs, ΣPBDEs, PFAS, ΣCHLs, ΣDDTs, HCB, ΣHCHs, OCS, dieldrin, mirex, toxaphene	Biological variables ($\delta^{13}C$, $\delta^{15}N$, fatty acid signatures)	Associations between POP concentrations and diet composition	Dietz et al., 2021
Greenland (East)	Ringed seal	1986-2017	ΣPCB, PCB-52, PCB-153, ΣDDT, HCB, α-HCH	Biological variables ($\delta^{15}N$), Climate indices (AO), Sea ice (coverage), Temperature (seawater)	Positive association between all compounds and $\delta^{15}N$; positive association between HCB, α-HCH and AO; positive association between HCB and seawater temperature	Rigét et al., 2020
Greenland (West)	Ringed seal	1994-2017	ΣPCB, PCB-52, PCB-153, ΣDDT, HCB, α-HCH	Biological variables ($\delta^{15}N$), Climate indices (AO), Salinity, Sea ice (coverage), Temperature (seawater)	Positive association between PCB-52 and $\delta^{15}N$; negative association between α-HCH and AO; positive association between ΣPCB, PCB-52, PCB-153, ΣDDT and temperature; positive association between ΣPCB, ΣDDT and salinity; positive association between sea ice coverage and all compounds except α-HCH	Rigét et al., 2020

AO: Arctic oscillation; CBz: chlorobenzene; CHL: chlordane; DDE: dichlorodiphenyldichloroethylene; DDT: dichlorodiphenyltrichloroethane; HCB: hexachlorobenzene; HCH: hexachlorocyclohexane; NAO: North Atlantic Oscillation; OCS: octachlorostyrene; OH-PCB: hydroxylated polychlorinated biphenyl; PBDE: polybrominated diphenyl ether; PCB: polychlorinated biphenyl; PCDD/F: polychlorinated dibenzo-*p*-dioxins and furans; PDO: Pacific Decadal Oscillation; PFAS: per- and polyfluoroalkyl substance group; PFOS: perfluorooctane sulfonic acid; PNA: Pacific/North American Pattern; [a]Varying between locations and/or compound groups; [b]Multiple other compounds analysed, but not included in analysis with climate parameters; [c]Western Hudson Bay; [d]Southern Hudson Bay.

2.5 Involvement of Arctic indigenous communities in POPs and climate change research

Authors: Derek Muir, Magali Houde, Eva-Maria Krümmel

2.5.1 Introduction

This chapter addresses the question: *How can Indigenous Knowledge (IK) contribute to the discussion of climate-related effects on trends of persistent organic pollutants (POPs)*? To address this question, we provide an overview of ongoing, long-term POPs monitoring research utilizing fish and wildlife species of importance to Arctic indigenous communities. Examples of indigenous community involvement in long-term contaminant studies are provided, along with examples of how climate-related observations collected at the community level can serve as an interface between IK and POP trends. A broader exploration of indigenous perspectives, consultations, and communications related to community-based contaminant projects is included in the Arctic Monitoring and Assessment Programme (AMAP) Mercury Assessment (AMAP, 2021). There are many issues in common with regard to community involvement, use of IK, and communication of results for POPs and mercury trends, therefore readers are encouraged to consult this resource for additional information, including definitions of IK and recommendations for its effective utilization (Krümmel and Houde, 2021).

Indigenous peoples from several regions of the Arctic have been involved in environmental monitoring and research on POPs to varying degrees, spanning from limited participation in projects (e.g. performing sample collection), to full partnership in the research, as exemplified by a co-production approach in which IK holders participate in all stages of a project. In most cases, the involvement of indigenous communities has centered around the collection of fish, caribou/reindeer, seabird, and marine mammal samples. The geographical extent of this involvement can be seen in a temporal trend study by Rigét et al. (2019) which analyzed 1074 long-term time series of contaminant concentrations in a wide range of Arctic biota. This review showed at least 16 indigenous communities, widely spread throughout the circumpolar region, have been directly involved in Arctic wildlife contaminants research for over two decades (Figure 2.5.1). Many of the time series analyzed by Rigét et al. (2019) are also discussed in Chapter 2.4 of this assessment in association with climate-related parameters, and thus exemplify the important contributions indigenous communities have already made towards understanding climate-related changes in Arctic contaminants.

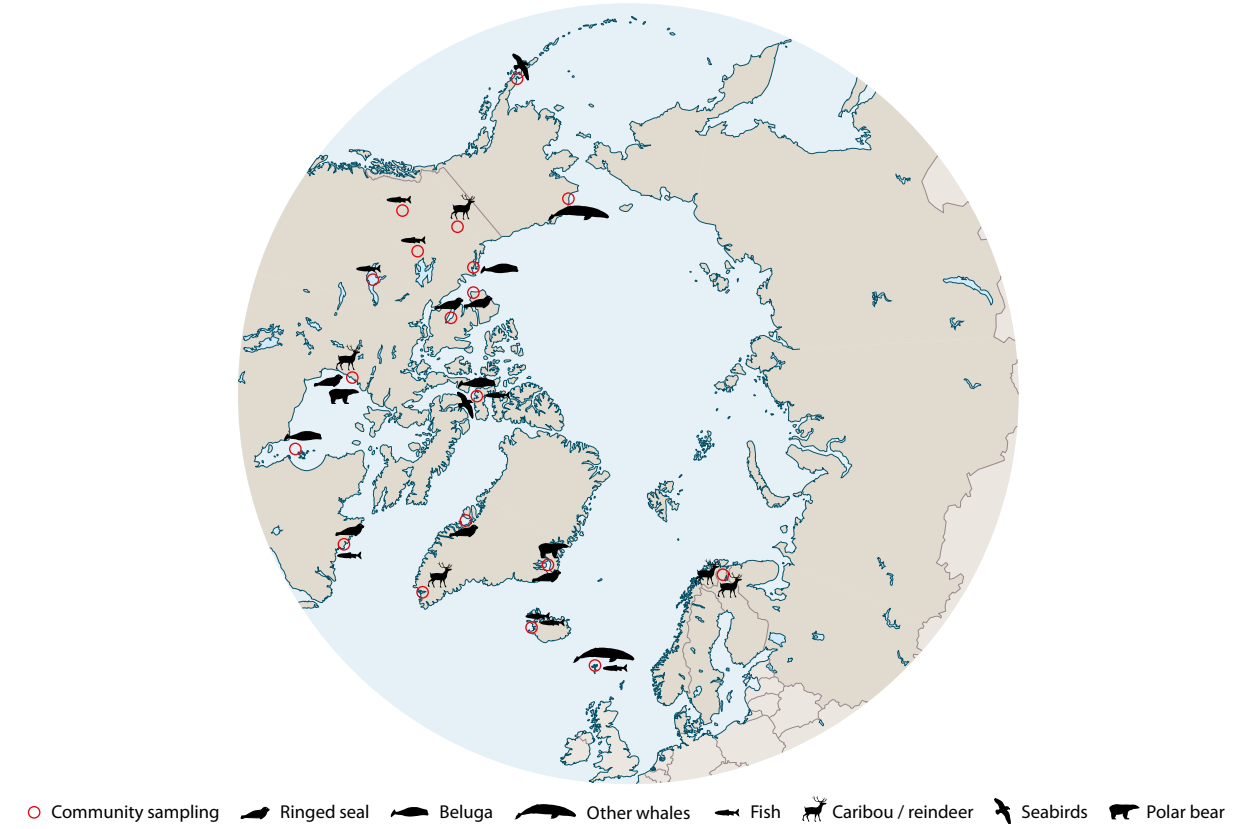


Figure 2.5.1 Northern communities involved in sample collection for long-term monitoring of POPs. These communities are contributing, or could contribute, Indigenous Knowledge on Arctic ecosystems, species, environmental conditions, and climate-related changes. Redrawn from Rigét et al. (2019).

To date, the involvement of indigenous peoples in contaminants research has mainly been limited to sample collection efforts. In most cases, community members have collected wildlife samples as part of traditional hunting and fishing activities, although there has also been involvement in water and air sampling. Hunters and fishers directly influence the research by making decisions on when, where, and which animals to harvest, by taking tissue samples, and recording information on the animals (e.g. size, sex, body condition). While this does not constitute equitable participation of indigenous peoples in research, without their participation, these long-term contaminant studies would not be feasible, especially in Alaska, Canada, and Greenland. Additionally, indigenous community involvement provides economic and logistical benefits for research by reducing the time and costs associated with travel and fieldwork, and increasing opportunities for capacity building.

As Lennert (2016) has noted, hunters and fishers in northern communities represent a rich repository of environmental knowledge that can enhance scientific research and observations of a changing Arctic. The United Nations Framework Convention on Climate Change technical paper on the best practices for the use of IK (UNFCCC, 2013) calls for "*involving indigenous and local knowledge in all assessment phases, from conception through to outputs*". Viewed through that lens, the utilization of IK and involvement of community members in the leadership and design of temporal trend studies on POPs has been very limited to date. Similarly, a global survey of climate research involving indigenous communities noted that the vast majority of studies used IK systems with minimal participation or decision-making by the communities that provided the knowledge (David-Chavez and Gavin, 2018). Thus, the level of indigenous involvement in Arctic contaminants and climate change research is in a similar situation and needs improvement.

The growing need for research on POPs in the Arctic to consider climate-related effects provides an excellent opportunity for studies to utilize climate- and food web-related IK, and for northern indigenous communities to assume more leadership in research. Such involvement would help to sustain projects over decadal time scales and ensure monitoring activities address local community needs.

Community-led projects are one approach for increasing the involvement of indigenous peoples in research. In these projects, the research addresses a concern identified by the local community, and the community is involved in all stages of the project, including planning, data collection, interpretation and analysis, the dissemination and communication of results, and decision-making. This has already happened to some extent in the case of long-term mercury trends in biota, as reviewed in the AMAP Mercury Assessment (AMAP, 2021). Mercury exposure via the consumption of fish and wildlife is a very common concern for communities across the Arctic, and the costs of mercury analysis are relatively low compared to those of POPs. Thus, community-based monitoring projects have tended to give priority to mercury research. The AMAP Mercury Assessment (AMAP, 2021) has identified over 40 community-based projects, many of which are designed and conducted by indigenous communities in collaboration with government or university-based scientists.

The communities identified in Figure 2.5.1 are those in which long-term monitoring of POPs in biota has been previously, and/or is currently, being conducted, with 'long-term' defined by Rigét et al. (2019) as monitoring that began before the year 2000 with continued participation in more than six sampling years. Contaminant data, along with IK from these participating communities, has been or could be used for assessing POP-climate interactions in the future.

2.5.2 Community involvement in long-term trend studies on POPs

There are various forms of community-based monitoring undertaken in circumpolar countries. Johnson et al. (2015) noted that the term 'community-based monitoring' is more widely used in Alaska, Canada and Greenland, than in Scandinavia. While the term 'community' is often interpreted as a permanent settlement, at the project-level it can refer to 'communities of practice', such as hunters or fishers, and the local or systematic knowledge that they hold. The article by Johnson et al. (2015) builds on an initiative conducted under the Sustaining Arctic Observing Networks (SAON) which surveyed 81 community-based programs across the circumpolar Arctic (Johnson et al., 2016). That initiative also led to the development of a searchable online database, the 'Atlas of Community-Based Monitoring and Indigenous Knowledge in a Changing Arctic' (www.arcticcbm.org). The atlas serves as an inventory of initiatives and assists with network building and identification of best practices and challenges for the field. While the atlas was initially developed with a focus on environmental monitoring programs, environmental and social change in the Arctic are significantly connected. Many communities take a holistic perspective on change and emphasize connections and linkages between the environment and society. The atlas was designed to be flexible to allow mapping of social and environmental monitoring initiatives. Several community-based projects listed in the database include research on contaminants, however, POP-focused projects are not specifically identified. While it is difficult to point to particular studies of long-term POPs trends that involve communities in the study design and utilization of IK, there are examples of close collaborations which have resulted in strong time series. Several examples are provided below.

A very good example of a project involving indigenous communities and utilizing IK in research is the long-term dataset collected from subsistence hunts of bowhead whales (*Balaena mysticetus*) by Alaskan native hunters in the southern Beaufort/Chukchi Sea (Figure 2.5.1). Landed whales are regularly measured and inspected (postmortem) by indigenous hunters working alongside biologists and veterinarians from the Department of Wildlife Management of the North Slope Borough (Barrow, AK). Bolton et al. (2020) reported the first temporal trend study of POPs in bowhead whales from samples collected by hunters annually from 2006–2015. Although associations with climate-related variables were not directly evaluated in the study, ancillary data (e.g. stable isotopes, percent lipid in blubber samples) were collected that could be used along with hunter's observations in future assessments of climate and POP interactions. In a separate study conducted

in the Bering-Chukchi-Beaufort region, surveys of community members identified sea ice loss, shoreline erosion, and shifting distributions of animals as important changes (Huntington and Eerkes-Medrano, 2017) that could affect trends of POPs in biota. However, it is worth noting that interviews with bowhead whale hunters revealed that from their perspective, climate change is a secondary concern, as issues related to hunting quotas are perceived as more important (Huntington and Eerkes-Medrano, 2017).

In Greenland, the Scoresby Sound (Ittoqqortoormiit) polar bear project is one of the longest wildlife time series in the Arctic, spanning over 25 sampling years from 1983 to the present. Long-term datasets of POP levels in polar bears (*Ursus maritimus*) and ringed seals (*Pusa hispida*) were possible because of ongoing collaborations with Inuit communities in East Greenland (Rigét et al., 2013b; Dietz et al., 2019; Rigét et al., 2020). Polar bear harvests are regulated with hunting quotas, and only full-time hunters are allowed to collect the animals. Thus, all research sampling is conducted in close collaboration with skilled Greenland Inuit hunters. Research results are often presented to Inuit hunters at meetings held during fieldwork. Moreover, the scientists involved in the project are in year-round contact with hunters to mutually exchange information on relevant issues related to the hunts, contaminant exposure data, and human and wildlife health issues.

In Inuit Nunangat (the Canadian homeland of Inuit), community members are essential partners in the study of contaminants in polar bears. Hunters lead all sample collection efforts (Letcher et al., 2017), and as part of broader studies examining polar bear ecology in the Canadian Arctic, their IK provides important information on bear population sizes and trends, health and behavior, as well as insight into the perceived effects of changing climate and ice conditions on animal populations (Henri et al. 2010; Wong et al., 2017). Community engagement for the project on trends of POPs in polar bears has included annual visits with groups of hunters, trappers and other stakeholders, workshops involving hunters, community members, youths, and college students (funded by the Northern Contaminants Program) and the creation of plain language project summaries to make research results accessible to the community (Letcher, 2017).

In the Inuvialuit Settlement Region (Mackenzie Delta/East Beaufort Sea region of the Northwest Territories, Canada), the annual beluga whale (*Delphinapterus leucas*) harvest has included a long-term collaboration of community members with government scientists since the 1970s. Hunters provide biological and hunt-related data to Inuvialuit hired as seasonal monitors for the duration of the whaling season (Harwood et al., 2014). Monitors collect samples from harvested belugas to be analyzed for health parameters, including stress indicators and contaminants since relatively high levels of polychlorinated biphenyls (PCBs) continue to be a concern for beluga health (Noël et al., 2014). Inuvialuit IK of beluga body condition, health, and disease has been collected using semi-structured questionnaires and included in an assessment of beluga whale health (Ostertag et al., 2018). Long-term temporal trends of POPs in the beluga have also been studied (Noël et al., 2018; Smythe et al., 2018), although they have not yet been investigated for associations with climate parameters or food web changes.

Similarly, in the Faroe Islands, long-finned pilot whale (*Globicephala melas*) samples have been collected for contaminants research in connection with traditional whale hunts. Pilot whales have been an important part of Faroese life for centuries, both in regards to food and culture, and remain so, despite concerns about dietary exposure to contaminants (Fielding, 2010; Weihe and Debes Joensen, 2012). Tissue sampling takes place after the harvest and before meat and blubber distribution to community members (Andreasen et al., 2019). Long-term trends of polybrominated diphenyl ethers (PBDEs) in pilot whale blubber, spanning eight collection years from 1986–2013 have been reported (Bjurlid et al., 2018), but not yet linked to climate parameters.

Contaminant trend studies on ringed seals in Canada and Greenland also rely on Inuit hunters to collect samples for research each year (Rigét et al., 2013a; Houde et al., 2019). Hunters are supplied with sampling kits for individual tissues, and they record biological data, including the length, girth, blubber thickness, sex, and weight of each animal. The studies are a part of larger efforts to understand ringed seal ecology (Kovacs, 2014). A workshop held in Canada in 2014 brought together seal harvesters and scientists to discuss the broader context of community engagement in research on ringed seals (McCarney et al., 2014). Recommendations from this workshop included strengthening connections between researchers and hunters and improving the communication of research results (e.g. posters, social media, video conferencing). Since 2016, annual outreach activities have been integrated into monitoring activities for the four Canadian communities involved in ongoing ringed seal sampling. Workshops provide opportunities for Inuit elders and hunters to share their knowledge in seal ecology, traditional methods for butchering seals and preparing seal skin, and identifying abnormalities in harvested game, as well as discuss their observations about the changing environment with students and researchers. By involving hunters, elders, community members, local schools, and college students, the social component of this project addresses a shared interest among Inuit and scientific researchers in enhancing communication and community capacity building related to contaminants research (Houde et al., 2017b; Henri et al., 2020).

2.5.3 Climate change observations at the community level

The perspectives of Arctic indigenous peoples on climate change in the Arctic have been included in several recent AMAP assessments. The topic was reviewed in the Arctic Climate Impact Assessment (Huntington and Fox, 2005), and IK of climate change impacts on sea ice was included in the Snow, Water, Ice and Permafrost in the Arctic (SWIPA) report (Meier et al., 2011). The recent Adaptation Actions for a Changing Arctic (AACA) reports include chapters that discuss the perspectives of northern indigenous and local communities related to climate change and community engagement in the Bering-Chukchi-Beaufort region (Huntington and Eerkes-Medrano, 2017), Baffin Bay-Davis Strait (Ford et al., 2017), and the Barents Sea region (Degteva et al., 2017). In their review of the perspectives of the Sámi people of northern Scandinavia and the Kola Peninsula of Russia, Degteva et al. (2017) note that local and ecological knowledge is being digitally mapped and

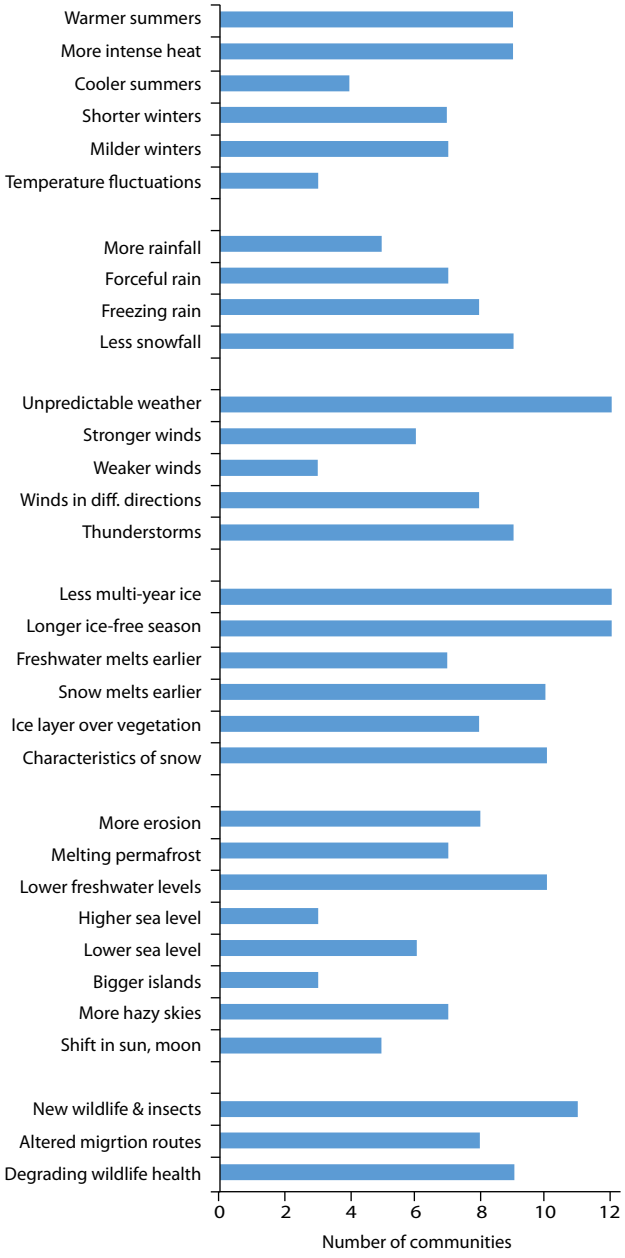


Figure 2.5.2 Frequency of climate-related environmental observations reported in surveys of 12 Inuit communities located across the western and central Canadian Arctic. Source: Gaden and Stern (2015).

modeled as a way to communicate traditional knowledge. This approach, which has also been widely used in Alaska, Canada, and western Greenland (Engler et al., 2013; Lennert, 2017; Robards et al., 2018) represents a way to record and preserve IK while identifying important ecological areas. A full review of this approach is beyond the scope of this chapter, but there is clear potential for the mapping of IK to aid the interpretation of spatial and temporal trends of contaminants and identify climate change impacts at local- and regional-scales.

The AACA reports for the Bering-Chukchi-Beaufort and Baffin Bay-Davis Strait regions (Huntington and Eerkes-Medrano, 2017; Ford et al., 2017) both cite a report by Gaden and Stern (2015) on climate and environmental observations made by Inuit in Canada which may be relevant for research on climate and contaminant interactions. Gaden and Stern (2015) summarized climate change observations collected from 12 Inuit communities in the Inuvialuit Settlement Region and the Kitikmeot region of Nunavut, a large area stretching from the Canada-Alaska border to northern Hudson Bay. These observations were recorded in a series of community-based surveys conducted between 2000 and 2011 and ultimately published as 17 separate studies (Gaden and Stern, 2015). A total of 32 general observations were reported as summarized in Figure 2.5.2. The changes most consistently reported included warmer summers and/or more extreme summer temperatures, more variable and unpredictable weather, longer ice-free seasons, less multi-year ice, earlier snow melt, lower freshwater levels, and the presence of new wildlife and insect species.

Cuerrier et al. (2015) studied the IK of climate change in three Inuit communities in Nunavik (northern Québec, Canada) using semi-structured interviews of 46 people (19 males and 27 females between the ages of 44 and 87; median age of 63). From these interviews, researchers extracted 30 environmental observations related to seasonal changes, weather, soil, plants, and insect and wildlife abundance. The observations of these communities were generally very similar to those recognized by the communities of the western and central Canadian Arctic (Figure 2.5.2; Gaden and Stern, 2015). Cuerrier et al. (2015) coded the responses and then applied statistical analyses to evaluate whether observations were significant and if they differed among communities. They proposed this as an approach to translate IK into quantitative evidence for decision-making and environmental policy development. There is obvious potential for this, or a similar approach, to bridge IK of environmental changes (e.g. ice conditions or marine food webs) with contaminant trends.

Although the focus of this review is on the contribution of IK to furthering the understanding of the effects of climate change on POP trends, it should be recognized that the foremost interests of community members may be on well-being and food security (i.e. the health and wellness of the people and environment, and the safety of traditional foods). The interests of community members deserve equal consideration to those of the broader scientific community and policymakers (e.g. whether trends are demonstrating the effectiveness of the Stockholm Convention). However, the complex interactions of climate change and contaminants make the interpretation of results, as they pertain to local communities, challenging. Indigenous communities may be facing dietary transitions because of reduced access to traditional foods or foods lower in contaminants. For example, the availability of certain fish and wildlife species that are the traditional foods of indigenous communities may become limited due to the direct and indirect effects of climate change. In turn, this would affect dietary options and thus, the level of contaminant exposure to indigenous communities. In this regard, the AMAP Human Health assessment (AMAP, 2015) recommended the development of community-based environmental monitoring networks to provide the data needed to better understand the effects of global climate change on the movement of contaminants and changing contaminant exposure of Arctic communities.

2.5.4 Conclusions and recommendations

The above examples serve to illustrate that IK is already an important component of ongoing long-term temporal trend studies on POPs in Arctic biota. However, the inclusion of northern communities in this research to date has mainly been through participation in sampling efforts, rather than involvement in the leadership and development of studies, data analysis, and communication of results. The climate-related information used in these studies has generally been provided by meteorological data websites; for example, sea ice data has been mainly obtained by satellite imagery although there are local programs conducted by northern community members that monitor sea ice thickness and date of sea ice break-up (Environment Canada, 2020). Daily observations of climate-related parameters have been collected by Arctic indigenous peoples for centuries and are part of their IK. These ongoing observations of sea ice and climate, animal distribution, behavior, diet, body condition and many other variables, would substantially contribute to the interpretation of POP trends by enhancing information on changes in local conditions not reflected in circum-Arctic scale climate indices. The challenge is to find a way of interfacing observational information with POPs data. Approaches such as mapping important ecological areas or systematically recording the knowledge of community members are already used to assess the impacts of resource development and for managing wildlife harvests, and thus may provide solutions to overcome this challenge. In many cases, information is already being collected as part of community-based projects focused on understanding the impacts of climate change (Johnson et al., 2015, 2016; Laidre et al., 2018b; Sawatzky et al. 2020), but additional capacity building is needed. Community-based projects such as SIKU: The Indigenous Knowledge Social Network, allow the sharing of knowledge and observations by Inuit hunters in near real-time. The incorporation of this, and other culturally relevant tools that allow Inuit to interpret results using their own knowledge system (Heath et al., 2018), may be a means to bridge IK and scientific knowledge in contaminant studies.

Recommendation: For IK to be used for interpreting POP trends and the impacts of climate change, additional capacity building is needed to facilitate the full engagement (and co-leadership) of indigenous peoples in the research process. Community involvement in monitoring and research needs to be much broader in order to provide a strong interface with contaminant information.

Recommendation: Going forward, Arctic indigenous peoples should both lead and be involved in all stages of study design and execution from the outset, including planning, sampling, analysis, and communication of research results.

3. Conclusions and recommendations

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3.1 Overview

This chapter presents a set of general conclusions and recommendations derived from the present assessment organized around the policy-relevant science questions outlined in Chapter 1. Detailed conclusions and recommendations can be found in the individual subchapters of Chapter 2. Here, we focus on conclusions related to the key questions of this assessment, beginning with the profound importance of the physical and ecological changes that influence exposure to persistent organic pollutants (POPs) and chemicals of emerging Arctic concern (CEACs) in a warming Arctic. Recommendations for further research and monitoring are shown in italics directly below the relevant conclusion. The key findings of this assessment are summarized in Box 3.1. This chapter also provides a list of knowledge gaps in relation to the effects of climate change on the transport of POPs and CEACs to the Arctic and their fate within the Arctic. Policy implications for the results and conclusions of this assessment are given at the end of this chapter.

This assessment has reviewed existing information on the effects of climate change on contaminants (POPs and CEACs) in the Arctic. Previous reports, including a joint assessment produced by the United Nations Environment Programme (UNEP) and Arctic Monitoring and Assessment Programme (AMAP) on POPs and climate (UNEP/AMAP, 2011), as well as an earlier AMAP (2003) report on the influence of climate change on contaminant pathways, considered most of the same issues that are addressed in Chapters 2.1 to 2.4 of the present report. However, at the time of the writing of those reports, few empirical data were available to support the predictions and hypotheses related to the possible impacts of climate change on POPs in the Arctic. This was not the case as of 2020. Over the past ten years, a growing number of studies have addressed the consequences of climate change for the fate of Arctic contaminants, either targeting specific aspects such as correlations between POP temporal trends in air or biota and climate indices (see Chapter 2), or reviewing the current state of knowledge (McKinney et al., 2015; Ma et al., 2016). In

Box 3.1 Key findings

Emissions and long-range transport of POPs and CEACs under climate change

- Concentration changes of POPs in Arctic air and ocean waters driven by climate change are predicted to be small compared to the reductions in concentrations that can be achieved by reducing primary emissions.
- Greater rates of primary emissions and secondary re-emissions of many POPs can be expected globally under climate change due to temperature-driven increases in volatilization.
- Increases in volatilization and degradation are two counteracting climate-sensitive processes affecting chemical concentrations in Arctic air. Both are expected to increase under higher temperatures related to climate change.
- Other climate change-related developments and events likely to affect POP sources and emissions include possible increases in economic activity and population growth in the Arctic.

Effects of climate change on levels and trends of POPs and CEACs in the physical environment

- Climate change perturbations are resulting in the re-mobilization of POPs within and between air, water, ice, snow, soils, and sediments in the Arctic. Examples include increased POP concentrations in lake sediments resulting from melting glaciers and permafrost slumps.
- Increasing concentrations of some POPs in Arctic air in recent years have been attributed to release from melting ice and to increased volatilization due to the diminished extent of sea ice.
- Increased primary productivity under climate change is enhancing the drawdown and transfer of contaminants from surface waters to deeper waters through a process known as the 'biological pump'. This process is likely increasing the sequestration of POPs from the atmosphere into deep ocean waters and sediments.
- Positive correlations exist between some POP concentrations in Arctic air and climate oscillation indices. Larger seasonal variations in POP concentrations occurred when the Arctic Oscillation index was negative.
- Local emissions of some POPs and CEACs (e.g. PCBs, PFAS, flame retardants, PAHs) have been shown near Arctic communities, military and industrial sites, and infrastructures (e.g. airports). Human activities (e.g. shipping, tourism, oil and gas development, fisheries) are likely to increase with future changes in climate, leading to potential increases in local emissions of contaminants within the Arctic.

continued overleaf

- There are indications that the reduction of sea ice due to climate warming is leading to a general increase of sea spray aerosols in the Arctic. This implies that re-emissions of water-soluble, surface-active POPs (e.g. PFAS) to air and coastal environments will increase as well.
- Extreme weather events (e.g. severe rain events, snowstorms, unseasonal warming in parts of the Arctic) and forest fires are becoming more frequent. Short-term elevated air concentrations of PAHs and PCBs in the high Arctic have been traced to wildfires in boreal forests of Canada and Russia. Increased discharge volumes from Eurasian rivers have been associated with increased loadings of PAHs to the Arctic Ocean.
- Natural halocarbons (nHCs) are likely to increase in the Arctic over this century due to climate change effects on nHC-producing species (i.e. phytoplankton and macroalgae), with potential, but uncertain impacts on stratospheric ozone.

Influence of climate change on accumulation of POPs in Arctic food webs

- First-year ice now dominates ice coverage over large parts of the Arctic and is coupled with earlier or erratic thawing. The melting of brine-rich, first-year ice may result in more efficient delivery of POPs to organisms at the base of the marine food web.
- Permafrost thaw has been shown to impact lake water chemistry in the Canadian Arctic and influence the condition of landlocked Arctic char via impacts on their dietary sources.
- Significant dietary shifts are being observed in Arctic animals due to climate-driven migrations of species from temperate waters and declines in sea ice that are changing movements and behaviors of ice-dependent species (e.g. polar bears shifting from hunting on pack ice to foraging on land).
- The combination of contaminant exposure and sea ice decline has synergistic adverse effects on lipid metabolism in polar bears, as biomarker responses of less- and more-polluted bears showed greater contrast during a period with poor sea ice conditions.
- There are too many unknowns and variable results among species and ecosystems to firmly conclude the net effect of climate change-driven impacts on species interactions and food web contaminant accumulation.

Associations between climate change and temporal trends of POPs in Arctic biota

- Positive associations have been found between annual mean concentrations of many POPs in biota and climate oscillation indices (e.g. Arctic Oscillation (AO) and North Atlantic Oscillation (NAO)) over time, and sometimes with time lags, suggesting that changes in air mass transport from North America and Europe toward the Arctic can influence POP levels in biota.
- Positive associations were also found between some POP levels in landlocked Arctic char and climate-related parameters, including temperature and precipitation. Thawing permafrost, and the resulting release of particulate matter and carbon into lake systems, was generally linked to increased POP levels in Arctic char and freshwater amphipods.
- Annual mean concentrations of many POPs in biota were associated with sea ice extent. Some associations between climate parameters and POP levels showed time lags.
- Dietary changes affected POP exposures of some Arctic animals. In general, effects on long-term POP trends were small, but temporary perturbations and changes in long-term rates were observed.
- Including climate parameters in time trend analyses did not affect the overall direction of the trends (i.e. increase or decrease of POP concentrations) but could affect the magnitude of the annual changes (i.e. towards a faster or slower rate of change).
- Results from the Antarctic showed that melting glaciers and calving icebergs coincided with temporary increases in POP levels in biota.

general, there are now many observations that suggest linkages exist between climate-induced changes and the fate of Arctic contaminants. However, these linkages are mainly correlative and the causal relationships behind these observations are still poorly understood. This is related to the multitude of direct and indirect effects of climate change on biological, chemical and physical processes that interact to influence the fate of contaminants within a rapidly warming Arctic, as illustrated in Figure 3.1.

The present assessment of POPs and CEACs in relation to climate change includes data from the fields of meteorology, hydrology and glaciology, which have not typically been considered in previous Arctic POP assessments, but are addressed by other AMAP reports (ACIA, 2005; AMAP, 2011b; AACA, 2017). Considering the complexity of physical, chemical, and biological processes, and the rapid developments with regard to both climate change and chemical contamination, interdisciplinary scientific exchange and collaboration are important. Going forward there is a need for a stronger collaborative approach to understanding the influence of climate change on POPs and CEACs due to high uncertainties and the incremental process of increasing knowledge of these chemicals. For example, the 'integrative approach' in modeling is the process of applying models iteratively to include evolving information about emissions, environmental levels, chemical properties, and new developments in climate change observations and model predictions. This approach benefits from collaboration among disciplines and between governments, universities, and local and indigenous peoples.

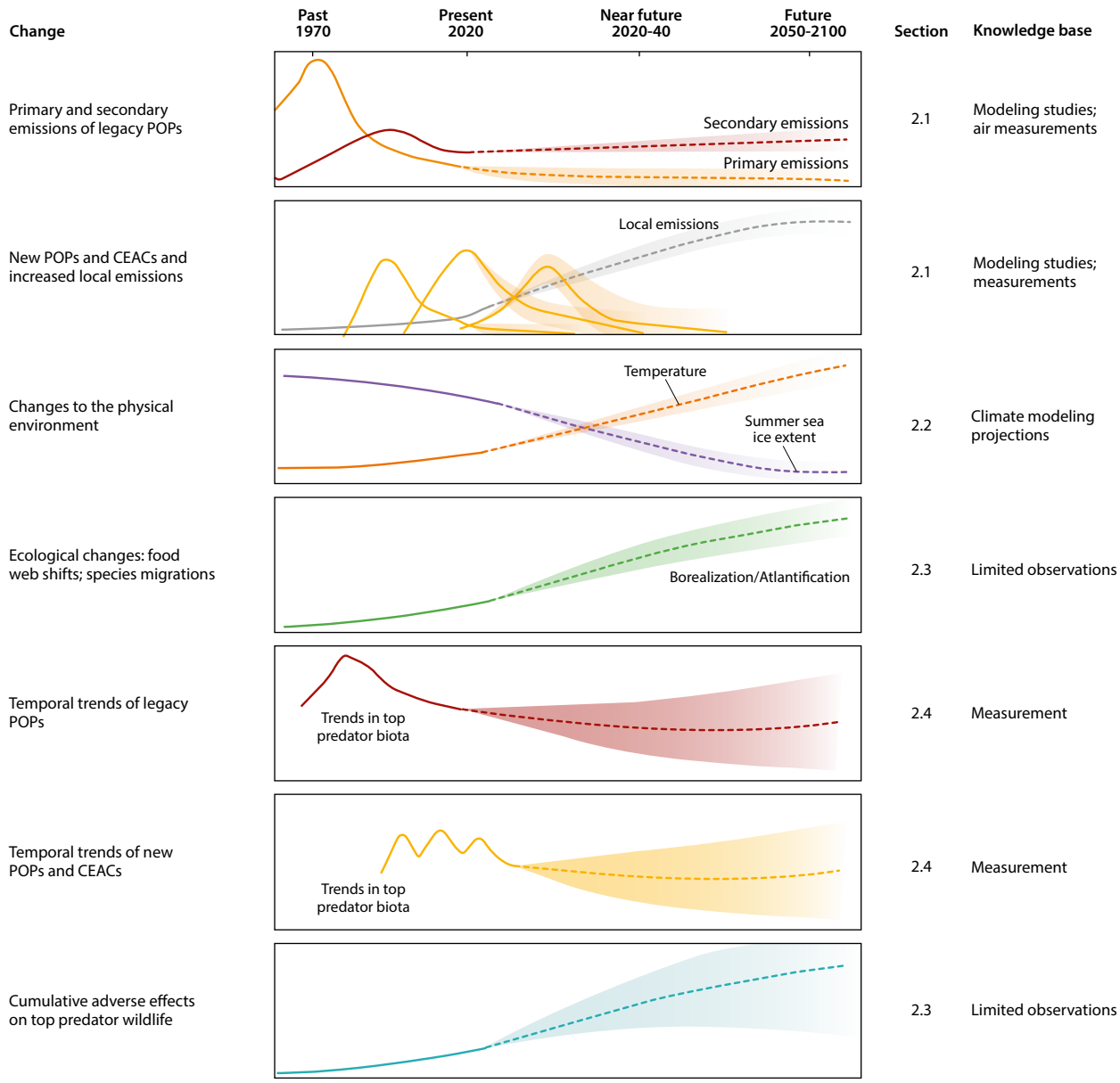

Figure 3.1. Projected changes in chemical emissions, environmental conditions, and ecosystems that illustrate the complexity in trends of POPs and CEACs in the Arctic under future climate change based on current information derived from this assessment. Temporal trends of PCBs are used as examples of legacy POPs. Dashed lines indicate predicted relative trends compared to modeled or measured trends (solid lines) for the period from 1970–2020. Shaded areas indicate relative uncertainty in projected trends.

3.2 General conclusions and recommendations

What are the key climate change-driven physical and/or ecological processes influencing POPs in Arctic wildlife, and how will climate change influence levels of POPs in Arctic biota and food webs? This assessment has demonstrated that there is a vast diversity of processes through which climate change may affect contaminant exposure in Arctic ecosystems. The direction and extent of changes in POP concentrations in biota are not consistent but vary between species, ecosystems and locations. In general, higher temperatures are leading to the release of stored POPs from melting ice, glaciers and thawing permafrost into aquatic systems, where higher temperatures can also affect the uptake and elimination of POPs in cold-blooded organisms. Variations in climate oscillation indices, which reflect changes in air mass movements, and thus contaminant transport, are correlated with changes in POP accumulation in some Arctic biota. Reductions in the amount and extent of sea ice are influencing the air-water exchange of contaminants and prey accessibility. In addition, the northward movement of subarctic species from the Atlantic and Pacific (i.e. Atlantification and borealization) is also leading to increased prey-switching, with consequences for POP exposure and tissue levels (Figure 3.1). Changes in the structure and function of Arctic ecosystems are not currently predictable, but disruptive impacts on a broad range of habitats, species, and processes are likely, which in turn will disrupt the dynamics of POPs in the Arctic.

Recommendation: Research on climate-induced changes in ecosystems and consequences for POP exposure and accumulation in wildlife should reflect the diversity of Arctic ecosystems and the

complexity of potential impacts and feedbacks of climate change on ecosystems. National monitoring programs should include ancillary biological and ecological data (e.g. body condition, fatty acid signatures, and stable carbon, nitrogen and sulfur isotope ratios), along with physical data (e.g. temperature, precipitation amounts, and ice coverage and quality) at the local scale, to better understand the factors that influence the accumulation of POPs in biota under climate change.

Can we link changes in temporal trends of POPs with climate parameters and/or food web changes? Many POP time trends established for Arctic air and biota have now achieved the statistical power needed to evaluate relationships with climate parameters (AMAP, 2016b). However, only a few POP time series from the Arctic have been investigated for correlations with climate parameters to date. Some of these time series do show associations between changes in POP concentrations and climate parameters (e.g. climate oscillation indices, sea ice extent, precipitation) over time and provide important indications of covariates to consider in the interpretation of POP time trends, as further detailed in Chapters 2.2 and 2.4.

Recommendation: Existing time trend monitoring data should be used to explore associations between climate parameters and POP concentrations in the Arctic to a greater extent. POP time series of sufficient length are available from most Arctic countries, in many cases covering many compounds in air and/or multiple species of biota. Studying correlations between climate data and POPs could provide a basis for formulating hypotheses about the most important mechanisms of climate effects on POPs in the Arctic.

Do the findings related to temporal trends of POPs in air and biota have implications for the national and international regulation of chemicals? Temporal trends of POPs in the Arctic are used as indicators in effectiveness evaluations of the Stockholm Convention, which regulates primary emissions of POPs. In general, POP concentrations have been decreasing in the Arctic environment. However, some POP time series have shown recent perturbations in these decreasing trends that seem to be associated with climate-related changes in physical processes or ecosystems. The reduction of primary emissions is still considered to be the main driver of POP time trends (Figure 3.1), however, for some compounds, the rate of decline in concentrations has been correlated (negatively or positively) with climate-related parameters.

Recommendation: There is a need for further research to identify and characterize the direct and indirect linkages between climate-induced changes in the long-range transport of contaminants to the Arctic and their accumulation in wildlife to aid the interpretation of trend data.

How well can we anticipate how POPs and CEACs, as well as microplastics, will impact the Arctic in a changing future climate? As outlined in the introduction (Chapter 1), this report has only addressed POPs included in the Stockholm Convention and selected substances assessed in the recent AMAP report on CEACs (AMAP, 2017a). Most of the currently available studies linking climate change and contaminants in the Arctic have addressed "initial" or legacy POPs (i.e. polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), hexachlorobenzene (HCB)), and new POPs (i.e. hexachlorocyclohexanes (HCHs), polybrominated diphenyl ethers (PBDEs), perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA)), as well as polycyclic aromatic hydrocarbons (PAHs). The largest amount of data is available for these compound groups, but focusing on these data-rich substances might result in an incomplete assessment of the consequences of climate change for Arctic contaminants. Some CEACs, such as the short-chain per- and polyfluoroalkyl substances (PFAS), halogenated natural products, and organophosphorus flame retardants, have much higher solubility in water than legacy and new POPs, while others, like the volatile neutral PFAS precursors, have much greater volatility. These differences in physical-chemical properties have implications for climate-related changes in long-range atmospheric or oceanic transport to the Arctic and accumulation thereafter.

Recommendation: A broader range of contaminants should be included in research and monitoring studies addressing climate-related effects on contaminants in the Arctic to account for differences in physical-chemical properties between compounds and associated differences in their long-range transport and fate.

The presence of microplastics is an important emerging issue in the Arctic which is being addressed separately by AMAP. However, microplastic particles can bind and leach POPs and CEACs, and thus may serve as a source or transport vector of contaminants to the Arctic. There is a general lack of knowledge with regard to climate change-related effects on microplastics and associated contaminants, especially concerning the processes and rates of microplastic incorporation within sea ice, the effect of microplastics on sea ice properties, and the potential release of microplastics and associated contaminants from melting ice.

Recommendation: The role of microplastic particles present in snow and ice as climate forcers requires further study. In addition, the exposure of Arctic biota to microplastics, and the role of microplastics as vectors for contaminant transport and biotic exposure needs further investigating. Future work under the AMAP POPs Expert Group and the AMAP Litter and Microplastics Expert Group should be coordinated in this respect.

What are the primary sources of POPs, how do POPs reach the Arctic, and how are emissions and source locations of POPs and CEACs affected by climate change? Information about primary emissions, including variations over time and space, is fundamental for assessing changes in the fate of POPs and CEACs, including their transport to the Arctic and distribution processes between different media. Temporally- and spatially-resolved global or circumpolar emission inventories only exist for a few POPs (PCBs, DDT, HCHs) and CEACs, such as PAHs. Therefore, new approaches, such as inverse modeling are required to establish emission patterns, in particular for those contaminants where emissions are associated with the entire lifecycle of consumer products (i.e. production, use, and disposal) rather than, for example, industrial or agricultural sources.

Recommendation: There is a need for temporally- and spatially-resolved emission inventories at circumpolar and global scales for more POPs and CEACs similar to what has been achieved

for PCBs, DDT, HCHs, and PAHs. Emissions estimates derived from inverse or 'top down' modeling (i.e. estimating emissions from measurements where large datasets of levels and trends are available) should be employed to address this data gap.

Does climate change within the Arctic exacerbate or diminish contaminant transport, accumulation, and occurrence in different abiotic media? Increasing temperatures and cryospheric changes have increased the mobility and transfer of POPs between physical environmental compartments of the Arctic through various mechanisms, including enhanced volatilization of contaminants from water, snow and ice, and re-mobilization of contaminants from melting of sea ice (i.e. multi-year and first-year ice), glacier ablation, and permafrost thaw, degradation and slumping.

Recommendation: More research is needed to better understand the implications of accelerated melting and re-mobilization of contaminants on the accumulation and exposure of marine and freshwater environments. Simultaneous multimedia assessments of contaminant redistribution (e.g. particulate-bound vs dissolved concentrations) and accumulation in food web organisms should be conducted to systematically quantify such impacts.

How do local sources contribute to Arctic contamination compared to long-range transport under climate change scenarios? Climate change and a diminishing cryosphere will likely increase human activity (e.g. shipping, tourism, industrial operations) and promote the expansion of urban areas in the Arctic. Additionally, adaptations to climate change might also include changes in lifestyles, behaviors and policies. These potential developments should be considered in assessments of contaminants in the Arctic, as they can involve the use of chemicals and thus have the potential to contribute to local releases of substances widely used in consumer and industrial products, some of which may be CEACs (Figure 3.1).

Recommendation: Given the likelihood of increased human activity and development in the Arctic due to climate change, there is a need to evaluate the emissions of newly-listed POPs, as well as current-use chemicals found in consumer and industrial products, within the Arctic. It will be relevant to assess the sources of these chemicals and the relative contributions of long-range transport and local emissions to their occurrence in the Arctic. It is also recommended that a broader list of substances be assessed for contamination potential and that time trend monitoring studies be expanded to include a broader range of chemicals and media (e.g. water).

How can Indigenous Knowledge contribute to the discussion of climate-related effects on trends of POPs?

Arctic indigenous peoples possess a rich repository of environmental knowledge and are directly affected by contaminants and climate change. Thus, their involvement and contributions are essential for assessing the effects of climate change on POPs and CEACs in the Arctic. The present assessment has identified questions that would benefit from specialized Indigenous Knowledge and the co-production of knowledge, especially concerning local and regional changes in environmental conditions (e.g. sea ice) and food webs that may affect POP trends, but would not be reflected in circumpolar-scale climate indices or biodiversity assessments.

Recommendation: Indigenous Knowledge and the participation of indigenous and local communities would substantially benefit the interpretation of climate change-related observations in the Arctic, including changes in the timing of events, sea ice, animal distribution, behavior, diet and body condition, and many other variables. It would therefore also substantially benefit the interpretation of POP trends and associations between climate change and POPs.

3.3 Knowledge gaps

This assessment has highlighted a set of knowledge gaps that warrant priority for future research to build a knowledge base that would serve Arctic communities, government decision-making, and policy needs. The knowledge gaps listed below are not ranked; general knowledge gaps are presented first, followed by specific knowledge gaps related to Chapters 2.1-2.4. Although more empirical data are available than in previous assessments to provide evidence of predicted changes, the effects of climate change on the fate of POPs and CEACs in the Arctic are not fully understood.

3.3.1 General knowledge gaps

- The present assessment has revealed a high complexity of direct and indirect effects of climate change. This complexity is far from understood, in particular regarding exposure and bioaccumulation processes. Climate change affects the amount and availability of POPs in the Arctic environment. It also affects the behavior of Arctic animals, including their foraging patterns. Both factors contribute to changes in POP levels in biota, but an understanding of mechanisms is lacking.
- There is a limited quantitative understanding of the effects of climate change on contaminant-related processes (e.g. whether volatilization or degradation of POPs will be the dominant process influencing Arctic air concentrations under increased temperature conditions).
- Many of the climate-related effects on contaminants identified in this assessment are based on correlation analyses without clearly identified causal relationships or a mechanistic understanding of relationships with contaminant fate.
- Indigenous Knowledge has contributed to previous assessments of both contaminants and Arctic climate change, however, there is a greater, underutilized potential to partner with indigenous and local communities within the Arctic in the co-production of knowledge on changes in local environmental conditions and food webs.

3.3.2 Knowledge gaps: emissions and long-range transport

- Emission estimates are limited to a few POPs such as PCBs, legacy chlorinated pesticides, and some CEACs, such as PAHs. These need to be updated as well as extended to ensure

sufficient temporal and spatial resolution for modeling of transport to the Arctic.

- Knowledge is lacking on the relative source attribution of POPs and selected CEACs in the Arctic to distant, regional and local sources.
- Evaluations of model-based predictions of POP and CEAC deposition to the Arctic are lacking.
- Uncertainties in the physical-chemical properties of chemicals of interest, especially CEACs, impede estimations of emissions and modeling efforts.
- Sensitivity analyses on model simulations need to be improved to evaluate the effects of climate-related changes in atmospheric composition and meteorological conditions on POP and CEAC concentrations in the Arctic.
- The potential effects of regime shifts in climate conditions that result from transgressing "tipping points" in the climate system (e.g. the complete loss of summer sea ice) on POP and CEAC concentrations have yet to be considered.
- There is a need for detailed studies of processes affecting the long-range transport of POPs and CEACs, including the joint analysis of modeling results and monitoring data (e.g. gas-particle partitioning, degradation, surface-air exchange, etc.)

3.3.3 Knowledge gaps: physical environment – levels and trends

- There is a need to address local sources of POPs, CEACs, and other as of yet unstudied current-use chemicals that could increase as a consequence of greater human activity in the Arctic related to climate change.
- Knowledge of contaminant re-mobilization due to permafrost thaw and erosion is limited to a few studies and locations. Thus, the relevance of such processes to biotic exposures in freshwater systems is currently difficult to assess due to the lack of data from across the circumpolar regions.
- Volatile natural halocarbons (nHCs) are likely to increase over this century due to the effects of climate change on producing species (i.e. phytoplankton and macroalgae) and biogeochemical cycles. The impacts of increased emissions of nHCs on stratospheric ozone are uncertain and need further investigation.
- The role of microplastic particles present in snow and ice as climate forcers is currently unknown. In addition, the exposure of Arctic biota to microplastics and the role of microplastics as vectors for contaminant transport and biotic exposure need further investigation.
- There is a lack of knowledge on the contaminant loadings from precipitation to polar marine surfaces (e.g. open water, ice melt ponds) and the relative input of these sources versus oceanic transport to the Arctic.
- The rapid replacement of multi-year ice by brine-rich first-year ice in a warming Arctic necessitates studies investigating the exposure of sympagic organisms (e.g. ice algae and associated zooplankton in brine channels) to contaminants and the contaminant loadings from first-year ice to ocean waters.
- While it is foreseeable that extreme weather events (e.g. severe rain events, floods, snowstorms and unseasonal warming in parts of the Arctic) will become more frequent due to climate change, studies examining the role of such events on the distribution pathways of POPs are currently lacking.

3.3.4 Knowledge gaps: influence of climate change on accumulation of POPs in Arctic food webs

- Much uncertainty remains as to the ecosystem-level effects of climate-derived changes in food webs on the exposure of Arctic wildlife to POPs.
- No data are currently available on the net effect of increased temperatures on the uptake and elimination rates of POPs in cold-blooded Arctic organisms.
- Changes in the POP levels of biota resulting from the direct effects of declining sea ice cover cannot be separated from the indirect effects of changes in food web structure and function with the present state of knowledge.
- Increased terrestrial runoff from snow and glacier meltwater is expected to influence both the exposure levels and biomagnification of POPs in lake and estuarine food webs, but there are few empirical data available to confirm this.
- There is a general lack of understanding regarding the impact of climate-driven changes in the phenology of ecological events (e.g. seasonal timing of migration, reproduction and food availability) on POP levels in Arctic biota.
- The net effect of climate-related increases in freshwater and marine primary production on the bioavailability and bioaccumulation of POPs in Arctic biota is not well understood.
- Climate-driven migrations of species from more southerly latitudes may introduce new sources of contaminants into Arctic food webs, however, the importance of these biovectors in transporting contaminants to the region is largely unknown.
- Changes in seabird and marine mammal foraging patterns due to climate change have the potential to impact their exposure to POPs, but few data exist.
- There is a general lack of understanding of how climate change and other environmental stressors affect the toxicity of POPs to Arctic biota.
- Mechanistic process-oriented models combining physical-chemical transport models with bioenergetic and food web models can be used to project the overall bioaccumulation of POPs in Arctic biota, but there is insufficient understanding or empirical data to parametrize these models.
- There is a general lack of knowledge of how climate-driven impacts on terrestrial ecosystems may influence levels of POPs in terrestrial food webs (e.g. lichen/caribou/wolf food web).

3.3.5 Knowledge gaps: climate change and temporal trends of POPs in Arctic biota

- While associations between POP time trends and climate parameters (e.g. sea ice coverage, air temperatures, oscillation indices) have been identified, the mechanisms underlying these correlations are not understood.
- Climate oscillation indices, sea ice, temperature and precipitation have been the predominant climate parameters used to evaluate associations between climate change and contaminant temporal trends in the Arctic thus far, but it is not known if other parameters might be equally or more important.
- The inclusion of climate parameters in temporal trend models has been shown to elevate or reduce the observed rates of decrease in POP concentrations in Arctic biota. A greater understanding of underlying processes is needed to explain these results.
- The relationship between climate parameters and contaminant time trends in biota varies across species and locations. The reasons for these variations (e.g. influence of local or regional conditions) are not known.
- Information on the effects of climate change on POP time trends in low trophic level organisms is generally lacking. In addition, there are no systematic studies of climate change effects on POP time trends in Arctic terrestrial ecosystems currently available.
- Associations have been shown between processes related to permafrost thaw, degradation and slumping, and POP levels in freshwater biota. The mechanisms underlying these connections are not fully understood, but increased particle loads and enhanced biological activity likely play a role.
- Observations of stable or increasing HCB concentrations in Arctic biota are frequently reported, but lack explanation.
- Although iceberg calving has been identified as a potential source of POP release and accumulation in Antarctic marine biota, no studies have examined this phenomenon in the Arctic.
- While the overall effects of climate change parameters on long-term contaminant time trends appear small at present, this may be a momentary observation. Updated information is needed to confirm that this is a general trend, as primary emissions will presumably continue to decrease, while the effects of climate change may become increasingly evident and can be expected to lead to complex feedback reactions.

3.4 Policy implications

- Although climate change is impacting POP levels and trends in the Arctic, the reduction of primary emissions is currently considered the main driver of POP time trends. Thus, continued efforts to include more POP-like chemicals in chemical management initiatives at the global level (e.g. the Stockholm Convention) and national or regional levels are needed.
- Results from this assessment are relevant to other regional, national and international forums addressing contaminants and climate change, such as the recent Global Environmental Facility Scientific and Technical Advisory Panel initiative on the co-benefits of sound management of chemicals and waste (GEF, 2020) and the Strategic Approach to International Chemicals Management (SAICM) policy framework.
- A precautionary approach to the management of persistent chemicals is prudent due to the poorly reversible consequences of global exposure and the range of hazards that are difficult to anticipate. These risks are compounded by deficiencies in our present understanding of how climate change will affect the global fate and transport of chemicals.
- The relative contributions of long-range transport versus local emissions of POPs and CEACs to the Arctic may be changing due to climate change. There is a need to identify local sources and re-evaluate their contribution to Arctic pollution in order to inform chemical management decisions within and outside the region to reduce contaminant loads to the Arctic environment.
- There are indications of climate change leading to increased human activity in the Arctic and thus potentially increased use of chemicals within the region. Therefore, there may be a need for additional risk assessments that are not solely based on the criteria used for POPs or similar chemicals (e.g. PBT under EU REACH), but also include, for example, criteria related to chemical persistence and mobility, and emissions from consumer products.
- Climate change can influence secondary emissions of POPs, as well as the physical, chemical and biological processes that affect their long-range transport and bioaccumulation, and thus, their long-term time trends in Arctic air and biota. These effects are outside the scope of the Stockholm Convention but need to be taken into account in relation to effectiveness evaluations.
- Without further action on climate change, alterations in the Arctic ecosystem are expected to continue and may impact the effectiveness of policies seeking to reduce POP exposures in Arctic people and wildlife through primary source reductions alone. It is important for decision-makers to recognize the diversity and complexity of climate change-induced effects on POPs, and that specific measures can have a variety of outcomes via physical, chemical and biological interactions and feedback reactions.
- There is a need for funding agencies to support international and interdisciplinary approaches to understanding the effects of climate change on the fate of POPs and CEACs in the Arctic. Long-term studies are crucial in this context.
- With the complexity of climate change impacts on contaminants in Arctic ecosystems, it is important to plan for integrated monitoring, which includes the collection of ancillary climate parameters, and biological and ecological data in addition to contaminant data. This includes, but is not limited to, data on climate indicators and local meteorological conditions, as well as information on the physiology, biology and ecology of Arctic organisms. To be operative, integrated

monitoring requires a stable institutional background and the agility to integrate short-term and local efforts and resources into a coherent service.

- In some cases (e.g. reducing combustion sources) there are co-benefits from actions on climate change that can also help mitigate chemical exposure problems.
- Additional capacity building of research programs, scientists, northern communities and particularly, indigenous youth, is needed to support and encourage community-based studies and the co-production of knowledge, including the utilization of Indigenous Knowledge for interpreting POP trends in light of climate change. Community involvement in monitoring activities should be extended and expanded to include interdisciplinary issues at the interface of contaminants and climate change, as addressed in this assessment.
- Monitoring programmes and individual research studies should comply with open data policies to ensure that quality-assured raw data on POPs and related ancillary data are publicly available for future study.

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Acronyms and Abbreviations

$\delta^{13}C$	Carbon stable isotopes
$\delta^{15}N$	Nitrogen stable isotopes
20C3M	20th Century Climate Scenario model
AACA	Adaptation Actions for a Changing Arctic
AICc	Akaike's Information Criterion corrected for small sample size
AMAP	Arctic Monitoring and Assessment Programme
AO	Arctic Oscillation
ArcRisk	Arctic Health Risks Project
AWS	Automatic weather stations
BA	Bromoanisole
BFR	Brominated flame retardant
BMI	Body mass index
BP	Bromophenol
CBz	Chlorobenzene
CEAC	Chemical of emerging Arctic concern
Cl-OPE	Chlorinated organophosphate ester
CHL	Chlordane
CLRTAP	Convention on Long-range Transboundary Air Pollution
CMB	Climatic Mass Balance model
CUP	Current use pesticide
DEHM	Danish Eulerian Hemispheric Model
dw	Dry weight
DDT	Dichlorodiphenyltrichloroethane
DDE	Dichlorodiphenyldichloroethylene
DiBA	Dibromoanisole
DOC	Dissolved organic carbon
EF	Enrichment factor
EHDPP	2-ethylhexyl diphenyl phosphate
ENSO	El Niño Southern Oscillation
ESB	Environmental Specimen Bank
FR	Flame retardant
FTOH	Fluorotelomer alcohol
FYI	First-year ice
GLM	General linear model
GMP	Global Monitoring Plan
HBCDD	Hexabromocyclododecane
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
Hg	Mercury
hHNP	Higher molecular weight halogenated natural product
HNP	Halogenated natural product
IK	Indigenous Knowledge
IPCC	Intergovernmental Panel on Climate Change
K	Condition factor
K_{AW}	Air-water partitioning coefficient
K_{OA}	Octanol-air partitioning coefficient
K_{OW}	Octanol-water partitioning coefficient
ln	Natural logarithm
log	Logarithm to the base 10

LRAT	Long-range atmospheric transport
LRT	Long-range transport
lw	Lipid weight
MASL	Meters above sea level
MEF	Melt-pond enrichment factor
MeO-PBDE	Methoxylated polybrominated diphenyl ether
MOSJ	Environmental Monitoring of Svalbard and Jan Mayen (MOSJ) Programme
MYI	Multi-year ice
NAO	North Atlantic Oscillation
NCP	Northern Contaminants Program
nHC	Natural halocarbon
NIST	National Institute of Standards and Technology
OCP	Organochlorine pesticide
OH-PBDE	Hydroxylated polybrominated diphenyl ether
OPE	Organophosphate ester
PAH	Polycyclic aromatic hydrocarbon
PNA	Pacific/North American Pattern
PBDE	Polybrominated diphenyl ether
PCA	Pentachloroanisole
PCB	Polychlorinated biphenyl
PCDD	Polychlorinated dibenzo-*p*-dioxin
PCDF	Polychlorinated dibenzofuran
PCN	Polychlorinated naphthalene
PCNB	Pentachloronitrobenzene
PCP	Pentachlorophenol
PDO	Pacific Decadal Oscillation
PFAA	Perfluoroalkyl acid
PFAS	Per- and polyfluoroalkyl substances
PFCA	Perfluorocarboxylic acid
PFHpS	Perfluoroheptane sulfonic acid
PFHxS	Perfluorohexane sulfonic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFSA	Perfluorosulfonic acid
PFTrDA	Perfluorotridecanoic acid
PNA	Pacific North American Oscillation
POC	Particulate organic carbon
POP	Persistent organic pollutant
RTS	Retrogressive thaw slump
SCCP	Short-chain chlorinated paraffin
SPM	Suspended particulate matter
SRES	Special Report on Emissions Scenarios model
SSA	Sea spray aerosol
SVOC	Semi-volatile organic chemical
TBL	Tropospheric boundary layer
TmCP	Tris meta-(cresyl) phosphate
TMF	Trophic magnification factor
TnBP	Tri-n-butyl phosphate
TOC	Total organic carbon
TriBA	Tribromoanisole
UNEP	United Nations Environment Programme
WRF	Weather Research and Forecasting model

Arctic Monitoring and Assessment Programme

The Arctic Monitoring and Assessment Programme (AMAP) was established in June 1991 by the eight Arctic countries (Canada, Denmark, Finland, Iceland, Norway, Russia, Sweden and the United States) to implement parts of the Arctic Environmental Protection Strategy (AEPS). AMAP is now one of six working groups of the Arctic Council, members of which include the eight Arctic countries, the six Arctic Council Permanent Participants (indigenous peoples' organizations), together with observing countries and organizations.

AMAP's objective is to provide 'reliable and sufficient information on the status of, and threats to, the Arctic environment, and to provide scientific advice on actions to be taken in order to support Arctic governments in their efforts to take remedial and preventive actions to reduce adverse effects of contaminants and climate change'.

AMAP produces, at regular intervals, assessment reports that address a range of Arctic pollution and climate change issues, including effects on health of Arctic human populations. These are presented to Arctic Council Ministers in 'State of the Arctic Environment' reports that form a basis for necessary steps to be taken to protect the Arctic and its inhabitants.

This report has been subject to a formal and comprehensive peer review process. The results and any views expressed in this series are the responsibility of those scientists and experts engaged in the preparation of the reports.

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