

Influence of Climate Change on Transport, Levels, and Effects of Contaminants in Northern Areas

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The Arctic Monitoring and Assessment Programme (AMAP) was established in June 1991 by the eight Arctic countries (Canada, Kingdom of 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 preventative 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.

AMAP technical reports are intended to communicate the results of scientific work that contributes to the AMAP assessment process. 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 and have not been approved by either the AMAP Working Group or the Arctic Council.

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The AMAP Secretariat is located in Oslo, Norway. For further information regarding AMAP or ordering of reports, please contact the AMAP Secretariat (Gaustadalléen 21, N-0349 Oslo, Norway) or visit the AMAP website at www.amap.no.

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Executive Summary

Climate change, particularly increased temperatures globally and especially in the Arctic, will affect the concentrations of persistent organic pollutants (POPs) and heavy metals in the different environmental media. The transport and pathways of pollutants to the Arctic from source regions in the northern hemisphere depend on the physical-chemical properties of each contaminant. Modeling studies using the Danish Eulerian Hemispheric Model (DEHM) in this project show that higher temperatures will enhance volatilization of some contaminants (e.g., γ -HCH, PCB52 and PCB153) from soil to air and thus increase their atmospheric transport to the Arctic. However, this impact is expected to be offset by increased degradation of these banned legacy contaminants owing to higher temperatures and by decreasing emissions. Contaminant fluxes between the ocean and atmosphere will also be altered, affecting their distribution and availability for uptake at lower trophic levels in the marine environment. This report provides maps of modeled estimates of the change in concentrations of γ -HCH, PCB52 and PCB153 in the atmosphere and oceans of the northern hemisphere between 1995–1999 and 2095–2099, as well as relative changes in their distribution between environmental compartments and food web biomagnification. In this report, only changes due to climate change have been studied. It is expected that changes due to lower emissions of these regulated contaminants in the future will be much greater than these climate-related changes.

Concentrations of POPs in Arctic seawater at the end of the 21st century as estimated with DEHM were used as input data in a bioaccumulation model to estimate future biomagnification of these contaminants in an Arctic marine food web. Overall, the modeled effect of the annual cycle of exposure, temperature, lipid content and food-web structure on food-web biomagnification (i.e., seasonal variations in bioaccumulation) was greater than the modeled changes in food-web biomagnification in response to projected climate change. It should be noted that the simulated food-web structure and diet composition did not change significantly between seasons.

As the human population in the Arctic increases, local sources of contaminants will play a greater role in contaminant exposure. With increasing and changing population structures expected,

the influence of local anthropogenic sources on pollutant patterns is likely to alter. Therefore, several local sources of contaminants on Svalbard were investigated, including characterization of atmospheric particulate and gaseous emissions from power plants, uptake of perfluorinated alkylated substances (PFASs) in freshwater fish, marine birds, and reindeer, and POPs concentrations in halibut and shrimp from northern Norway. The results of these studies provide a basis for better regulation of local pollution sources.

Studies of contaminant levels in the blood of residents in northern Norway and parts of Arctic Russia show that levels of POPs and PFASs subject to international regulations or bans have been decreasing over the past three decades in local residents, while concentrations of unregulated PFASs are still increasing in residents of northern Norway. These results provide encouraging evidence of the value of international regulations in reducing human exposure levels and, thus, ultimately health effects in exposed populations.

The humic-rich Fennoscandian surface soil is an important sink for metals and other air pollutants, a large percentage of which are transported in from other areas. Terrestrial moss has been used for several decades to monitor the atmospheric deposition of cadmium (Cd), lead (Pb) and mercury (Hg) in Norway. While Pb deposition has decreased significantly over the past 30 years and Cd deposition decreased strongly until 2005, Hg concentrations in mosses have stayed the same over the past three decades. Mercury concentrations in moss are essentially uniform over all of Norway, while Pb and Cd deposition are highest over southern Norway. In contrast to the other metals, moss Hg probably reflects a combined signal of deposition and air concentrations and is thus not an ideal indicator for deposition input to ecosystems. The Hg cycle, particularly in the Arctic is expected to undergo several changes owing to climate change, but given the complexity of the Hg cycle it is not currently possible to determine whether the combined effects of climate-related changes will increase or decrease Hg concentrations in the atmosphere. There are several climate-dependent processes related to Hg in the Arctic, for example, atmospheric mercury depletion events in spring, which are dependent on sea-ice/open leaks (important for Hg deposition in the Arctic), biological reduction processes in the ocean (important for re-emissions), and changing patterns of atmospheric transport and wet deposition.

1. Introduction

The Arctic Monitoring and Assessment Programme (AMAP) was established in 1991 by the eight Arctic states to implement part of the Arctic Environmental Protection Strategy (AEPS) adopted that year by these countries. In 1996 the AEPS, including its working groups, was reorganized to form the Arctic Council.

Under the remit of AMAP, some of the main objectives are to monitor and assess levels, trends and effects of environmental pollution in the Arctic. This work is undertaken by a large number of scientists and experts from institutes and national agencies in the member countries. Since 1991, AMAP has conducted several major assessments of persistent organic pollutants (POPs), hazardous metals and other anthropogenic pollutants, climate change, and effects on humans and ecosystems in the Arctic. These were prepared based on peer-reviewed research publications, national reports and conference presentations by a large number of Arctic specialists and scientists. AMAP also coordinates research-based projects concerning issues of importance to achieving a better understanding of the Arctic environment.

This report provides a compilation of the main results of an integrated study investigating the combined effects of pollutants and climate. It was funded partly by the participating institutes and partly by the Nordic Council of Ministers (project numbers KOL-1015, MST-527-00014) and is a continuation and follow-up

to the earlier study *Combined Effects of Selected Pollutants and Climate Change in the Arctic Environment* (AMAP, 2011a). That study identified a number of knowledge gaps requiring future investigation, and the present study aimed to follow up on some of those recommendations. These include:

- Improve knowledge about exchange processes of environmental pollutants between the atmosphere, ice and oceans in a changing Arctic climate
- Develop optimized analytical methods for new and emerging pollutants and subsequently include these substances in ongoing Arctic monitoring programs
- Provide relevant science-based information to help understand the fate and behavior of emerging pollutants in the Arctic terrestrial and freshwater food webs
- Obtain urgently needed scientific evidence on the combined effects of legacy POPs, emerging substances and heavy metals on their environmental fate and health effects.

To address these issues, the work undertaken as part of the present study is based on updated models, new analytical techniques and large human cohorts from northern Russia and northern Norway. This assessment concluded by recommending research priorities, identifying knowledge gaps, and addressing regulatory actions for managing potential environmental risks and exposure for human populations in the North. The study comprised five project groups:

Project element	Topic	Researchers
Component 1	Atmospheric modeling	Jesper Christensen and Kaj Hansen, Aarhus University
Component 2	Food web modeling	Katrine Borgå, Norwegian Institute for Water Research (NIVA) and University of Oslo; Jostein Starrfelt and Anders Ruus, NIVA
Component 3	Climate and persistent organic pollutants in Arctic terrestrial ecosystems	Roland Kallenborn, Norwegian University of Life Sciences (NMBU) and University Centre in Svalbard (UNIS); Pernilla Carlsson, Akvaplan-niva and AMAP
Component 4	Mercury and other heavy metals	Katrine Aspmo Pfaffhuber, Norwegian Institute for Air Research (NILU)
Component 5	Human health effects	Therese Haugdahl Nøst and Jon Øyvind Odland, UiT -The Arctic University of Norway

The project was coordinated by Janet Pawlak, AMAP Deputy Executive Secretary, and Lars-Otto Reiersen, AMAP Executive Secretary

2. Climate change and the fate of environmental pollutants in the Arctic

2.1 Influence of climate change on the Arctic environment

Climate conditions in the Arctic have changed over recent decades and as documented in the Fifth Assessment Report (AR5) of the Intergovernmental Panel on Climate Change (IPCC, 2014) it is extremely likely that human influence has been the dominant cause. The main driving factor for the changes observed in the Arctic is the rise in global average temperature, which is now the highest since measurements began. Under the four scenarios of human influence used by the IPCC in their projections of future climate change, global surface temperature increase is likely to exceed 1.5–2°C at the latter part of the 21st century (2081–2100) relative to the period 1850–1900; compared to the period 1986–2005 the global temperature rise is likely to be within the range 0.3–4.8°C (IPCC, 2007, 2014). Scientists project that the largest temperature increase will occur during the 21st century compared to the 22nd century (Hedegaard et al., 2012). The warming is occurring faster and with greater magnitude within the Arctic region than in other parts of the world. Under the most extreme scenario of human influence, the average surface temperature increase in the Arctic region in the latter part of the 21st century (2081–2100) is projected to exceed 9°C compared to the average surface temperature during the period 1986–2005.

The influence of climate change on the distribution and transformation patterns of POPs and heavy metals in the Arctic atmosphere has become an important research area for polar scientists (Kallenborn et al., 2012). Local studies of atmospheric POPs distribution patterns reveal expected changes in source strength, distribution and deposition pathways in northern environments owing to regulatory controls and paralleling those in mid-latitude environments. It is anticipated that Arctic research, including the influence of climate change on pollutant distribution, will also contribute to enhanced understanding of contaminant-related hazards in lower-latitude regions. The Arctic is considered to be an 'early warning system' for climate change-related effects in the rest of the world. Even after more than 50 years of research and monitoring, legacy POPs that are already being regulated by international agreements, as well as compounds of emerging concern, are still a priority topic for environmental scientists and national and international regulators. Knowledge of legacy POPs still dominates our understanding of contaminant patterns in the Arctic as well as the processes and mechanisms regarding the fate and impact of persistent contaminants that break down only very slowly in the environment. Both in terms of levels and their contribution to effects, polychlorinated biphenyls (PCBs) are major pollutants in top predators such as polar bears (*Ursus maritimus*) (Dietz et al., 2015). Long-term air monitoring datasets from national and international Arctic monitoring programs will continue to enable

in-depth interdisciplinary research on contaminant profiles, spatial and temporal trend studies, environmental fate, and distribution modeling and understanding of regional transport pathways in the future.

As concluded in the first phase of this project (AMAP, 2011a), climate change in the Arctic will affect the transport of pollutants to the Arctic and the fate of pollutants within the Arctic. The results from the FP7 EU project ArcRisk (*Arctic Health Risks: Impacts on Health in the Arctic and Europe owing to Climate-induced Changes in Contaminant Cycling*) indicated an increase of PCB re-emissions from secondary sources in the Arctic during the coming decades (AMAP, 2014) relative to baseline conditions under the present climate. These re-emissions from secondary and primary sources at lower latitudes are in addition to continuing emissions from remaining primary sources of PCBs in untreated waste and PCB-containing equipment and materials still in use, as well as an increased long-range atmospheric transport potential as a result of climate change (Armitage et al., 2011; Ma et al., 2011; Wöhrnschimmel et al., 2012a,b, 2013). Nevertheless, PCB concentrations in the Arctic are expected to decrease over time owing to regulations banning the production and use of PCBs that began in the 1970s (Armitage et al., 2011; Ma et al., 2011; Wöhrnschimmel et al., 2013). This report presents new scientific results on the transport and fate of POPs and mercury (Hg) to and within the Arctic under changing climate conditions. Transport of contaminants via atmospheric and oceanic currents and how the transport changes will affect the bioaccumulation, bioavailability, degradation and re-mobilization of POPs and Hg in the future have been investigated. The bioaccumulation model applied in this project allowed a comparison of the impact of seasonality versus climate change on POPs uptake in an Arctic marine food web. Although it is difficult to predict the impact of biological changes in Arctic food webs (such as the spread of new species as biovectors from the south, and changes in species composition and trophic levels) with respect to the fate of POPs, this study initiates the work required to understand such changes (Kallenborn and Blais, 2015).

The following scenarios may be used to illustrate the broad spectrum of climate change-related influences on the sources and fate of pollutants in the Arctic and the global interconnections of these issues. (i) Climate change will almost certainly affect the availability and quality of agriculture areas. A scenario under which large-scale agricultural production sites are relocated north would have inevitable consequences for Arctic regions. For example, potential primary sources of pesticides would move closer to the Arctic. (ii) The anticipated industrial and domestic infrastructure development in the north would cause a significant increase in the density and number of human settlements in the Arctic in the years to come (Armitage et al., 2011; Kallenborn et al., 2012). The range of future development scenarios and the clear influence of climate change on pollutant distribution pathways is an important incentive to obtaining a more comprehensive understanding of the hazards that legacy and emerging pollutants will pose in the future in the Arctic. Earlier research has often focused on the Arctic marine environment because marine food is a common source of essential nutrients for the Arctic population, and the main exposure pathway for

POPs and heavy metals (AMAP, 2009). To provide a more holistic approach, the results of research on emerging POPs and heavy metals in terrestrial and freshwater environments in the Arctic have also been included in this report.

Human health in the Arctic has received an increased focus during recent decades owing to the bioaccumulation of POPs in the marine food web and the resultant increased exposure in humans consuming marine foods (AMAP, 2009; Odland and Nieboer, 2012). To better understand and identify risks to human health related to dietary sources of environmental contaminants (including legacy POPs, emerging contaminants and trace metals), large cohorts of people need to be studied over long periods, and their general lifestyle, including the preferred choices of food, needs to be taken into account. However, information concerning tissue concentrations of emerging POPs and their potential effects in Arctic indigenous people is still scarce. The present study aims to determine levels of some emerging compounds and heavy metals in people as well as in relevant food items. Such information is very important for understanding and evaluating risks to human health in the Arctic.

2.2 Stress factors in the Arctic environment

Conditions in the Arctic environment are extreme. Polar night, low ambient temperatures, extensive snow and ice cover in marine and terrestrial ecosystems, and a relatively short summer are some of the physical stress factors for wildlife. Arctic wildlife has adapted to these conditions, and can even benefit from some. Arctic animals have life histories resulting from adaptation to the large seasonality of the Arctic ecosystem, with short periods of abundant food availability when large lipid stores are accumulated as energy sources (Moore and Huntington, 2008). However, when additional stress is introduced into the system, such as changes in the timing and length of the snow and ice cover season, subsequent changes within the ecosystem may affect factors that influence the exposure of wildlife to POPs and heavy metals. To better understand the effects of contaminants and climate change in the Arctic, it is necessary to assess not just the effects and behavior of legacy POPs, but also to understand the environmental fate and combined effects of emerging contaminants over a longer time perspective.

2.3 Climate change impacts on environmental processes

Arctic ecosystems are changing. The Arctic marine environment is currently undergoing dramatic changes in relation to the extent and seasonal variations in sea ice, ocean acidification, and changes in hydrology and the patterns of ocean currents in the north (AMAP, 2011c). Environmental processes such as snow/ice melt, runoff from land, and rising ocean temperatures can enhance the redistribution of POPs from secondary sources

(ACIA, 2005; Macdonald et al., 2005; AMAP, 2011c; Kallenborn et al., 2012; Carlsson, 2013). Legacy POPs are under regulation and the impact of primary emission sources is expected to decline, although the importance of secondary sources will increase (Wöhrnschimmel et al., 2012b, 2013). It is therefore important to examine projected changes in transport and distribution pathways and, in a wider context to understand how they will affect concentrations of POPs in food webs and human foodstuffs in the Arctic. Some of the main findings from the EU-funded project ArcRisk showed that the choice of foodstuff in combination with strictly regulating or banning the production and use of toxic compounds are important for reducing levels of POPs in humans and the Arctic environment (Armitage et al., 2011; AMAP, 2014; Carlsson et al., 2014a,b). However, emerging POPs such as perfluorinated alkylated substances (PFASs) can behave differently in the environment compared to legacy POPs such as PCBs owing to their different physical-chemical properties, and this must be taken into account in models and bioaccumulation studies (Stemmler and Lammel, 2010). In contrast to lipid-soluble POPs that are retained in blubber and lipid stores, PFASs are associated with protein-rich compartments (Ahrens et al., 2009; Carlsson et al., 2016). This is of toxicological importance, as it affects both their bioaccumulation and their potential interference with basic cellular processes such as fatty acid metabolism (Buhrke et al., 2015) and hormone transport (Zoeller, 2007).

The environmental changes currently observed in the Arctic and other northern areas are followed by changes in food webs as southern species expand their distribution northward from mid-latitude regions. Changes in species occurrence will alter trophic interactions in food webs and will inevitably influence the mobility and food-web biomagnification of environmental contaminants. The role of biovector-mediated transport of contaminants is an important new pathway with the potential to increase the presence of hitherto unknown anthropogenic hazardous substances in northern environments (reviewed by Kallenborn and Blais, 2015). In this context, transfer by biovectors is not only considered a direct distribution pathway, but also a means of concentrating environmental contaminants by direct uptake into the organisms concerned, where they were previously distributed in the abiotic environment in ultra-trace levels over vast areas. To date, this pathway has only been sparsely investigated for Arctic environments, and is likely to be specific to certain geographical locations. For example, it has been shown to be significant for the transport of energy and contaminants from sea to lakes by migrating seabirds. Guano from these birds contains POPs, which are delivered to Arctic lakes by the returning birds (Evenset et al., 2007). Seasonal migration has also been shown to contribute to increased contaminant levels in seabirds, comparable to one trophic level biomagnification, and in predatory birds the overwintering area significantly explained variations in contaminant levels in the breeding colonies (Baert et al., 2013; Leat et al., 2013). Due to global climate change, increased occurrences of invasive species as well as changes in migration pathways have already been observed in many regions, including the Arctic.

2.4 Models and projections

Several models have been developed in recent decades to project future scenarios. They take into account physical-chemical properties of pollutants, changing temperatures as a consequence of climate change, past pollutant emissions and estimated future emissions. However, biological parameters, such as seasonal variations in lipid content and exposure to environmental contaminants are very difficult to include in such models, but are very important for a good representation of biological variation. This report aims to combine atmospheric/oceanographic models with food-web models to investigate and differentiate between seasonal variations and climate change-related variations in the biomagnification of POPs in an Arctic food web.

2.4.1 DEHM

This study used the Danish Eulerian Hemispheric Model (DEHM). It is a three-dimensional (3D) atmospheric chemistry-transport model originally developed in 1991 to study the long-range transport of sulfur dioxide (SO₂), sulfate (SO₄) and Pb to the Arctic (Christensen, 1997; Christensen et al., 2004; Hansen et al., 2008; Hedegaard et al., 2008; Brandt et al., 2012). The model covers the entire northern hemisphere and all important sources for the Arctic are included in the model domain. The number of POPs has been increased and their physical-chemical properties have been updated in the model compared to the previous report on combined effects of selected pollutants and climate change in the Arctic environment (AMAP, 2011a). Fourteen different POPs are now included in the model simulations: the three commonly produced hexachlorocyclohexanes (HCHs): α -HCH, β -HCH, and γ -HCH and 11 PCB congeners: PCB8, PCB28, PCB31, PCB52, PCB101, PCB105, PCB118, PCB138, PCB153, PCB180, and PCB194. The simulations are made on a regular photochemistry scheme with 67 species, which describe the ozone-nitrogen-hydrocarbon-sulfur chemistry in the troposphere and lower stratosphere, and a Hg chemistry scheme with seven Hg species. The global historical AMAP Hg emissions inventories for 1990, 1995, 2000 and 2005 were used as input to the Hg module. The regular chemistry scheme ensures a proper description of particulate matter that POPs can associate with as well as a description of the hydroxyl (OH) radicals, which are the primary reaction constituent for POPs in the atmosphere. The main oxidant for elemental Hg is ozone, which is included in the parameterization of the atmospheric mercury depletion events (AMDEs) that occur over sea ice in the Arctic during polar sunrise. The POPs module has several compartments: 3D atmosphere to 15 km over the Earth's surface, 75-m upper ocean layer, 15-cm-thick soil layer, snow pack and vegetation. Further details of the DEHM model are described elsewhere (Christensen, 1997; Christensen et al., 2004; Hansen et al., 2008, 2015; Hedegaard et al., 2008; Brandt et al., 2012). The DEHM model is usually driven with meteorological data from numerical weather prediction models and is currently coupled

to data from the numerical weather prediction model MM5v3.7 (fifth-generation mesoscale model) developed by Grell et al. (1995). That model was driven by global data from the European Centre for Medium-Range Weather Forecasts (ECMWF) until the year 2000, and thereafter by data from the National Centers for Environmental Prediction (NCEP). The DEHM model also has a climate mode, where the input is climate meteorological data calculated from a global circulation model (ECHAM5/MPI-OM) that simulates the SRES (Special Report on Emissions Scenarios) A1B climate scenario (IPCC, 2000).

The POPs and temperature results from the DEHM model were used as input parameterization for the AQUAWEB bioaccumulation food-web model by extracting time series of monthly water and air-dissolved contaminant concentrations for γ -HCH, PCB52 and PCB153, and temperature. The AQUAWEB model that was previously parameterized for an Arctic marine food web (Arnot and Gobas, 2004; Borgå et al., 2010) was parameterized for the Kongsfjorden food web on Svalbard.

Although seasonal variations in the long-range transport of POPs are of great importance, the implications for food-web biomagnification have been poorly studied. The combination of the DEHM and AQUAWEB models allows better simulations of the uptake and fate of pollutants in an Arctic marine food web. There are some unknown uncertainties in model calculations of POPs transport, mainly related to the simple 75-m-thick ocean compartment module used in DEHM. This module does not include ocean transport, which could be an important route for contaminant transport to Svalbard. Atmospheric transport and deposition are the only modes of POP transport to the Arctic used in the model calculations.

The DEHM model was initially run with real meteorological input data from MM5¹ for a 25-year period (1989–2013) to validate the model system and to build up non-zero initial contaminant conditions for the later climate-related model runs. From time-series outputs of the DEHM model, the 2007 monthly air and water dissolved contaminant concentrations of γ -HCH, PCB52 and PCB153 and temperature were used as input to the AQUAWEB bioaccumulation model. The DEHM model was also run in the climate mode, where climate meteorological data estimated from ECHAM5 for the decades 1990–1999 and 2090–2099 were used as input (AMAP, 2011a). Based on the model output, monthly temperature and contaminant exposure in the two decades were estimated. Constant emissions with initial concentrations obtained from the output for the year 2011 for model runs with MM5 meteorological data were assumed. It is possible to estimate the relative changes in POPs transport with the DEHM model in the Arctic only due to the changes in climate by comparing results for the two decades 1990–1999 and 2090–2099, where the model was run using exactly the same emissions data. It is expected that changes due to decreases in emissions in the future will be much greater than changes due to the climate effect. The

¹www.mmm.ucar.edu/mm5/mm5-home.html

model was previously applied to study the effect of climate change on future atmospheric levels of ozone and particulate matter and also recently to study the effect on POPs in the Arctic (Hedegaard et al., 2008, 2012, 2013; Langner et al., 2012; Simpson et al., 2014; Hansen et al., 2015). From these climate model outputs, ratios of change were estimated for each month (RoC_{Month}) using the mean ratio of the last five years of the simulations for contaminant concentrations ($C_{Month2095-2099}$, $C_{Month1995-1999}$) (Eq. 1).

$$RoC_{Month} = \left(\frac{C_{Month,2095-2099}}{C_{Month,1995-1999}} \right) \quad \text{Eq. 1}$$

Using these RoC coefficients, concentrations for the respective phases in 2107 were estimated from the modeled concentrations in 2007 (Eq. 2), and used as input to the AQUAWEB bioaccumulation model (Arnot and Gobas, 2004).

$$C_{Month,2017} = C_{Month,2007} \times RoC_{Month} \quad \text{Eq. 2}$$

Changes in temperature simulated by the DEHM model were implemented using the Δ change approach; mean values of monthly temperature changes were calculated from the model outputs for 2095–2099 versus 1995–1999 and were added to the simulated temperatures for 2007.

2.4.2 AQUAWEB

The AQUAWEB model (Arnot and Gobas, 2004) was modified to reflect the Arctic marine pelagic food web, as described by Borgå et al. (2010). The main concept of the model is to estimate

the net accumulation of POPs in biota based on competing rates of uptake and elimination, including two uptake rate constants (direct respiratory and dietary uptake) and four elimination rate constants (direct respiratory, excretion, biotransformation and growth rate). The model was parameterized to reflect the local food web of Kongsfjorden, Svalbard, including algae, herbivorous and carnivorous zooplankton and fish (Table 2.1). The food-web model comprises a set of differential equations with all relevant variables interpolated in a circular fashion inside the solver using either monthly values (from the DEHM model) or values from observations, that is, May, July and October. The ‘year’ 2007 was repeated 50 times to reach concentrations in biota that did not increase over the year (i.e., so that end of December = start of January).

Temperature dependencies in parameters, uptake and elimination rates were implemented as described by Arnot and Gobas (2004) and Gewurtz et al. (2006). For the dynamic simulations, lipid fraction and temperatures (and thus partitioning coefficients) are dynamic variables. Unless defined specifically, seasonal dynamics of the parameters were interpolated by a sine wave for parameters with seasonal variation, defining the Julian days and values of the amplitude peak and/or low. The zooplankton and fish species included are the same as were analyzed seasonally for contaminants, lipid content and dietary descriptors (stable isotopes of nitrogen $\delta^{15}\text{N}$ and carbon $\delta^{13}\text{C}$) in 2007 (Hallanger et al., 2011).

The bioaccumulation modeling focused on γ -HCH, PCB52 and PCB153 because their physical-chemical properties span a wide range, resulting in different temperature dependence, volatilization, water solubility, persistence in the environment and biota, bioaccumulation, and other environmental behavior. The physical-chemical properties

Table 2.1. Kongsfjorden food-web parameterization of the bioaccumulation model AQUAWEB. The parameter values listed reflect static scenario.

Species name	Body weight, g	Feeding rate, g food / g bodymass / day	Growth rate, 1/day	NLOM fraction, kg/kg	Lipid diet assimilation efficiency,%	NLOM diet assimilation efficiency,%
Algae	1×10^{-6}	0	0.08	0.1	-	-
<i>Calanus finmarchicus</i>	0.001	-	0.0067	0.17	0.72	0.72
<i>Calanus glacialis</i>	0.0023	0.1629	0.0067	0.17	0.72	0.72
<i>Calanus hyperboreus</i>	0.012	0.1278	0.0048	0.17	0.72	0.72
<i>Thysanoessa inermis</i>	0.08	0.0971	0.0033	0.18	0.75	0.75
<i>Themisto libellula</i>	0.30	0.0789	0.0025	0.18	0.75	0.75
<i>Themisto abyssorum</i>	0.15	0.0789	0.0025	0.18	0.75	0.75
Chaetognatha	0.25	-	-	0.18	0.75	0.75
Polar cod	160	0.0433	0.0011	0.16	0.92	0.6
Saithe	47	0.031	0.0011	0.16	0.92	0.6
Capelin	160	0.031	0.0011	0.16	0.92	0.6

NLOM: Non-Lipid organic matter

Table 2.2. Physical-chemical properties of the model chemicals at 25°C.

Chemical name	Molar mass	Log K_{AW} ^a	Log K_{OW} ^b	Log K_{OA} ^c	ΔU_{OA} (kJ/mol) ^d	ΔU_{OW} (kJ/mol) ^e
PCB52	292.0	-2.0	6.0	8.2	-81.3	-27.5
PCB153	360.9	-2.1	6.9	9.5	-94.8	-26.6
γ -HCH	290.9	-4.0	3.8	7.7	-64.0	-9.6

^aAir-water partitioning factor; ^boctanol-water partitioning factor; ^coctanol-air partitioning factor; ^dinternal energy of phase transfer between octanol and air; ^einternal energy of phase transfer between octanol and water.

applied in the bioaccumulation model were the same as in the DEHM model (Table 2.2).

In the present study, the whole body lipid content of the respective species in Kongsfjorden was assumed to vary seasonally as measured in specific matrices throughout the food web, extrapolating between the measured values to obtain the annual variation (Hallanger et al., 2011).

The most likely dietary relationships between species were determined for spring, summer and autumn/ winter, based on maximum likelihood estimation of the food web using $\delta^{15}N$ measured in the various species (Hallanger et al., 2011), adapted from Starrfelt et al. (2013) using Monte Carlo Markov Chain (MCMC) analysis.

2.4.3 CoZMoMAN

CoZMoMAN is a time-variant multimedia mechanistic model that simulates and predicts environmental fate and human food chain bioaccumulation and levels for POPs in different birth cohorts (Breivik et al., 2010). The model includes both a marine and a terrestrial human food chain and is based on emission data (Breivik et al., 2010).

The model calculations describe how emissions of contaminants are transported and distributed in the environment and predict concentrations in environmental compartments (air, water, soil, sediment) and organisms in an aquatic and an agricultural food chain (e.g., grass, cows, fish). On the basis of time-variant emission estimates from 1930 to 2010, concentrations in air, water and tissues of organisms are simulated and then human intake rates determined. Thus, human contaminant concentrations can be estimated for a single time point or period. The required input information is birth year, year of sampling, and daily intake of dairy products, meat and fish. For women, their age at child birth(s) and breastfeeding duration for each child must also be specified. The original model domain includes Sweden and parts of southeastern Norway.

Modeling studies can contribute to the formulation and testing of hypotheses concerning the impact of regulatory measures and behavioral changes on human PCB exposure. Comparing observations of human concentrations with model estimates thus provides valuable model evaluations.

2.5 Heavy metals

Metals are naturally occurring elements. They are found in elemental form in many chemical compounds. Each form or compound has different properties, which affect how the metal is transported, what happens to it in the food web and its toxicity. Being elements, metals cannot be broken down into less toxic substances in the environment. Some metals are even essential nutrients in low concentrations. The metals of most concern regarding effects in the Arctic are mercury (Hg), cadmium (Cd) and lead (Pb) because they have no known biological function, but they bioaccumulate in organisms and are toxic. Environmental Pb levels appear to have stabilized after years of decline following the switch to unleaded fuel in most countries.

Heavy metals can be toxic even in small quantities and are present at high levels in regions remote from most anthropogenic sources, such as the Arctic. Mercury is of particular concern in the Arctic because of its ability to bioaccumulate and biomagnify in food chains as organomercury, with methylmercury (MeHg) the most important (AMAP, 1998, 2011b; Macdonald et al., 2005; Ruus et al., 2015). Through the 1998 Protocol on Heavy Metals under the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP), governments are taking measures to minimize and prevent emissions of Cd, Pb and Hg by regulating combustion and industrial processes as predominant anthropogenic sources of their emission, in line with the precautionary approach. Building on the 1998 Protocol on Heavy Metals, the Minamata Convention on Mercury was adopted in 2013, a treaty negotiated under the auspices of the United Nations Environment Programme (UNEP). The Minamata Convention on Mercury is a global treaty to protect human health and the environment from the adverse effects of Hg. The major content of this treaty includes a ban on new Hg mines, the phase-out of old Hg mines, control measures on air emissions, and international regulation of the informal sector for artisanal and small-scale gold mining. As of April 2016, 128 states had signed and 25 had ratified the Convention.

This report focuses mainly on Hg. Atmospheric deposition represents an important input of Hg to land and water, and has increased the amount of Hg in many environmental compartments by a factor of two to three since the start of the industrial age (Mason et al., 2012).

The terrestrial environment is important in the regional Hg cycle because Hg deposited on land is largely retained in soils and vegetation. Changes in land cover and land use have a large impact on Hg mobilization when exposed soils erode and Hg

leaches into surrounding freshwaters and marine waters (Stern et al., 2012). Snow, ice, and soils on land are key reservoirs for atmospheric deposition and, in turn, can become sources of Hg through melting of terrestrial ice and snow and via soil erosion and re-emissions.

Mercury can undergo several biogeochemical transformations within snow, ice and soil that influence its fate. The main reactions are the oxidation or reduction of inorganic Hg and the formation or decomposition of MeHg. The reduction of Hg(II) to Hg(0) is a transformation of fundamental importance because of its role in emitting Hg back into the atmosphere. Microbial methylation is also a key process that transforms Hg(II) into its more toxic organic form. Mercury enters terrestrial food webs through uptake in vegetation. Lichens, which are an important food for many Arctic herbivores, have no root system and accumulate Hg mainly from the atmosphere (Estrade et al., 2010). Below the tree line, foliage of trees and shrubs also accumulates Hg mainly from atmospheric deposition directly onto their leaves rather than through uptake via roots (Grigal, 2002; Erickson et al., 2003; Graydon et al., 2008). Trees and shrubs play an important role in scavenging Hg and increasing its flux to snow and soils via throughfall and litterfall (Poullain et al., 2007b; Graydon et al., 2008).

2.5.1 New emissions and re-emissions of mercury

Current levels of Hg affecting Arctic ecosystems are a legacy of anthropogenic emissions that began at the start of the industrial era. Present-day emissions to the atmosphere are dominated by re-emissions from natural surfaces; 60% of the Hg entering the atmosphere is released from natural surfaces (soil, oceans, etc.) but comprises Hg previously deposited to these surfaces and is a mixture of anthropogenic and naturally occurring Hg. Natural emissions (i.e., from weathering of rocks, volcanoes, and geothermal activity) account for about 10% of total emissions to the atmosphere (UNEP, 2013). Another approximately 30% is of anthropogenic origin, with the main sources including fossil fuel combustion, smelting and production of ferrous and nonferrous metals, cement production, oil refining, artisanal and small-scale gold mining (ASGM), and consumer products waste. ASGM is the largest source followed by fossil fuel (mainly coal) combustion within this group (Fig. 2.1). Re-emission is a natural process,

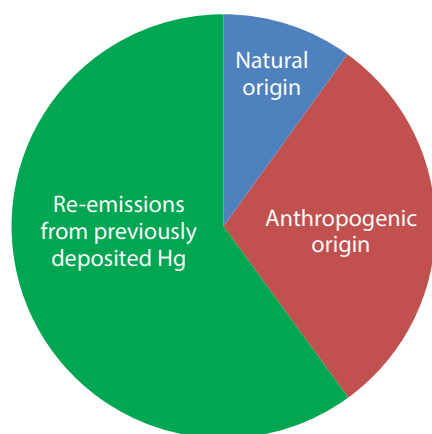


Figure 2.1. Relative distribution of the major categories of Hg emission to the atmosphere. Source: UNEP (2013).

and Hg can be deposited and re-emitted many times as it cycles through the environment. Re-emission is not considered a natural source, although it may originate from a natural source, because by the time Hg is being re-emitted it is impossible to identify its specific origin. Nonetheless, human activity has increased the environmental burden of Hg, resulting in higher levels of re-emission. Re-emission is compounded by changes in land use practices as well as rising temperatures owing to climate change.

Estimating re-emission and re-mobilization rates is difficult. It is often done using modeling approaches. These models are based on data on atmospheric levels and other observations as well as current understanding of chemical transformations and other processes that determine how Hg moves between air, land and water. The models aim to balance the amount of Hg in circulation at any given time while remaining consistent with observational data. Temperature is a key factor; re-emission rates are generally lower with lower temperatures, even if indications of short-term improvements will be difficult to determine in, for example, Arctic ecosystems.

Re-emission is also a major factor in determining the length of time needed for anthropogenic emission reductions to be reflected in decreasing environmental levels of Hg. Mercury emitted to the atmosphere in any one year may be deposited or retained in soil and water for some time before being re-emitted or re-mobilized in later years. This cycle can be repeated, keeping levels in air and water elevated even after emissions from anthropogenic sources have been reduced. A continued addition of Hg to the global pool will leave an ever-longer legacy of anthropogenic Hg contamination worldwide. It is thus very important that ongoing international efforts to reduce Hg emissions continue and are strengthened.

2.5.2 Mercury in water

The pathways and fate of Hg in aquatic environments are important because it is in water, sediments and wetland soil that inorganic Hg is converted into MeHg, which is toxic and bioaccumulates in animals (Macdonald and Loseto, 2010). The major human exposure to Hg and the related health risks come from the consumption of marine foods (UNEP, 2013). In the aquatic environment, re-mobilization of Hg occurs when Hg deposited and accumulated in soil or sediments is re-mobilized, for example, by rain and floods that cause Hg to re-enter the aquatic system. Re-suspension of aquatic sediments due to wave action or storm events is an additional way for Hg to re-enter the aquatic ecosystem.

Mercury concentrations in the upper 100 m of the oceans are twice those of a century ago. Intermediate and deeper waters have, on average, 10–25% more Hg, which reflects the slow transport of Hg downwards in the oceans (Mason et al., 2012). Seawater concentrations will thus be especially slow to respond to changes in Hg inputs from atmospheric deposition and river flow. As a result, Hg concentrations in marine biota are likely to remain elevated or increase further for decades to centuries even without an increase in atmospheric emissions. In freshwater ecosystems, atmospheric deposition and re-mobilization from soil slow any reduction in Hg levels, even

in regions where atmospheric concentrations have decreased due to emission controls. It is likely to be years or decades before reductions in anthropogenic emissions and releases of Hg have an effect on Hg levels throughout the environment and in the fish and marine mammals which are part of the human food chain (Mason et al., 2012). At present, concentrations of Hg in marine animals in the Arctic are about 10 to 12 times higher than during the pre-industrial period. This means that roughly 92% of the Hg in marine predators such as seabirds, seals, and whales is of anthropogenic origin. The timing of the increase suggests that the Hg originates from emissions during past decades in Europe, North America and Russia (Dietz et al., 2009).

2.5.3 Atmospheric transport, cycling and surface fluxes

The atmosphere plays a fundamental role in the Hg cycle through long-range atmospheric transport to the Arctic and local deposition onto terrestrial, freshwater, and marine surfaces within the Arctic. Much of the transport of Hg to the Arctic occurs during winter and spring. The transport is influenced by atmospheric circulation patterns such as the Pacific North American Oscillation (PNAO) and the North Atlantic Oscillation (NAO). During the past decade, the Arctic atmospheric pressure patterns have been characterized by anomalously high sea-level pressure on the North American side of the Arctic and low pressure on the Eurasian side, which has supported more southerly winds (Overland et al., 2008). Altered atmospheric pressure fields imply the possibility of altered patterns of atmospheric transport of Hg into and out of the Arctic (Macdonald et al., 2005). Direct transport of emissions from Europe is recorded in Norway and the Norwegian Arctic; however, the same is not observed from Asia, because the higher surface temperatures there do not favor such a low-altitude transport into the Arctic (the emission areas in Asia are outside the Polar Front) (Hirdman et al., 2009). Although no direct transport is observed from Asian sources, they do contribute significantly to the overall Hg level observed (Durnford et al., 2010). There has been little change in Hg transport pathways and circulation patterns over the past 30 years, but climate change is expected to alter that, which will cause more Hg to be transported into the Arctic (Hirdman et al., 2010).

Concentrations of Hg in the atmosphere are normally too low to present a risk of adverse health effects for humans. The concern for Hg in the atmosphere is primarily related to its long-range transport potential, its subsequent deposition, conversion to MeHg, and thereafter accumulation in biota (AMAP, 2011b). Because Hg can be re-emitted, the transport pattern is complex. Consequently, it is important to investigate the surface-related chemistry of Hg by determining the fluxes of different Hg species over different surfaces. Much effort has been expended to understand the levels, transport, and transformations of Hg in Arctic air and the role of the atmosphere in this region.

Most Hg is emitted to the atmosphere in the form of gaseous elemental mercury (GEM), which is slowly oxidized. Hence, it has a long residence time in air (6 to 24 months), and easily enters the global 'atmospheric pool' where it can be transported worldwide. GEM is therefore found at similar concentrations

throughout the northern hemispheric troposphere. However, during short-term events shortly after polar sunrise in spring, GEM can be rapidly depleted from the lower atmosphere through oxidative processes and deposited on the ground or physically bound to aerosol surfaces (Schroeder et al., 1998). This transformation is referred to as an 'atmospheric mercury depletion event' (AMDE) and has sparked enormous interest in atmospheric Hg and the role of AMDEs in the Arctic Hg cycle. AMDEs are characterized by a series of photochemically-initiated reactions in the atmosphere which result in the conversion of GEM to short-lived forms of Hg that can be deposited on terrestrial and aquatic surfaces. Conversion into these short-lived oxidized forms of gaseous oxidized Hg (GOM) and particulate Hg (HgP) will enhance deposition to soil and water surfaces. Some of this deposited Hg will be re-emitted, but a significant proportion can be further transported to aquatic environments and methylated, which will potentially lead to bioaccumulation of MeHg in biota and humans. It is important to understand that GOM and HgP are fractions of Hg that are operationally defined by the analytical methods. Primary emitted GOM and HgP are therefore not necessarily the same Hg species as the GOM and HgP present elsewhere in the atmosphere.

Scientific efforts to characterize the atmospheric transport and delivery of Hg to the Arctic are challenging because Hg emissions to air occur from natural sources and anthropogenic activities, and complex exchanges of Hg occur between the air and its interfaces with soil, water, and the cryosphere.

2.6 Field work and analyses

This report combines model projections and empirical data collected from terrestrial and freshwater food webs. PFASs have been analyzed in birds, reindeer and Arctic char from Svalbard (Fig. 2.2). Blood samples from people in northern Norway and Russian areas near Norway have been analyzed for PCBs, pesticides and heavy metals. Other studies have



Figure 2.2. Svalbard as a research platform for the majority of the field studies reported here © Norwegian Polar Institute.

focused on marine food webs (Øverjordet et al., 2015a,b; Ruus et al., 2015).

2.6.1 Particle transport and characterization

While several of the legacy organic contaminants undergo long-range transport in the gas phase (UNEP/AMAP, 2011), some of the higher molecular weight compounds (such as greater than four-ring polycyclic aromatic hydrocarbons, PAHs; octa-deca-PCBs; and octa-deca polybrominated diphenyl ethers, PBDEs) and fluorinated chemicals are primarily associated with particles in the air (de Wit et al., 2010). In addition, local sources of particles and pollution exist in the Arctic, and there is a need to determine the impact of local sources compared to sources of particles arriving via long-range transport (Hung et al., 2005, 2010; Shen et al., 2006; Stock et al., 2007; von Waldow et al., 2010; Xiao et al., 2012; Li and Jia, 2014). Characterization of particles can provide insight into the sources, both to differentiate between different local sources and to identify contributions due to atmospheric transport from southern latitudes (e.g., combustion-derived particulate matter carried into the High Arctic via prevailing meteorology from industrial regions further south, which concentrates in the atmosphere in late winter/early spring causing Arctic Haze). The origin of the particles can be determined from their size, morphology and chemical composition. Particles are deposited onto the ground via wet deposition (precipitation) and dry deposition, with particle deposition serving as a source of deca-BDE to Arctic snowpacks (Meyer et al., 2012). Changes in weather systems can therefore affect the deposition of particles and associated POPs (UNEP/AMAP, 2011). Hence, there is increasing interest in and need for more knowledge about particle-facilitated POPs transport to the Arctic (Weinbruch et al., 2012).

2.7 Levels of contaminants in humans in the Arctic

Longitudinal monitoring studies of POPs in human populations are important to better understand changes with time and age, and for future predictions. Studies on the health effects of PCBs require an understanding of past and present human exposure. Time-resolved models may supplement information on concentrations in individuals obtained from measurements and/or statistical approaches, if they can be shown to reproduce empirical data (Breivik et al., 2010; Nøst et al., 2013).

As part of this project, the CoZMoMAN model was used in person-specific predictions of life course concentrations of PCBs in individual Norwegians (Nøst et al., 2013). The CoZMoMAN model was evaluated by reproducing measured time-variant concentrations of PCBs in environmental compartments, local food items, and human breast milk. Person-specific predictions of contaminant levels were also made using statistical models that linearly regressed PCB concentrations against dietary

and lifestyle variables (Nøst et al., 2013). In addition, age-period-cohort effects were investigated, and predicted PCB concentrations were compared to measured values as part of the model.

Two comprehensive studies have recently been conducted in northern Norway: the Northern Norway Mother-and-Child Contaminant Cohort study (MISA study) and the Tromsø Study (an ongoing long-term study). The MISA study was designed to investigate maternal concentrations of organic contaminants and metals in the context of a northern-southern latitude perspective, to identify exposure predictors and to investigate the influence of physiological changes and related pregnancy adaptations during the gestational and postpartum periods. The work took place between 2007 and 2009, and included pregnant and delivering women from northern Norway (n=516). A suite of selected organochlorine contaminants, five toxic metals (arsenic, Cd, Pb, Hg, cobalt) and five essential elements (copper, manganese, zinc, molybdenum, selenium) were analyzed (Hansen et al., 2010).

The Tromsø Study explored changes in POPs concentrations from 1979 to 2008 by following individuals over this period. Five surveys took place in Tromsø (northern Norway), in 1979, 1986–1987, 1994–1995, 2001 and 2007–2008. Sixty adult men were randomly selected from a total of 1438 participants, with the qualifier that they had donated blood in all five surveys. Fifty-three of the randomly selected men had sufficient remaining sample volumes in more than three sampling events to measure contaminants. The range in birth years was from 1925 to 1950, and the median ages at first and last sampling were 43 and 71, respectively (Nøst et al., 2013). Calendar year (periodic) changes and influences of age and birth cohorts were assessed by graphical and mixed effects analyses. The concentrations of four PCBs were thereafter predicted for each sampling year by the CoZMoMAN model.

The rank correlation between measurements and predictions from both the CoZMoMAN model and regression analyses was strong (Spearman's $r > 0.67$). Simulations indicated large inter-individual differences in concentrations experienced in the past. Agreement between measurements and predictions of concentrations, subject ranking and quartile assignment was good. Contamination histories for individuals predicted by the CoZMoMAN model revealed variation between study subjects, particularly in the timing of peak concentrations. Realistic *a priori* assessments of PCB exposure by mechanistic models provide individual PCB exposure metrics that could serve as valuable supplements to measurements. The time-variant model CoZMoMAN has been useful in estimating prenatal, postnatal and childhood exposure to PCB153 under scenarios of hypothetical and realistic maternal fish consumption (Binnington et al., 2014).

3. Persistent organic pollutants

The chemical lifespan and environmental stability (persistence) of POPs are temperature dependent. Therefore, it is possible that increased transport to the Arctic under a future warmer climate is compensated by a shorter lifetime of the POPs due to the higher ambient temperature and higher hydroxyl radical (strong oxidant) concentrations in the atmosphere. However, there are several processes that affect the fate of POPs in the Arctic. By combining models, empirical data and toxicological investigations these processes may be better understood.

3.1 Climate change effects on atmospheric transport of persistent organic pollutants

Many factors affect the environmental fate of POPs; however, the combined effect of climate change on these factors is difficult to predict. Rising temperatures may cause enhanced revolatilization of deposited compounds from sinks such as soil, snow and ice caps. This can also alter the partitioning between different media. The precipitation pattern in the Arctic is expected to change, which will modify pollutant deposition patterns. Over a longer period, changes in ocean currents may affect ocean transport (AMAP, 2011c; Kallenborn et al., 2012;

Grannas et al., 2013). Finally, higher temperatures will lead to more rapid degradation rates. A chemical database prepared for the ArcRisk project provides the values for the temperature dependence of degradation rates that were used in this study, but these values are associated with considerable uncertainty. By using the meteorological output from ECHAM5 for two periods, 1990–1999 and 2090–2099, the DEHM model was used to predict some of these effects by comparing the two ten-year model runs described in Sect. 2.4.1.

Figures 3.1 to 3.3 show atmospheric surface concentrations of γ -HCH, PCB52 and PCB153 for the period 1995–1999 and the percentage change between 1995–1999 and 2095–2099. The highest concentrations of γ -HCH are over the emissions area in Europe and especially Asia, and the lowest are over the Arctic Ocean. In most of the model domain the atmospheric concentrations of γ -HCH are higher in 2095–2099 than in 1995–1999, up to 20% higher over some parts of the Arctic Ocean and more than 30% to 40% higher over the northern part of the Pacific Ocean, while the increases for PCB52 and PCB153 are less. These increases are associated with increased volatilization of γ -HCH in 2095–2099 compared to 1995–1999 (Fig. 3.1, right). The highest concentrations of PCB52 are over the emissions areas in North America and Europe (Fig. 3.2, left) and the graphic shows that there is some direct

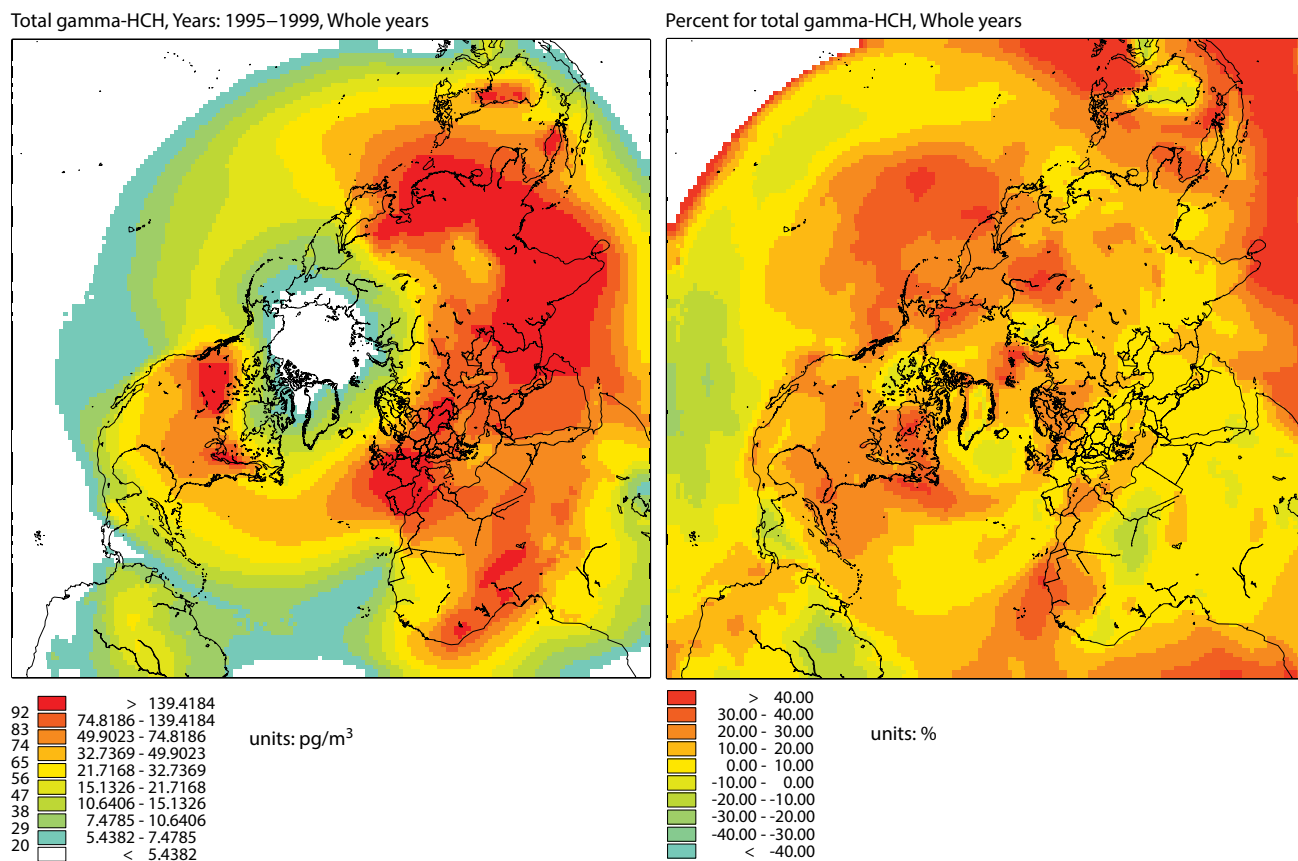
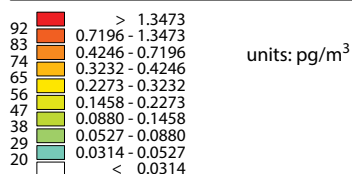
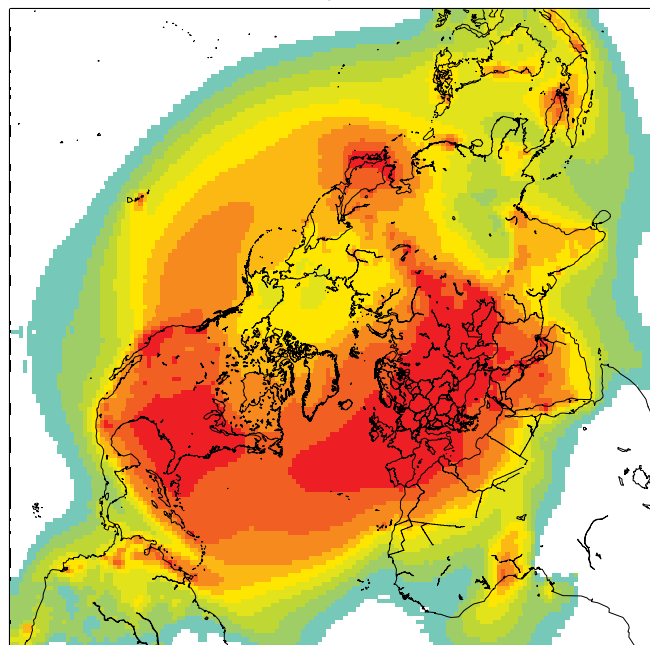


Figure 3.1. Total annual mean concentration of γ -HCH in the lowest atmospheric model layer for the period 1995–1999 (left), and the percentage change between the mean for 1995–1999 and the mean for 2095–2099 (right).

Total PCB52, Years: 1995–1999, Whole years



Percent for total PCB52, Whole years

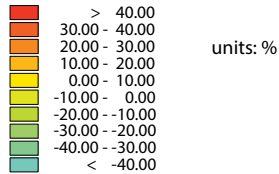
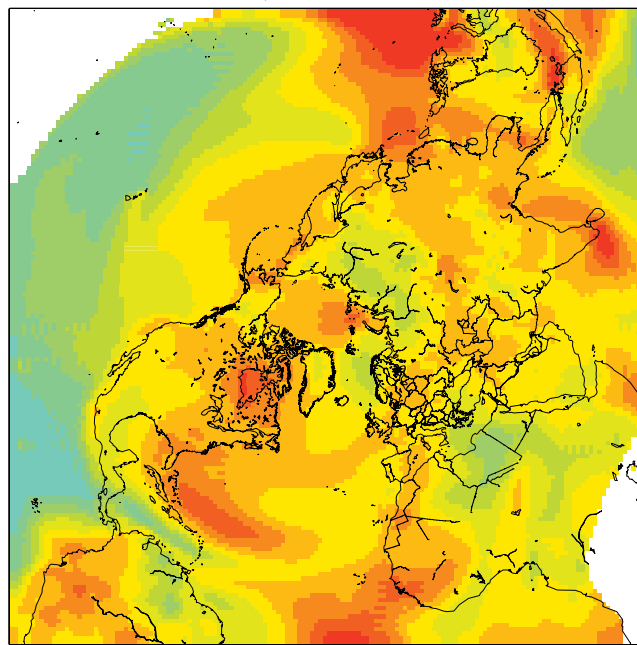
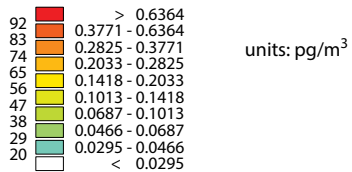
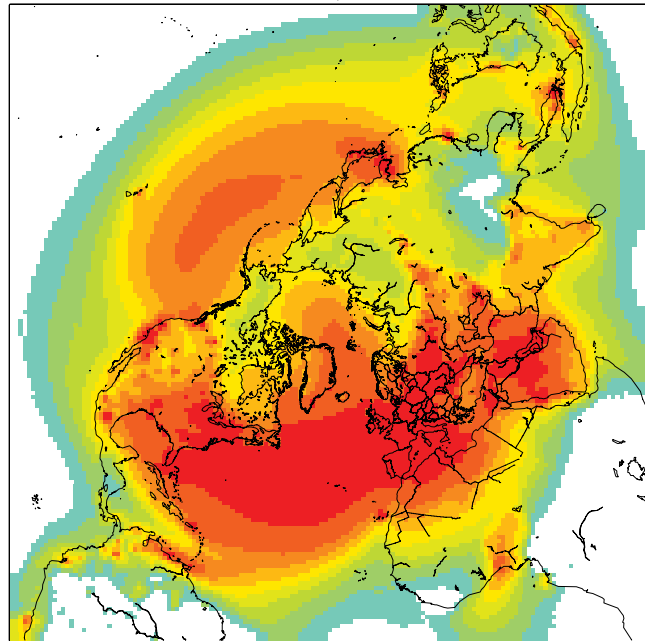


Figure 3.2. Total annual mean concentration of PCB52 in the lowest atmospheric model layer for the period 1995–1999 (left), and the percentage change between the mean for 1995–1999 and the mean for 2095–2099 (right).

Total PCB153, Years: 1995–1999, Whole years



Percent for total PCB153, Whole years

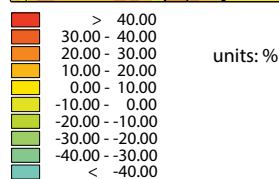
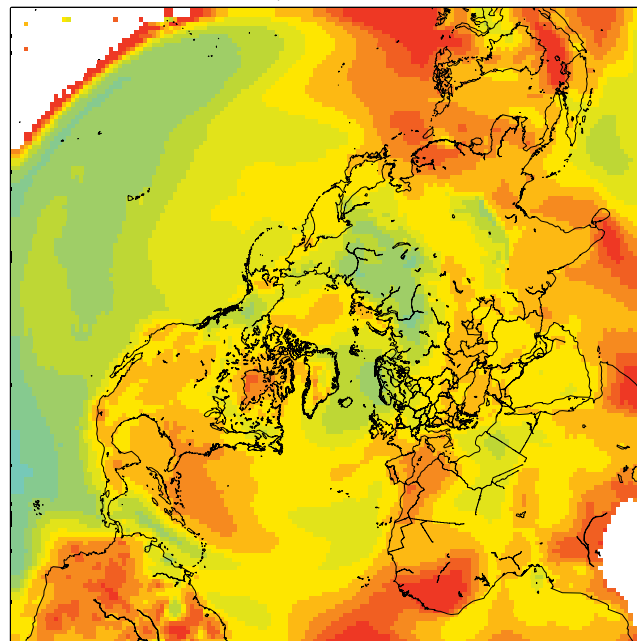
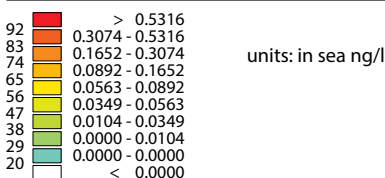
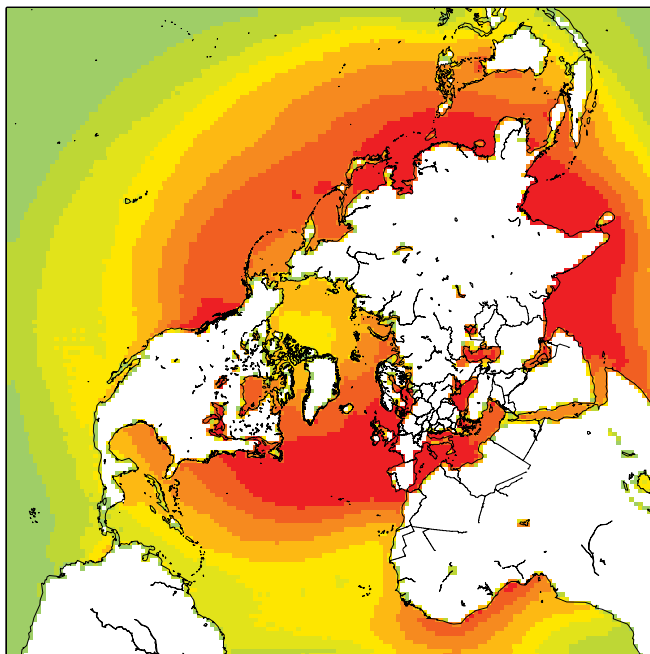


Figure 3.3. Total annual mean concentration of PCB153 in the lowest atmospheric model layer for the period 1995–1999 (left), and the percentage change between the mean for 1995–1999 and the mean for 2095–2099 (right).

atmospheric transport into the Arctic. Emissions in Europe are more favorable for atmospheric transport to the Arctic compared to emissions in Asia (Christensen, 1997). Figure 3.2 (right) shows higher atmospheric PCB52 concentrations in 2095–2099 compared to 1995–1999, but the increase is not as large as for γ -HCH. In some areas over northern Siberia and the North Atlantic, there is even a decline in PCB52, which is probably due to higher particle deposition rates owing to reduced snow and ice cover that increases dry deposition, and higher precipitation rates in the Arctic. Model results for PCB153 (Fig. 3.3) are similar to those for PCB52, although PCB153 shows a more pronounced decrease owing to its stronger association with particles. Model results for sulfate (not shown) indicate a decline in concentration over the whole Arctic. The overall conclusions are that increased volatilization due to higher temperatures increases atmospheric transport, especially for γ -HCH, and declining snow/ice cover and higher precipitation rates increase the deposition of particle-bound POPs such as PCB52 and PCB153. Furthermore, higher degradation rates in soil under warmer conditions would reduce POPs concentrations in the atmosphere by decreasing volatilization from the soil.

Figures 3.4 to 3.6 show concentrations of γ -HCH, PCB52 and PCB153 in the upper 75-m ocean layer for the period 1995–1999 and the projected percentage change between 1995–1999 and 2095–2099. The highest concentrations of γ -HCH (Fig. 3.4) are downstream from emissions areas, with ocean concentrations increasing in the mid- and higher latitudes. The spatial distribution of ocean PCB52 (Fig. 3.5) and PCB153 (Fig. 3.6) concentrations from lower to higher latitudes is due to the temperature gradient. This indicates that PCB concentrations in ocean water are to some extent in equilibrium with atmospheric concentrations. The changes between 1995–1999 and 2095–2099 for both PCB52 and PCB153 show a decrease, especially around Svalbard. The main reasons for this is that volatilization from the ocean in the Arctic will increase due to the higher surface temperature of the ocean as well as the decreased ice cover, and to some degree also the higher degradation rates in soil. The increased atmospheric transport owing to the greater volatilization is not large enough to compensate for the increased volatilization from the Arctic Ocean and the more rapid degradation of the contaminants.

Total gamma-HCH, Years: 1995–1999, Whole years



Percent for total gamma-HCH, Whole years

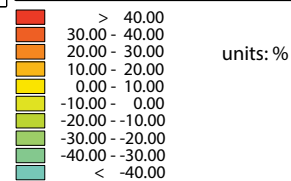
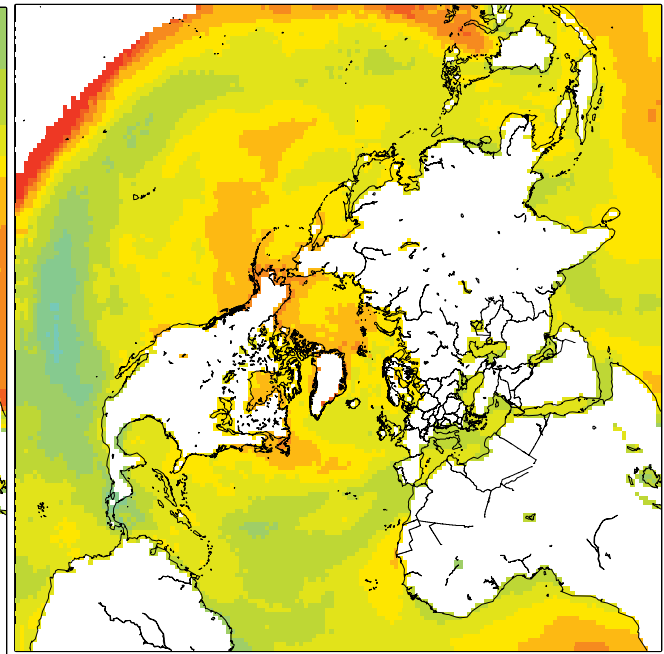


Figure 3.4. Total annual mean concentration of γ -HCH in the ocean model layer for the period 1995–1999 (left), and the percentage change between the mean for 1995–1999 and the mean for 2095–2099 (right).

Total PCB52, Years: 1995–1999, Whole years

Percent for total PCB52, Whole years

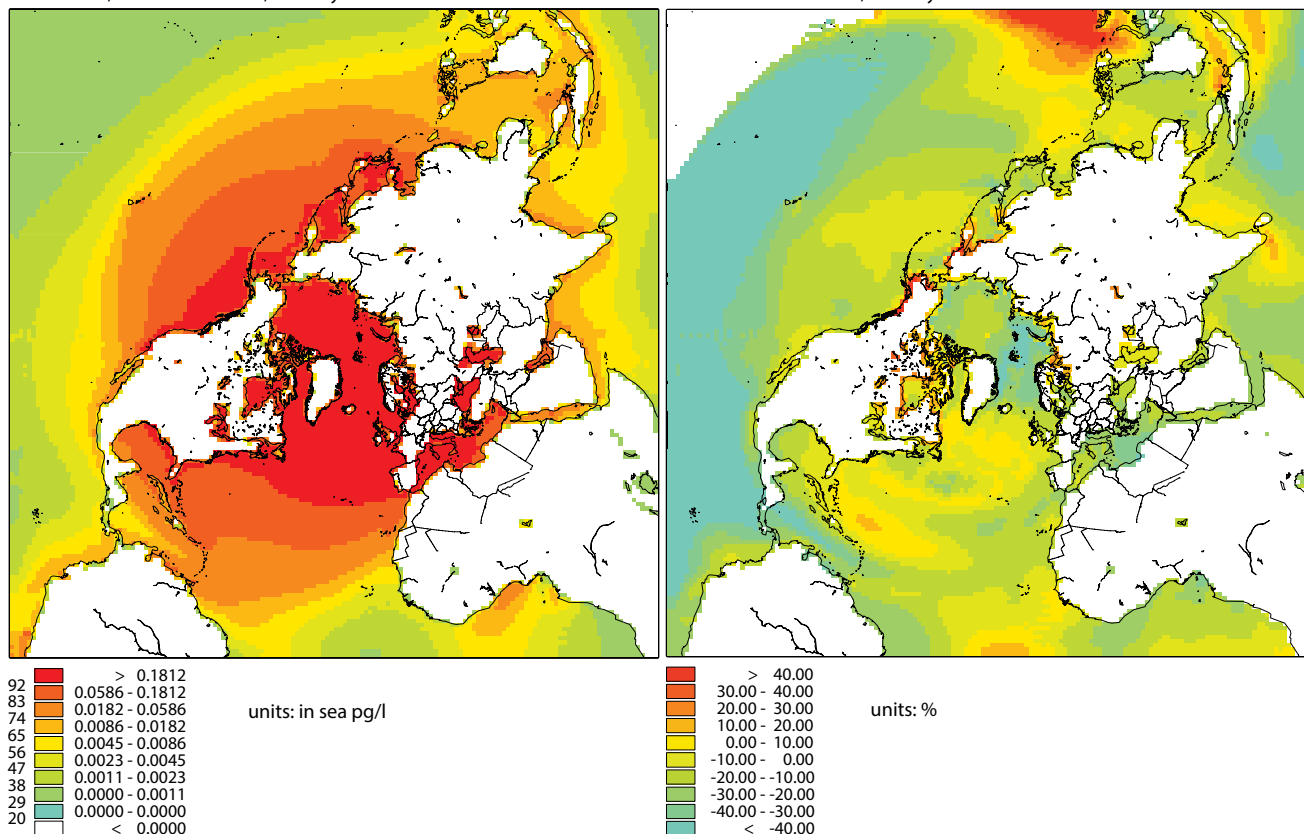


Figure 3.5. Total annual mean concentration of PCB52 in the ocean model layer for the period 1995–1999 (left), and the percentage change between the mean for 1995–1999 and the mean for 2095–2099 (right).

Total PCB153, Years: 1995–1999, Whole years

Percent for total PCB153, Whole years

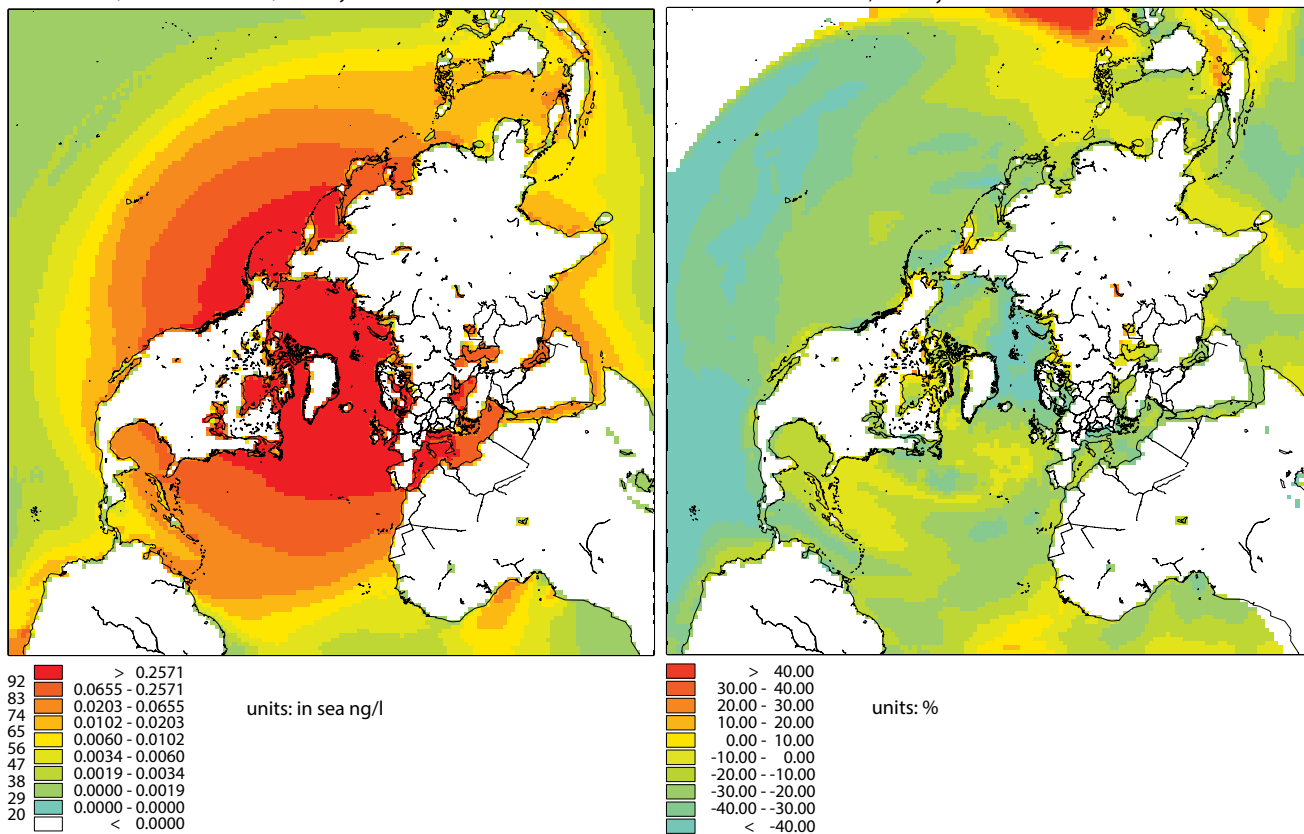


Figure 3.6. Total annual mean concentration of PCB153 in the ocean model layer for the period 1995–1999 (left), and the percentage change between the mean for 1995–1999 and the mean for 2095–2099 (right).

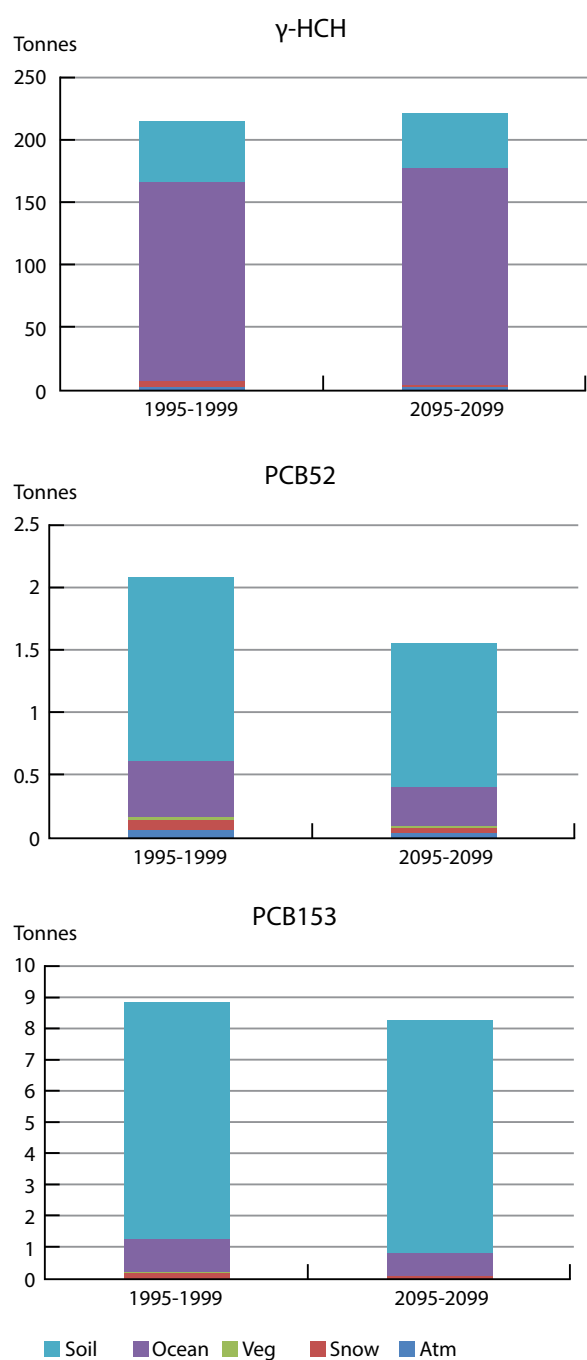


Figure 3.7. Distribution of the mean of the total mass of γ -HCH, PCB52 and PCB153 between five environmental media (soil, ocean, vegetation (veg), snow, atmosphere (atm)) in the Arctic for the periods 1995–1999 and 2095–2099.

Figure 3.7 shows the distribution of the total mass of γ -HCH, PCB52 and PCB153 in the five different media (soil, ocean, vegetation, snow, atmosphere) represented in the DEHM model for the Arctic for the periods 1995–1999 and 2095–2099. It is clear that most of the mass of the contaminants occurs in the oceanic and soil compartments. Only a minor proportion is present in the atmosphere and in the other two compartments (vegetation and snow). There is a large difference between γ -HCH and the two PCBs, which can be explained by their water solubility. While γ -HCH is water soluble (most of the mass is in seawater), most of the mass of the two less hydrophilic PCBs is bound to organic matter in soil. There is a small increase in the total mass of γ -HCH between 1995–1999 and 2095–2099 owing to a small decrease in the soil and a larger increase of total mass in the ocean. The main reason for this is that the warmer climate of 2090–2099 increases the atmospheric concentration owing to the greater volatilization of γ -HCH, which can also be seen in Fig. 3.1. This drives an enhanced transport of γ -HCH to the Arctic Ocean water (Fig. 3.4). There is a decrease between 1995–1999 and 2095–2099 in the mass of both PCB52 and PCB153, although this is most pronounced for PCB52. This is mainly because higher temperatures shorten the lifetimes of PCBs owing to more rapid degradation and increase volatilization from the oceans. PCB153 is more persistent and less volatile than PCB52, which explains the more pronounced decrease in PCB52.

The model predicts a 55% lower mass in the 2090s compared to the 1990s in the atmosphere of the northern hemisphere for all compounds studied. Nevertheless, the mass of HCHs is predicted to increase by up to 38% owing to volatilization from the ground, while the mass of PCBs was predicted to decrease by 17–38%. Higher temperatures lead to increased atmospheric concentrations of these contaminants, although their degradation rates will also rise (Hansen et al., 2015). The dominant process and outcome depend on the physical-chemical properties of each contaminant. However, regulation of emissions has a larger impact on contaminant concentrations in the Arctic atmosphere than climate change-related processes (AMAP, 2014; Hansen et al., 2015).

Table 3.1 shows total mean concentrations in the uppermost ocean layer for the periods 1995–1999 and 2095–2099 and the ratio of change (RoC) for four locations. These data were used as input to the food web model. The RoC for γ -HCH varies between almost no change (RoC=1) to 1.11 for Liefdefjorden (11% increase in mean concentration). The RoC for PCB52 varies between 0.4 and 0.53 (50–60% reduction in mean concentrations). The results for PCB153 show a slightly higher reduction than for PCB52 (55–67%).

Table 3.1. Mean concentrations in the ocean of γ -HCH, PCB52 and PCB153 for four locations around Svalbard for the periods 1995–1999 and 2095–2099, and the ratio of change (RoC) between concentrations.

Location	γ -HCH, ng/L			PCB52, pg/L			PCB153, pg/L		
	1995–1999	2095–2099	RoC	1995–1999	2095–2099	RoC	1995–1999	2095–2099	RoC
Kongsfjorden	0.125	0.134	1.07	0.382	0.192	0.50	0.687	0.299	0.43
Liefdefjorden	0.098	0.109	1.11	0.375	0.200	0.53	0.771	0.350	0.45
Barents Sea-1	0.249	0.253	1.02	0.778	0.308	0.40	1.077	0.359	0.33
Barents Sea-2	0.241	0.248	1.03	0.653	0.261	0.40	0.838	0.283	0.34

3.2 Climate change versus seasonal changes – impact on persistent organic pollutants in the ecosystem

3.2.1 Projections for a future climate

The bioaccumulation model was run with the simulated temperature and contaminant concentrations in 2107 based on the decadal ratio of results from the climate meteorological data between 1990–1999 and 2090–2099. The year 2107 was chosen to model the conditions a century later than the empirical food web data from 2007. Any differences in these input data (temperature and exposure) are due to different climate regimes from the ECHAM5 model for the two decades. As both decades have the same initial modelled contaminant concentrations, the climate ratio was based on the mean of the monthly ratio for the last five years (permitting the model to run for five years to allow the climate differences to affect the contaminant signal).

As there are few studies available from which to make predictions of lipid content in a future climate, a 10% decrease was assumed on all 2007 lipid values, an approach also taken and discussed by Borgå et al. (2010). This was based on a study of polar fish, where a temperature increase resulted in decreased lipid content (Brodte et al., 2008).

Climate change is likely to result in species replacement in polar food webs as southern species expand their distributions northward (Kallenborn et al., 2012). However, as predictions of species replacement in a future climate are highly speculative, this has not been included in the food web model. Thus, the climate change scenario includes seasonal changes in temperature, contaminant exposure and lipid content, but not changes in dietary composition.

3.2.2 Bioaccumulation model endpoints

From the different forcing scenarios, the treatments of contaminant exposure, temperature, lipid content and feeding relations were in

turn kept constant, varied one at a time, and then all varied in the full dynamic model. This resulted in accumulated concentrations in specific species as model endpoints. From lipid-normalized contaminant concentrations and estimated trophic positions, food-web biomagnification was assessed by trophic magnification factors (TMFs). TMFs were estimated from the regression slope resulting from the regression of contaminant concentrations against the trophic level of the specific species, as described in previous publications (Fisk et al., 2001; Borgå et al., 2004, 2012). The trophic levels were the Monte Carlo Markov Chain (MCMC) modeled ones included in the bioaccumulation model.

3.3 Kongsfjorden case study

Kongsfjorden is situated on the west coast of Svalbard close to Ny-Ålesund (Fig. 2.2, 78°55'N, 11°55'E). The research station Ny-Ålesund and the Zeppelin mountain atmospheric monitoring and research station are located at the coast in the middle part of the fjord. The fjord used to be ice-covered in winter, but was ice free in 2007 and thereafter has only had partial ice cover in the inner parts of the fjord during winter. There has also been a higher inflow of Atlantic water masses into the fjord (Hop et al., 2006; Cottier et al., 2007). Kongsfjorden is thus considered an Arctic fjord where the environmental parameters are currently changing towards Atlantic features, which makes the fjord very suitable for climate change studies in the Arctic.

The effect of the annual cycle in exposure, temperature, lipid content and food-web structure on γ -HCH, PCB52 and PCB153 biomagnification in the marine food web (phytoplankton, zooplankton, and fish) of Kongsfjorden was simulated using the modified AQUAWEB bioaccumulation model. Resulting TMFs were compared to measured data in the same food web from 2007. The DEHM model estimated temperature and contaminant concentrations in seawater in 2007 using real meteorological input data, and projected results for 2090–2099 using climate meteorological data. The best-fit scenario between simulated and measured food-web biomagnification allowed lipids to vary (Fig. 3.8). Estimated seasonal variations

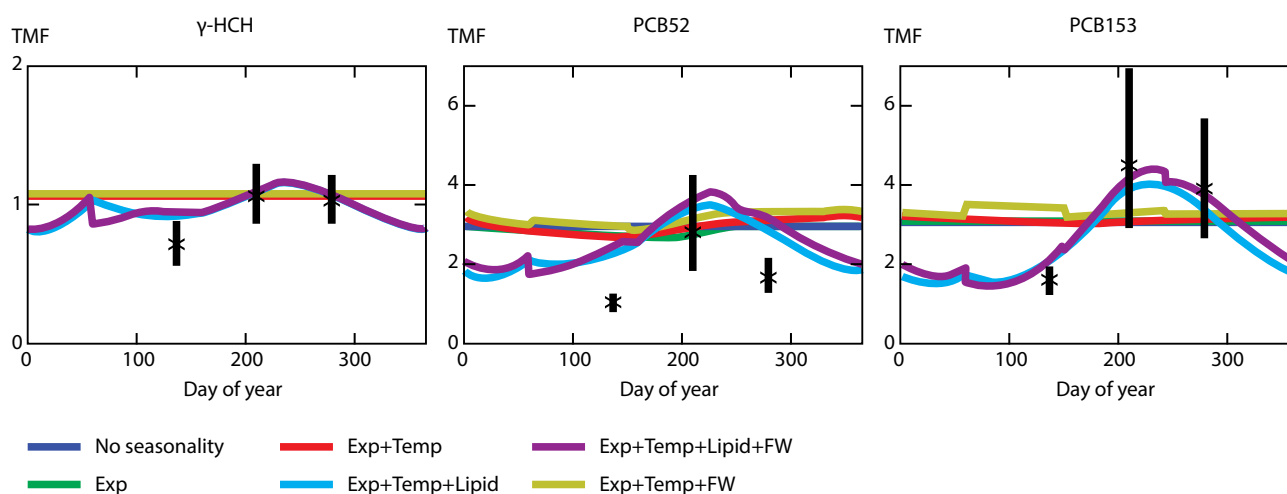


Figure 3.8. Modeled and measured food-web biomagnification (trophic magnification factor, TMF). Measured TMFs are from Hallanger et al. (2011). The scenarios reflect increasing degrees of dynamic parameterization; from no seasonal variation in any input parameters to varying seawater concentrations of POPs (exposure, Exp), adding temperature (Temp), lipid content (Lipid) and trophic interactions in the food web (FW).

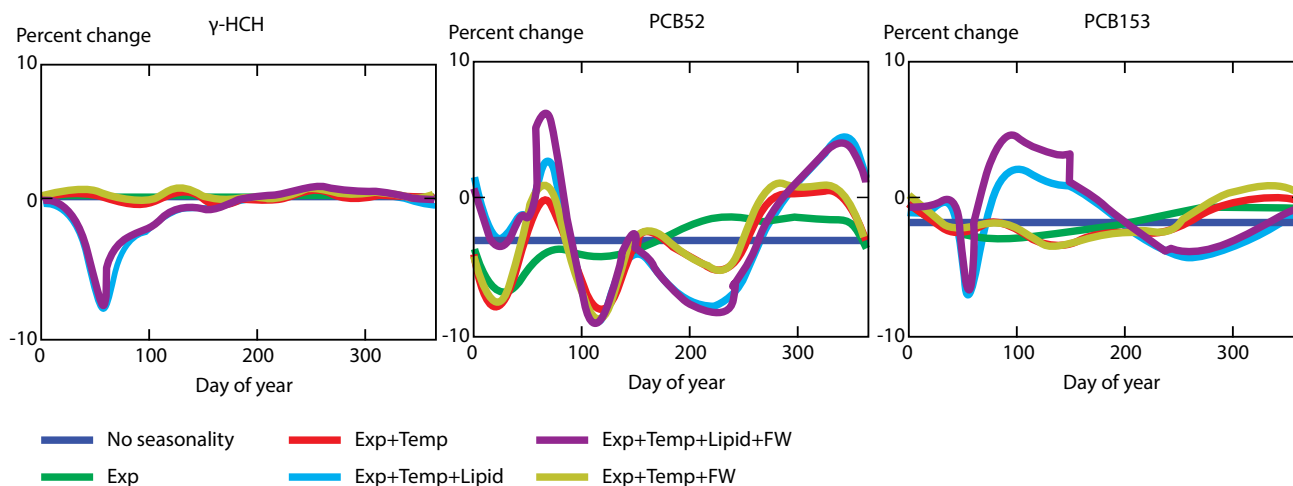


Figure 3.9. Percentage change in trophic magnification factors (TMF) between climate scenarios for 2007 and 2107. The scenarios reflect increasing degrees of dynamic parameterization, from no seasonal variation in any input parameters, to varying seawater concentrations of POPs (exposure, Exp), adding temperature (Temp), lipid content (Lipid) and trophic interactions in the food web (FW).

in temperature, exposure and dietary relations had a minor effect on TMF. In the future Arctic climate scenario, simulated TMFs were only marginally different from at present, and the magnitude and direction depended on the model substance; the average change from 2007 to 2107 was 0.4% for γ -HCH, -3.1% for PCB52 and -1.7% for PCB153 (Fig. 3.9). This indicates that seasonal variation in biomagnification is greater than the biomagnification response to projected climate change. As the best-fit seasonal scenario included the annual cycle in lipid content, this physiological factor is also crucial for making good predictions of contaminant food-web bioaccumulation in the future. Dietary composition was not a significant variable affecting TMF, mostly due to the lack of estimated diet composition between seasons. Seasonal changes in the abiotic drivers included alone were not enough to explain measured food-web bioaccumulation over time.

The present study analyzed biomagnification of contaminants in a marine food web. Previous studies have shown that the IPCC climate change projections of increased primary production would result in overall reduced food web contaminant exposure, because organic contaminant concentrations in the dissolved phase will decline as more are adsorbed to the organic carbon (Borgå et al., 2010).

3.4 Local sources of organic pollutants – A case study of Svalbard settlements

Svalbard has few permanent settlements and Longyearbyen is the main city (about 2000 inhabitants), followed by the Russian coal mining town Barentsburg (about 400 inhabitants). Both towns have coal-fired power plants as the main energy source. Svea is a coal mining settlement with no permanent population, but 100 to 200 people work there at any one time. The power plant in Svea is fueled by diesel. Detailed maps of these settlements are shown in Fig. 3.10. Hydrocarbons

have been detected at high concentrations in Longyearbyen, attributed mainly to local snowmobile use (Reimann et al., 2009). Recent measurements of PAHs in the soil below the main snowmobile routes were dominated by phenanthrene, chrysene, fluoranthene and benzo[*b*]fluoranthene. These PAHs were also present in the air in Longyearbyen, although they were not the dominant compounds. It is therefore assumed that the pattern differences between PAHs in air and soil are caused by selective sorption as well as by long-term weathering processes (Schütze, 2013). The present study therefore investigated whether particles could be characterized by their origin and be used as indicators of the origin of PAHs associated with well-defined particle fractions.

Gaseous and particle emissions from Longyearbyen and Barentsburg were analyzed for PAHs and PCBs. Particles were divided into two size fractions, with an aerodynamic diameter of 0.1–0.5 μm (fine particles) and 0.5–10 μm (coarse particles), respectively. The particles were analyzed with transmission electron microscopy (TEM) and scanning electron microscopy (SEM) and classified based on size, morphology and chemical composition (Table 3.2). The fine particles were dominated by secondary aerosols (56–79%) and secondary aerosols and soot (19–42%) in all samples from Longyearbyen and Barentsburg, while the coarse fraction was dominated by fly ash (power plant samples) and sea salt (Weinbruch et al., 2015).

The soot particles from the Longyearbyen power plant contained heterogeneous inclusions of Pb. Potassium (K) was also frequently detected in these particles. Soot particles collected in Longyearbyen (at the roof of UNIS) contained Pb and iron as heterogeneous inclusions. The exhaust from the power plant comprised fly ash particles (0.5–6 μm in aerodynamic diameter), where iron-rich aluminum silicates dominated the samples (84%), followed by aluminum silicates (8%) and silicates (8%). Particles from Barentsburg showed a different composition. Almost all the secondary

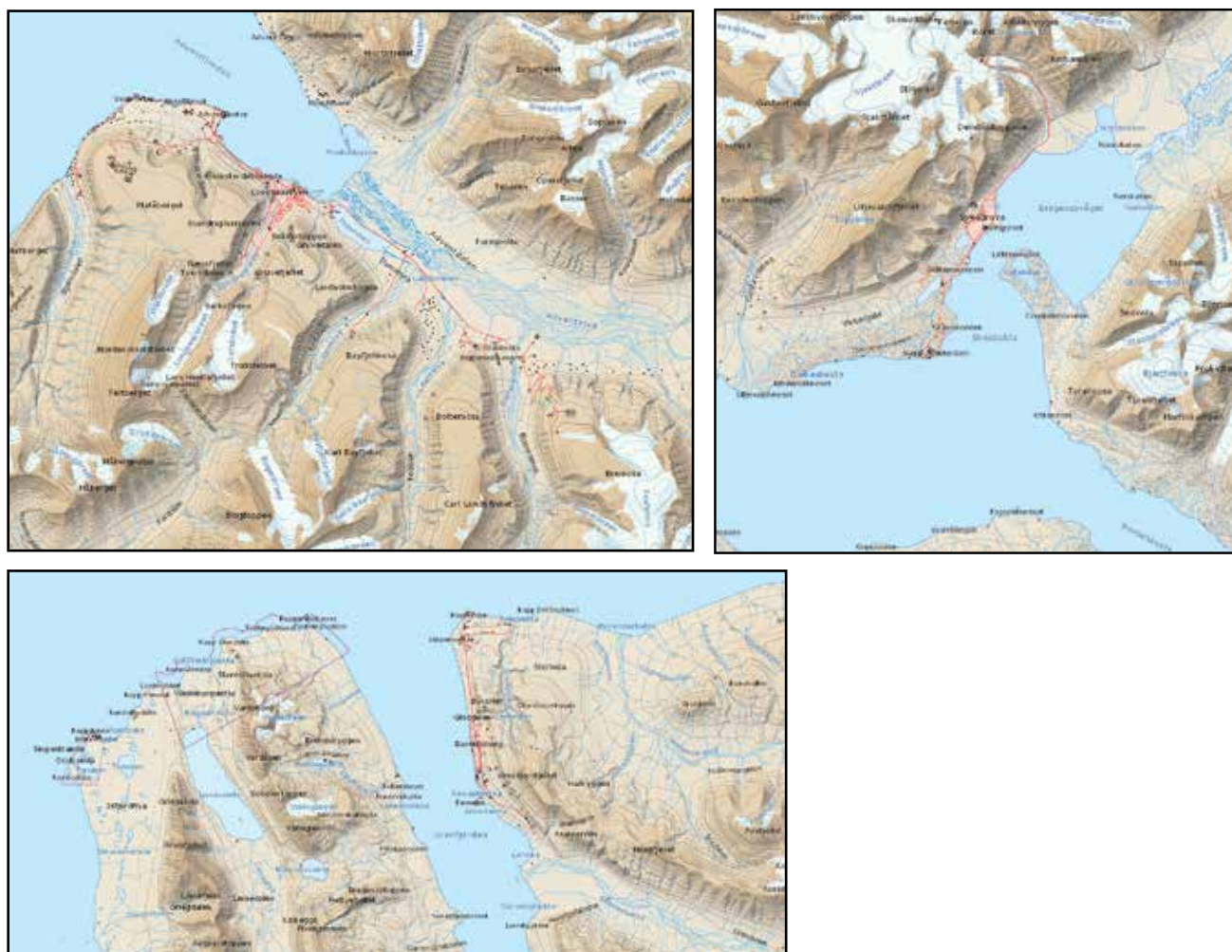


Figure 3.10. Longyearbyen (upper left), Svea (upper right) and Barentsburg, Isfjord radio and Linnevatnet (lower picture). Source: Norwegian Polar Institute (www.toposvalbard.no).

Table 3.2. Classification criteria for particle groups found in Longyearbyen and Barentsburg (Schütze, 2013).

Particle group	Classification criteria
Sea salt	Na and chloride (Cl) as major elements; sometimes magnesium (Mg), potassium (K), Ca, typical cubic morphology
Aged sea salt	As sea salt with sulfur (S) and/or oxygen (O)
Calcium (Ca) sulfates	Ca, S and O as major elements
Sodium (Na) sulfates	Na, S and O as major elements
Silicates	Silicon (Si) or Si and aluminum (Al) as major elements; Mg, K, Ca, often as minor elements, sometimes iron (Fe)
Mixed particles	Mixtures of sea salt, silicates and secondary aerosols
Soot	Carbon (C) and typical morphology of agglomerates of primary particles
Fly ash	Spherical morphology; aluminum silicate or metal (Fe sometimes titanium, Ti) composition
Iron oxides/hydroxides	Fe and O, sharp edges
Secondary aerosols	S, O, Na and/or C as major elements; sometimes nitrogen (N) and phosphorus (P); unstable under electron bombardment
Secondary aerosols and soot	As secondary aerosol with soot inclusions
Carbon rich	C as major element
Others	Particles not included in one of the groups listed above

aerosols contained high amounts of phosphorus, which can be considered as a fingerprint for the coal power plant in Barentsburg. As with soot particles from the Longyearbyen power plant, fly ash from the Barentsburg power plant also contained K and heterogeneous Pb inclusions. On the other hand, iron oxides, carbon-rich particles and particles with a cover of carbonaceous material were only found in the stack from the Barentsburg power plant. Samples 1 km north of the power plant contained fly ash particles dominated by iron-rich aluminum silicates (60%), aluminum silicates (24%) and iron oxides (16%).

Fly ashes are typical particles associated with coal burning. Secondary aerosols can originate from combustion in the local power plants, but can also reach Svalbard via long-range transport. TEM analysis indicated small heterogeneous Pb inclusions in soot internally mixed with secondary aerosols, which could be an inorganic fingerprint of local coal power plant combustion. McConnell and Edwards (2008) showed that Pb in particles originates from fossil-fuel combustion. Hence, these secondary aerosols and soot particles with heterogeneous Pb inclusions can be considered as fingerprints for coal combustion.

Polycyclic aromatic hydrocarbons most often originate from combustion processes, and the total (gas and particle-bound) atmospheric concentrations of PAHs in Longyearbyen declined with distance from the power plant (Figs. 3.10 and 3.11) during measurements in March 2012. However, concentrations of volatile two-ring PAHs were higher in Longyearbyen city and its vicinity than at the power plant. Two-ring PAHs are more likely to originate from sources other than coal combustion, such as traffic (cars and snowmobiles). The larger four- to six-ring PAHs dominated the particle phase at the power plant and in samples within Longyearbyen town, less than 1 km from the power plant. The dominant PAHs in Longyearbyen power plant were: indeno[1,2,3]pyrene, benzo[ghi]fluoranthene, benzo[bjk]fluoranthene, benzo[a]fluoranthene, benzo[e]pyrene and

benzo[a]pyrene. Despite the short distance to Longyearbyen, this PAH concentration pattern was not completely reflected in Longyearbyen town. This rapid change illustrates the quick composition and property changes occurring in primary emissions immediately after leaving the stack. These changes ultimately affect the PAH concentration pattern and are mainly caused by rapid cooling and condensation processes and particle deposition.

3.5 Perfluorinated alkylated substances in the terrestrial and freshwater environment

Perfluorinated alkylated substances are present in the Arctic environment mainly due to long-range atmospheric and oceanic transport (Stemmler and Lammel, 2010; Norwegian Environment Agency, 2014). Local settlements also contribute to PFAS levels; Longyearbyen on Svalbard (Kwok et al., 2013) and other Arctic locations. The importance of local contributions from, for example, airports has also been reported for Arctic Canada, where nearby lakes had different PFAS isomer patterns and concentrations. The dominance of perfluorooctane sulfonic acid (PFOS), followed by perfluorohexane sulfate (PFHxS), perfluoroheptane sulfate (PFHpA) and perfluorooctanoic acid (PFOA) in Resolute and Meretta Lakes that received wastewater from nearby airports indicates the impact of local sources. Nearby lakes that had not received such wastewater had much lower concentrations of PFASs, and different isomers dominated (PFOA and perfluorodecanoic acid, PFDA, in Lake Amituk and Char Lake, respectively) (Stock et al., 2007; NCP, 2013). Increased interest in northern areas (e.g., shipping, tourism, rising population in northern areas of Scandinavia) means the effects of local pollution must be considered. Concentrations and patterns of contaminants in local fish provide information about current pollution status.

The remote Lake Linnévatnet on Svalbard (Fig. 3.10) receives water from one small glacier and is situated 10 km east of Barentsburg and 45 km east of Longyearbyen, the two main settlements on Svalbard, with about 400 and 2000 inhabitants, respectively. Muscle (n=19) from Arctic char (*Salvelinus alpinus*) from Lake Linnévatnet had low concentrations of PFASs, and the sum of 13 PFAS isomers analyzed was 0.5 ng/g ww in 2010. Only three of those isomers were present above detection limits in more than 50% of samples: 6:2 fluorotelemer sulfonic acid (6:2 FTSA), perfluoroundecanoic acid (PFUnA), and perfluorononanoic acid (PFNA). Fish were also collected in 2013. PFUnA was the only PFAS isomer detected in most samples; more than 50% of Arctic char muscle samples contained PFUnA (Garsjø, 2013). Elevated PFBA levels were found in surface freshwater from Lake Linnévatnet collected in 2014–2015. PFBA was the predominant PFAS in freshwater and surface seawater from this location (Ahrens et al., in prep; Skaar, 2016). PFOS was detected in one muscle sample from 2013, and in five of 20 muscle samples from 2010, with a median concentration of 0.09 ng/g ww. Low levels of PFOS in lake trout (*S. namaycush*) have been reported from Great

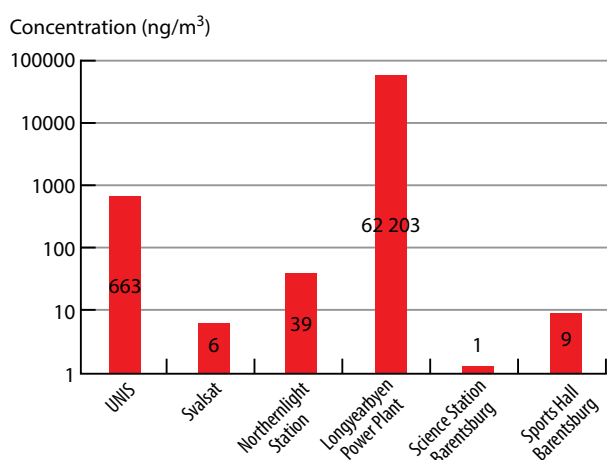


Figure 3.11. Total PAH concentration in the particulate and gas phase. Source: Schütze (2013).

Slave Lake in Canada (<0.01–0.58 ng/g ww in measurements from 2003 to 2009); these levels are comparable to PFOS in Arctic char from Linnévatnet, Svalbard (NCP, 2013; Garsjø, 2013). PFOA showed lower levels than expected: 0.05 ng/g ww in muscle from Arctic char collected in 2010 and ranging from 0.02 to 0.04 ng/g ww in 2013. However, PFOA was only detected in 20–25% of the Arctic char muscle samples. The median concentrations of the sum of perfluoroalkylated carbon acids (Σ PFCA) in 2013 (0.1 ng/g ww) and 2010 (0.3 ng/g ww) were comparable to those in sea-run char from Cambridge Bay and Pond Inlet, Canada (0.12 ng/g ww) (NCP, 2013; Garsjø, 2013). The size of the fish analyzed can affect concentrations, because large individuals often spend parts of their life in the ocean. To follow up on this, a new study was initiated to investigate PFAS levels in the water and sediment of Lake Linnévatnet.

Fish from Lake Vättern in the urban area of Sweden contained tenfold higher concentrations than those from Arctic Canada and Svalbard. Salmon (*Salmo salar*) and brown trout (*S. trutta*) from Lake Vättern contained 8.49 and 5.73 ng/g ww PFOS, respectively (Berger et al., 2009). The distance to sources is important for PFAS concentrations in fish, and this should be kept in mind with regard to the expected increase in population in Arctic regions.

Samples of liver, muscle and fatty tissue from Svalbard reindeer (*Rangifer tarandus platyrhynchus*) were provided by local hunters in 2010. While short-chained PFASs were common in muscle from Arctic char in 2010, the reindeer samples were dominated by long-chained PFASs (perfluorotridecanoic acid, PFTrA; perfluorododecanoic acid, PFDoA) in the muscle samples (Fig. 3.12). The distribution differs between the tissues analyzed, and PFNA, PFTrA, linear PFOS and PFHxS were the most common isomers in liver. PFOS concentrations in reindeer liver ranged from below the limit of detection (LOD) to 0.7 ng/g ww. These levels were comparable to those in moose (*Alces alces*) liver from the Decho area in Canada (<0.01–1.1 ng/g ww), but slightly lower than in caribou (*Rangifer tarandus*;

0.7–2.2 ng/g ww) from Northwest Territories/western Nunavut, Canada (Müller et al., 2011; NCP, 2013). A long-term study of reindeer and moose in Sweden from 1987 to 2006 reported perfluorooctane sulfonamide (FOSA), PFOS and PFOA above detection limits in reindeer muscle, but not at concentrations that could be quantified. PBDEs, hexabromocyclododecane (HBCDD) and even PCB28, PCB52 and PCB101 were below detection limits in moose and reindeer muscle in the same study (Danielsson et al., 2008).

The samples from Svalbard reindeer were also analyzed for hexachlorobenzene (HCB) and *p,p'*-DDE. The analyses confirmed the presence of *p,p'*-DDE in one liver (n=3) and one fatty tissue sample (n=7). HCB was confirmed in most samples: in one liver, four of the six muscle samples and in six of the fatty tissue samples. In addition to reindeer, local hunters also provided samples from one pink-footed goose (*Anser brachyrhynchus*) and one ptarmigan (*Lagopus muta hyperborea*) from Svalbard. Both HCB and *p,p'*-DDE were present in fatty tissue from the ptarmigan, but not in liver from the pink-footed goose. However, due to the very small number of samples, the results should be treated with caution (Melien, 2014).

Reindeer are popular food items in northern Scandinavia and recent studies have investigated POPs in semi-domesticated reindeer (*Rangifer tarandus tarandus*) from northern Norway. POPs concentrations in all tissues analyzed (meat, liver, fatty tissue, bone marrow) were low, often close to or even below detection limits. DDTs were detected in 5–45% of samples, with *p,p'*-DDE the most abundant in fatty tissue and bone marrow (33% and 45% of samples above the LOD, respectively). The levels of *p,p'*-DDE (range 0.49–3.01 ng/g ww in fatty tissue) were lower than for HCB (35.8 ng/g ww) (Hassan et al., 2013).

PFASs were analyzed in five glaucous gulls (*Larus hyperboreus*) and four black guillemots (*Cepphus grylle*) collected in 2007 from Longyearbyen. Glaucous gulls are known to have high concentrations of POPs as a result of their feeding behavior (Verreault et al., 2005), while black guillemots feed mainly on zooplankton and represent a lower trophic level than glaucous gulls. Glaucous gulls analyzed in the present study had mean Σ PFAS concentrations of 147 and 15 ng/g ww in liver and muscle, respectively, while levels in black guillemot were lower at 36 and 2.5 ng/g ww in liver and muscle, respectively (Axelson, 2014). PFOS contributed 72% of the Σ PFAS (126 ng/g ww in glaucous gull liver). A study from the Barents Sea in 2004 showed lower levels in liver from glaucous gulls (40 ng/g ww) but comparable levels for black guillemot (12 ng/g ww) (Haukås et al., 2007). The glaucous gulls analyzed in the present study were collected around the dumpsite in Longyearbyen, which can explain their high PFAS concentrations, in addition to a general trend of increasing PFAS levels in Arctic biota (Butt et al., 2010). Exposure to long-chain perfluorocarboxylic acids has now been found to be directly related to lower breeding success of Svalbard kittiwakes (*Rissa tridactyla*) (Tartu et al., 2014).

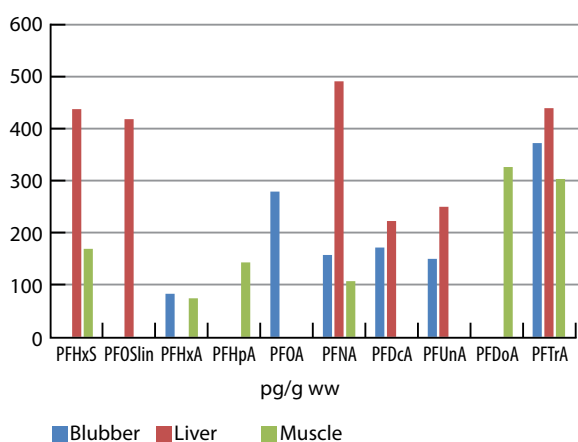


Figure 3.12. Concentrations of PFASs (pg/g ww) in Svalbard reindeer. Source: Svalbard Environment Fund (2013).

3.6 Persistent organic pollutants in benthic marine food from northern Norway

Halibut (*Hippoglossus hippoglossus*) and shrimp (*Pandalus borealis*) are common dietary foodstuffs for communities in northern Norway and are also important species for the coastal fishing industry. Halibut can grow large and old and this species has earlier been a cause for concern in some areas with regard to POPs exposure for consumers (Van der Meeren et al., 2014). PFASs, PBDEs, PCBs and DDTs were recently analyzed in shrimp and halibut from the Tromsø area in northern Norway (Carlsson et al., 2016). Median ΣPCB concentrations were 4.9 and 2.5 ng/g ww for halibut and unpeeled shrimp, respectively. PCBs contributed 50% of the total POPs load in the halibut followed by DDTs (26%) and PFASs (18%), while POPs in unpeeled shrimp were dominated by PFASs (74%). The higher levels of PBDEs, PCBs and DDTs in halibut compared to shrimp are indicative of the biomagnification of these compounds, reflecting the higher trophic level of halibut compared to shrimp. However, PFASs were present at higher concentrations in shrimp compared to halibut, with median PFOS concentrations of 2.7 ng/g ww in shrimp compared to 0.9 ng/g ww in halibut muscle. This emphasizes that emerging POPs and other organic contaminants require new methodology and insight in order to predict their potential exposure risk for humans.

Concentrations of legacy POPs and PFASs measured in shrimp and halibut are comparable to results from other studies conducted elsewhere in the Arctic. PFASs have been reported at low levels and even below detection limits in fish from the Faroe Islands, Iceland and western Greenland (Jörundsdóttir et al., 2012; Eriksson et al., 2013; Carlsson et al., 2014a). Thus, a major town like Tromsø, with associated port facilities and shipping activities, may not contribute significantly to the POPs burden observed in these species. This is perhaps less clear for PFASs, as levels of PFOS and PFCAs were higher in shrimp than halibut muscle. The overall concentrations of POPs, including the dioxin-like PCBs, as well as PFASs were well below the European guidelines for human consumption in shrimp and halibut, and human dietary exposure through moderate consumption of these organisms falls within tolerable daily intake (TDI) levels or benchmark doses. Muscle from larger and older halibut may contain higher POPs concentrations, although these are (mostly) not a cause for concern with regard to human consumption.

3.7 Human exposure to dietary contaminants in northern Norway and Russia

3.7.1 Northern Norway

Concentrations of a variety of POPs were measured in women from the Northern Norway Mother-and-Child Contaminant Cohort study (MISA study). Low contaminant concentrations were generally observed in these women (Table 3.3). Data are

presented only in cases where detection frequencies were greater than or equal to 70%. Certain POPs were not reported because the coefficient of variation exceeded 70% and concentrations were too low. The POPs not reported were oxychlorodane, *trans*-chlorodane, *cis*-chlorodane, heptachlor, Mirex, and PCB28, PCB52, PCB101, PCB105, PCB149, PCB156, and PCB194.

When comparing the northern Norway MISA data for 2006–2008 with previously reported data (Table 3.4), the recent geometric mean of *p,p'*-DDE concentrations in maternal plasma is almost half the value for Bodø in 2004, although the range is wider (AMAP, 1998, 2003, 2009). The geometric mean for PCB153 concentrations in the plasma of northern Norway MISA participants is slightly higher than reported for participants from Bodø. Although with a wider range, the geometric means for participants from both Bodø and northern Norway showed levels that were almost half those observed in 1996 in the plasma of women from Kirkenes (AMAP, 2009).

Table 3.3. Lipid-adjusted concentrations (µg/kg plasma lipid) of PCBs and organochlorines in pregnant women (early pregnancy) from northern Norway (n=515; mean age=30.6; age range 18–43; average parity=0.9; range 0–4) (Hansen et al., 2010).

Contaminant ^a	2006–2008 Geometric mean	Range
<i>p,p'</i> -DDE	38.7	10.9–351
PCB99	2.2	0.4–18.1
PCB118	4.1	1.0–38.3
PCB138	14.9	2.8–118
PCB153	24.8	5.3–201
PCB180	16.5	3.0–159
HCB	9.6	3.5–53.3
<i>trans</i> -nonachlor	2.8	0.6–17.6
<i>cis</i> -nonachlor	0.6	0.1–4.5

^aAverage values are presented only for compounds with detection frequencies greater than or equal to 70%, all levels below the Limit of Detection (LOD) were set to LOD/√2 for statistical purposes.

Table 3.4. Time trends of contaminants in maternal plasma from participants from northern Norway (µg/kg plasma lipid). Data show geometric mean (and range) for specified sampling periods. Oxychlorodane data were not available for northern Norway due to low concentrations and the coefficient of variation exceeding 70%. Sources: Kirkenes (AMAP, 1998), Bodø (SPFO, 2005), northern Norway (Hansen et al., 2010; AMAP, 2015).

Contaminant	Kirkenes 1996 29 (19–44) ^a n=66 ^b	Bodø 2004 30 (20–35) ^a n=10 ^b	N. Norway 2006–2008 30.6 (18–43) ^a n=515 ^b
Oxychlorodane	3.9 (1.6–11)	2.6 (1.0–6.1)	
<i>p,p'</i> -DDE	79 (19–436)	67 (26–176)	38.7 (10.9–351)
PCB153	52 (25–130)	23 (10–50)	24.8 (5.3–201)

^aArithmetic mean age and range of group sampled; ^bnumber of individuals sampled.

Table 3.5. Serum concentrations of PFASs (µg/L ww) in the northern Norway MISA study group (n=378; mean age 31; range 18–43; average parity = 0.9; range 0–4). Data are presented for PFASs with detection frequencies greater than 50% (Berg et al., 2014).

	Median	Arithmetic mean	Range
PFHxS	0.44	0.61	<LOD–14.8
PFHpS	0.10	0.12	<LOD–1.10
ΣPFOS	8.03	8.81	0.30–35.8
PFOS Linear	4.66	5.10	<LOD–19.1
PFOS Branched	3.37	3.71	<LOD–18.2
% linear PFOS	59.0	59.1	36.0–80.0
PFOA	1.53	1.70	0.28–11.0
PFNA	0.56	0.67	0.15–4.36
PFDA	0.23	0.26	0.05–2.34
PFUnDA	0.26	0.30	0.03–1.46
PFDoDA	0.03	0.04	<LOD–0.20

LOD, limit of detection; PFHxS, perfluorohexane sulfonate; PFHpS, perfluoroheptane sulfonate; ΣPFOS, sum of branched and linear perfluorooctane sulfonate; % linear PFOS, percentage linear PFOS related to PFOS; PFOA, perfluorooctanoate; PFNA, perfluorononanoate; PFDA, perfluorodecanoate; PFUnDA, perfluoroundecanoate; PFDoDA, perfluorododecanoate.

The MISA study identified links between traditional and market foods consumed and PFAS concentrations measured in serum. Levels of PFOS were greater than those of any other PFASs in the MISA study women, followed by PFOA (Table 3.5). Berg et al. (2014) showed that women consuming more marine food had significantly elevated concentrations of PFOS and other PFASs. Women who consumed a large amount of game had higher concentrations of PFHxS, perfluoroheptane sulfonate (PFHpS), and PFNA, while elevated concentrations of PFHpS and PFOS were also detected in high consumers of white meat. There was also a relationship between the consumption of beef and salty snacks and higher PFOA concentrations. However, the strongest significant predictor of all PFASs investigated was parity (Table 3.5), indicating lower maternal levels with increasing parity. Moreover, higher total months of breastfeeding were significantly associated with lower serum concentrations of PFHpS, PFOS and PFOA, across parity groups. While no association with age was found for PFHxS, PFHpS, PFOS or PFOA concentrations, age was positively associated with PFNA, PFDA and perfluoroundecanoate (PFUnDA) concentrations. These observations may be explained by compound differences in half-lives, bioaccumulation potential and continued production after 2002 for PFNA, PFDA and PFUnDA (Armitage et al., 2009; Zhang et al., 2012).

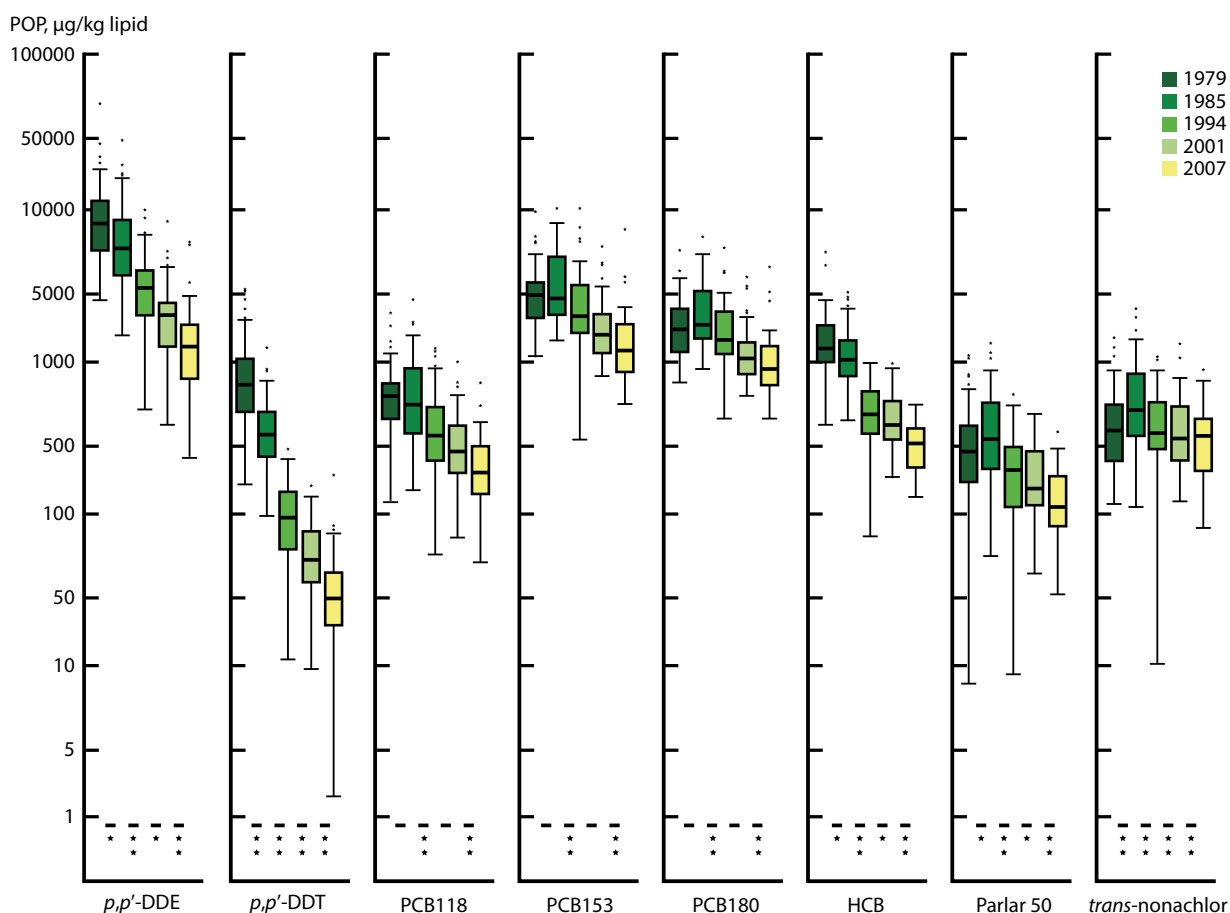


Figure 3.13. Concentrations (log scale) of selected POPs analyzed in repeat serum samples of men (1979 n=51, 1985 n=51, 1994 n=45, 2001 n=48, 2007 n=52) from northern Norway. Parlar 50 represents toxaphenes and *trans*-nonachlor represents the chlordanes. Boxes represent the 25th to 75th percentiles, horizontal lines represent the median, whiskers indicate 1.5 times the length of the interquartile range above and below the 75th and 25th percentiles, respectively, and outliers are represented as data points. The asterisks denote significant differences (* $p < 0.05$ and ** $p < 0.001$) for comparisons between consecutive sampling years. Source: Nøst et al. (2013).

In the Tromsø Study, a long-term investigation of health in men from northern Norway, a number of contaminants were analyzed in repeat serum samples for a subset of these men ($n=53, 52, 48, 49$ and 52 for samples taken in 1979, 1986, 1994, 2001 and 2007, respectively). Nøst et al. (2013) found that levels of several POPs declined in these men between 1979 and 2007–2008 (identified as 2007), with significant declines observed for most POPs between pairs of five different consecutive sampling years (Fig. 3.13). The largest decrease was observed for p,p' -DDT, which declined tenfold between

1979 and 2007. DDTs and HCB showed significant declines over the entire time frame. Parlar 50 (representing toxaphenes) and *trans*-nonachlor (representing the chlordanes) also showed significant differences across all consecutive sampling years. The smallest absolute decline overall was for chlordanes. Many pesticides were observed to decline from 1979 onward. Each of PCB118, PCB153, and PCB180 increased from 1979 to 1986 but otherwise showed similar blood concentration patterns across 1979 and 2007, with two significant periods of decline, between 1986 and 1994, and 2001 and 2007.

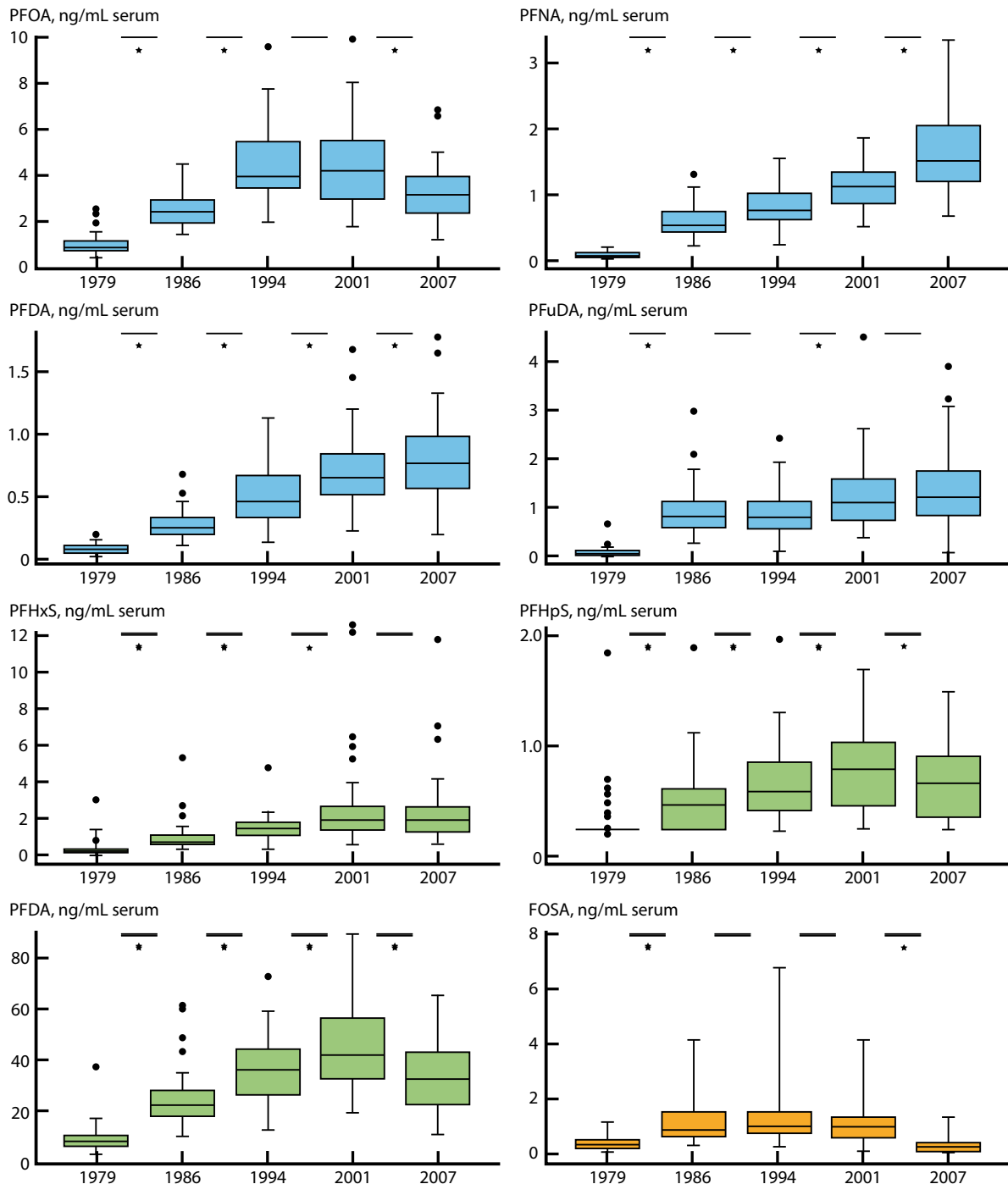


Figure 3.14. Concentrations of the most abundant PFASs analyzed in repeat serum samples for men from northern Norway (1979 $n=53$, 1985 $n=52$, 1994 $n=48$, 2001 $n=49$, 2007 $n=52$). The asterisks (*) denote significant differences in consecutive sampling years ($p < 0.001$, Wilcoxon signed rank test). The boxplots for FOSA are censored box plots with the horizontal line indicating the limit of detection (LOD). Source: Nøst et al. (2014).

A similar analysis was performed by Nøst et al. (2014) for PFAS concentrations in the same cohort (the Tromsø Study) of Norwegian men (Fig. 3.14). The highest measured concentrations were again for PFOS, in 2001; however, the largest percentage change in concentration was for PFUnDA (850%) between 1979 and 1986. Three PFASs (PFDA, PFNA, PFOS) showed significant changes in concentration across all consecutive paired years; in all cases levels were increasing. For PFOS in men, concentrations increased fivefold from 1979 to 2001 and subsequently declined between 2001 and 2007. PFOA and FOSA showed similar overall trends with significant declines between 2001 and 2007.

3.7.2 Arctic Russia

This section presents a follow-up to the study conducted in Russia involving the Chukotka 2001–2003 birth cohort which was reported by AMAP (2009) and the 2007 follow-up involving biomonitoring sampling and investigation of health effects. Individual data on contaminant levels in blood samples from 17 mothers and cord blood from their corresponding 17 babies born in the Chukotka coastal area in 2001 and 2002 were compared with levels in blood sampled from the same women and their five-year old children in 2007 (Dudarev et al., 2010). The possible influence of breastfeeding on maternal POPs serum levels and association of children's POPs blood levels and the frequency of infectious diseases was assessed. Data from sampling for contaminants in women and men from three northwestern Russian communities in 2009 and early 2010, namely, Nelmin-Nos, Usinsk and Izhma reported by Rylander et al. (2011), are also presented here. The three study sites are separated geographically and the residents are anticipated to have different dietary habits and living conditions. Blood samples were collected from volunteers at the same time as they took part in a general health examination. The population of Nelmin-Nos was also

sampled during the 2001 to 2003 Persistent Toxic Substances study, of which the Chukotka 2001–2003 birth cohort was a part. In association with the international Kolarctic project (KO467), samples were taken in 2013 and 2014 in the Pechenga district of Murmansk Oblast in northwestern Russia, near the Norwegian border. In the two cities Nickel and Zapolyarny, 400 people were interviewed and for the first time in this region, blood samples were taken to assess concentrations of contaminants. The sampled population included adult men, and pregnant and non-pregnant women (AMAP, 2015). Although the three study sites were geographically separated and the residents were expected to have different dietary habits and living conditions, few regional differences were observed when the data were controlled for age and sex. However, men had significantly higher concentrations of HCB in Izhma, a rural inland district of the Komi Republic mainly populated by reindeer herders, and the Izhma study group overall showed higher concentrations of Cd. The study group from Usinsk, where most people make their living as oil and gas workers, had higher concentrations of selenium and *p,p'*-DDE (women only).

The 2001–2003 Chukotka birth cohort has previously been described (AMAP, 2009). Maternal blood levels of all POPs decreased, by 33–74%, during the five-year interim period between 2001–2003 and 2007. Over the same period, the child blood serum concentrations of most POPs increased, by 24–132%. The increase in POPs levels in child blood may be explained by the transfer of contaminants from the mother through breastfeeding as well as through the consumption of local traditional food. Only oxychlordan and *p,p'*-DDT decreased in children from birth to 2007. After five years, maternal PCB levels were approaching those observed in the cord blood in 2001–2003. Conversely, child PCB levels after five years now resemble maternal PCB levels from 2001 (Table 3.6). The same PCB congeners were measured in mothers and children (Table 3.6).

Table 3.6. Maternal (n=17), cord and child blood (n=17) concentrations of POPs and metals, 2001–2002 and 2007, from coastal Chukotka (Lavrentia and Lorino, Russia). Data are reported as geometric means (range). POPs are reported as µg/L plasma. Metals are reported as µg/L whole blood (Dudarev et al., 2010).

Contaminant	Maternal		% ^a change	Cord	Child	% ^a change
	2001–2002 24.6 years (15–33)	2007				
Oxychlordan	0.9 (0.09–3.5)	0.2 (0.02–0.8)	-73	0.43 (0.06–3.6)	0.3 (0.06–1.0)	-27
<i>trans</i> -nonachlor	0.9 (0.1–2.9)	0.2 (0.05–1.0)	-72	0.3 (0.03–3.2)	0.4 (0.01–1.3)	+14
<i>p,p'</i> -DDT	0.2 (0.06–1.2)	0.06 (0.02–0.1)	-72	0.12 (0.02–0.4)	0.08 (0.03–0.3)	-33
<i>p,p'</i> -DDE	2.7 (1.0–6.6)	0.8 (0.2–2.5)	-70	1.2 (0.3–7.4)	1.5 (0.3–3.7)	+24
DDE:DDT	11.9 (5.5–76.8)	12.8 (4.0–37.3)	+8	10.6 (2.3–34.3)	19.5 (5.8–102.5)	+84
ΣPCBs	3.5 (0.8–8.9)	2.0 (0.3–6.6)	-44	1.4 (0.2–10.8)	3.3 (0.3–7.6)	+132
HCB	2.1 (0.5–6.0)	1.7 (0.4–10.7)	-19	1.3 (0.2–5.1)	2.2 (0.3–6.3)	+72
β-HCH	2.1 (0.8–7.6)	1.4 (0.3–2.9)	-33	1.2 (0.2–8.1)	2.3 (0.7–7.7)	+90
Mirex	0.1 (0.04–0.5)	0.08 (0.03–0.2)	-43	0.07 (0.01–0.5)	0.09 (0.04–0.2)	+28
Hg	1.6 (0.5–3.9)	1.61 (0.5–4.8)	+1	1.4 (0.5–3.3)	0.9 (0.5–2.7)	-31
Pb	37.5 (18.3–76.8)	29.6 (4.9–137)	-21	37.6 (14.3–78.3)	38.2 (6.9–102)	+2

^aGeometric mean values of 2007 relative to geometric mean values of 2001–2002 (100%).

In this follow-up study based on these measured maternal PCB levels, the calculated average elimination half-life of PCB congeners (congener group 105–187) in maternal blood was four to six years (for total PCB, 5.7 years) (Dudarev et al., 2010). As a comparison, Ritter et al. (2011) calculated maximum intrinsic human elimination half-lives for PCBs as ranging from 10 to 15 years, which are longer time frames than experienced for the Chukotka women in the present study. A shorter calculated half-life for the sum of PCBs may indicate that these women had an increased mechanism for elimination (e.g., additional pregnancies). While the duration of breastfeeding for the infant from 2001–2003 was not significantly correlated with maternal blood serum levels for POPs in 2007, it is known that almost 50% of women in the study had at least one additional child between their pregnancy in 2001–2003 and the 2007 follow-up, with two women having two additional children in this period. The additional pregnancies may have reduced maternal PCB body burdens during gestation and subsequent breastfeeding, although additional breastfeeding durations were not tested for significance. It is known that the major source of contaminant exposure in this maternal group was via the consumption of traditional food items (Dudarev et al., 2010). While important dietary changes may have occurred during extended or cumulative hospital stays for pregnant women

(Dudarev et al., 2010), it remains unclear whether such short-term dietary transitions would significantly reduce the body burden of long-lived contaminants such as PCBs in Russian women (Binnington et al., 2014).

In this study, maternal *p,p'*-DDT and *p,p'*-DDE levels declined by about 70% and 72%, respectively (Dudarev et al., 2010). Levels of *p,p'*-DDT decreased in children by 33% over the five-year period, potentially indicating that exposures to *p,p'*-DDT decreased during early childhood, or exhibited dilution through growth. Levels of *p,p'*-DDE increased by 24%, probably reflecting metabolism of the existing *p,p'*-DDT body burden in the children and potentially continued exposure to *p,p'*-DDE in food or another compartment. The average duration of breastfeeding of all babies from 2001 to 2007 was 24.4 months (range 1 to 72), so ongoing exposure through breastfeeding seems likely (Dudarev et al., 2010). The DDE:DDT ratio among mothers did not change, remaining around 12, which might indicate continued historical sources of exposure, whereas in children this ratio increased substantially over this five-year period, by 84% (from 10.6 to 19.5).

Statistical analysis of arithmetic mean concentrations of PCBs, HCB, and *p,p'*-DDE in women (Table 3.7) and men (Table 3.8) from Nelmin-Nos, Izhma, and Usinsk indicated that men had

Table 3.7. Concentrations of POPs ($\mu\text{g}/\text{kg}$ plasma lipid) in women in regions of Russia in 2009–2010. Data are presented as arithmetic mean [median] and (range) (Rylander et al., 2011).

	Nelmin-Nos (n=87) 2.8 (0–9)^a	Izhma (n=25) 2.3 (1–3)^a	Usinsk (n=25) 2.1 (1–4)^a
PCB99	31 [20] (<LOD–304)	16 [9.4] (<LOD–56)	25 [25] (<LOD–35)
PCB118	48 [30] (<LOD–268)	41 [35] (<LOD–66)	44 [40] (<LOD–106)
PCB138	46 [35] (<LOD–169)	32 [31] (<LOD–61)	40 [37] (<LOD–66)
PCB153	98 [78] (<LOD–534)	65 [59] (<LOD–156)	72 [67] (<LOD–137)
PCB180	57 [47] (<LOD–286)	23 [8.5] (<LOD–108)	30 [25] (<LOD–113)
HCB	135 [110] (<LOD–373)	122 [102] (32–297)	117 [103] (35–320)
<i>trans</i> -nonachlor	na	na	3.6 [1.3] (<LOD–21)
<i>p,p'</i> -DDE	246 [163] (<LOD–1342)	127 [107] (41–517)	234 [203] (91–600)

^amean parity (range); LOD: limit of detection.

Table 3.8. Concentrations of POPs ($\mu\text{g}/\text{kg}$ plasma lipid) in men from regions of Russia in 2009–2010. Data are presented as arithmetic mean [median] and (range) (Rylander et al., 2011).

	Nelmin-Nos (n=22)	Izhma (n=25)	Usinsk (n=25)
PCB99	36 [26] (<LOD–136)	29 [22] (<LOD–175)	33 [31] (<LOD–70)
PCB118	57 [26] (<LOD–532)	43 [15] (<LOD–478)	39 [28] (<LOD–106)
PCB138	58 [36] (<LOD–291)	54 [36] (<LOD–270)	53 [55] (<LOD–104)
PCB153	104 [92] (<LOD–222)	123 [106] (<LOD–297)	110 [97] (36–236)
PCB180	64 [57] (<LOD–152)	79 [73] (<LOD–170)	63 [59] (<LOD–158)
HCB	98 [86] (<LOD–203)	183 [160] (46–361)	122 [107] (53–427)
<i>trans</i> -nonachlor	na	na	8.9 [4.6] (3.1–26)
<i>p,p'</i> -DDE	245 [176] (51–732)	168 [138] (20–428)	228 [190] (53–782)

Table 3.9. Levels of POPs in pregnant women, non-pregnant women, men and the general population (men and non-pregnant women of all ages) in the Pechenga district of Murmansk Oblast in 2013–2014. Data are lipid adjusted ($\mu\text{g}/\text{kg}$ plasma lipid). Data, including age, are presented as geometric mean (range) (AMAP, 2015).

	Pregnant women	Women	Men	General population (men + women)
	n=50^a	n=33^a	n=17^a	n=50^a
	28.2 (16–41)^b	44.2 (26–65)^b	39.2 (27–54)^b	42.3 (26–65)^b
PCB118	26.1 (9.4–119)	35.6 (12.1–98.2)	46 (9.9–135)	38.6 (9.9–134)
PCB138	9.2 (0.97–48.2)	28 (6.4–74.9)	44.3 (14.8–106)	32.5 (6.4–106)
PCB153	12.2 (1.3–56.7)	27.5 (8.5–61)	47.9 (21.7–141)	32.8 (8.5–141)
<i>p,p'</i> -DDE	102 (16–1221)	141 (39.4–538)	167 (51.6–940)	150 (39.4–940)
<i>p,p'</i> -DDT	11.4 (1.3–376)	18.9 (6.5–124)	39.5 (11.6–132)	24.9 (6.5–132)
HCB	18.2 (5.3–252)	33.3 (12.8–85.1)	40.6 (10.9–189)	35.5 (10.9–189)
β -HCH	8.5 (0.8–146)	55.8 (17.6–157)	46 (27.8–100)	52.6 (17.6–157)

^aNumber of individuals sampled; ^bgeometric mean (range) of age of individuals sampled.

significantly higher concentrations of PCB138, PCB153, and PCB180 than women in the period 2009 and early 2010 (Rylander et al., 2011). Older people had higher concentrations of PCB153, PCB180, HCB and *p,p'*-DDE. When the blood data were adjusted for age and sex, no differences were found between the three communities for PCB138, PCB153 and PCB180, although men from Izhma had significantly higher HCB concentrations than men from the other sites. When adjusted for parity, the women of Usinsk had significantly higher levels of *p,p'*-DDE. Parity was only a significant predictor of *p,p'*-DDE (Rylander et al., 2011). POPs levels in pregnant women in the Pechenga district of Murmansk Oblast (Table 3.9) were lower than those in the general population (combined men and non-pregnant women). POPs levels in people from Murmansk were comparable to those in subjects from Nelmin-Nos, Izhma and Usinsk for PCB118, but PCB138, PCB153 and HCB levels were lower in the Murmansk population (AMAP, 2015).

In comparing changes in mean concentrations of POPs in the Nelmin-Nos populations of men and women between the 2001–2003 sampling and the more recent sampling in 2009–2010, there was a clear decrease for both women and men

in *p,p'*-DDE and, to a lesser extent, in HCB. Small increases in concentration were observed for PCB153 and PCB180 across the two sampling periods although the significance of these differences has not been assessed.

Table 3.9 shows recent levels of POPs in pregnant women, and the general population of non-pregnant women and men combined, of the Pechenga district of Murmansk Oblast (AMAP, 2015). Pregnant women's levels of POPs were consistently lower than for non-pregnant women and men in the general population. Women's levels of POPs were consistently lower than men's, with the exception of β -HCH, where women had higher levels, 55.8 $\mu\text{g}/\text{kg}$ plasma lipid compared to 46.0 $\mu\text{g}/\text{kg}$ plasma lipid, and a wider range of levels than men. The maximum concentration of β -HCH in women was 157.1 $\mu\text{g}/\text{kg}$ plasma lipid compared to men's 100.5 $\mu\text{g}/\text{kg}$ plasma lipid. Arithmetic means of PCB118 in the Pechenga district are comparable to those in Nelmin-Nos, Izhma and Usinsk. PCB138 levels in those regions are slightly elevated compared to men and women in the Pechenga district, while differences in PCB153 and HCB concentrations in these regions were even greater.

4. Heavy metals, with an emphasis on mercury

4.1 Atmospheric monitoring, trends and global change impact in Norway

A decrease in the concentration of atmospheric Hg has been observed at many remote monitoring stations in the northern hemisphere in response to the decline in anthropogenic Hg emissions in North America and Europe since the 1990s (Temme et al., 2007; Ebinghaus et al., 2011; Cole et al., 2013). However, until 2014 this decrease is not evident at Norwegian long-term monitoring stations. The lack of a decline in air concentrations in Norway may be due to re-emissions of Hg or because monitoring sites are influenced by air masses other than those in which decreases have been observed. Long-term monitoring of atmospheric elemental Hg concentrations has been conducted at two locations in Norway since the early 1990s. One site is located in the southernmost part of Norway (Birkenes at 58°N) and the other site in the far north at the Zeppelin station (Ny-Ålesund at 79°N) on the Arctic archipelago, Svalbard. A new station (Andøya 69°N) was added to the Norwegian monitoring program in 2010. Results from these stations show the annual mean Hg level to be similar to the northern hemisphere background concentration, ranging from 1.5 to 1.7 ng/m³ (Fig. 4.1). Observations from these stations show no significant overall temporal trend in concentration. Only if the last eight years from Birkenes are considered can a concentration decrease of about 2% per year be seen, which is similar to observations from other stations in Europe and North America, such as Mace Head (Ebinghaus et al., 2011) and Egbert (Cole et al., 2013).

While the relatively large uncertainties in global emission inventories prohibit the analysis of expected trends in air concentration at specific monitoring stations, models can be used to examine general concentration levels. Fig. 4.1 presents results from the DEHM model using global historical AMAP Hg inventories for 1990, 1995, 2000 and 2005 as input to the Hg module of the DEHM model (Christensen, 1997; Christensen et al., 2004; AMAP, 2010; Brandt et al., 2012). Modeled and measured GEM concentrations at Birkenes show good agreement overall, although the model results do not capture the sometimes large year-to-year variation in the measurements (Fig. 4.1).

Worldwide atmospheric Hg measurements to the early 2000s suggest that atmospheric Hg levels increased from the late 1970s to a peak in the 1980s, then decreased and leveled off from 1996 to 2001 (Slemr et al., 2003). At Mace Head (Ireland), air masses that have crossed the North Atlantic Ocean from North America showed a decrease in total gaseous Hg (TGM) of -0.028 ± 0.01 ng/m³/y for the period 1996 to 2009 (Ebinghaus et al., 2011). Long-term atmospheric monitoring data for GEM in polar regions are rare; although GEM has been measured at Alert (Canada) since 1995. Trend analysis showed a decline of -0.0086 ng/m³/y for the period 1995 to 2007 (Cole and Steffen, 2010). Trend analyses on the Hg data from Ny-Ålesund have been undertaken for several periods. For 1994–2002, 1995–2005 and 2000–2009, Berg et al. (2004, 2008, 2013) reported no trends in GEM concentration. The first two studies contained manually collected data for 1994 to 1999. Annual averages were analyzed in the first study, and in the second, annual, winter, spring, summer and autumn means were used. In the third study, Berg et al. (2013) evaluated results from ten years of GEM measurements

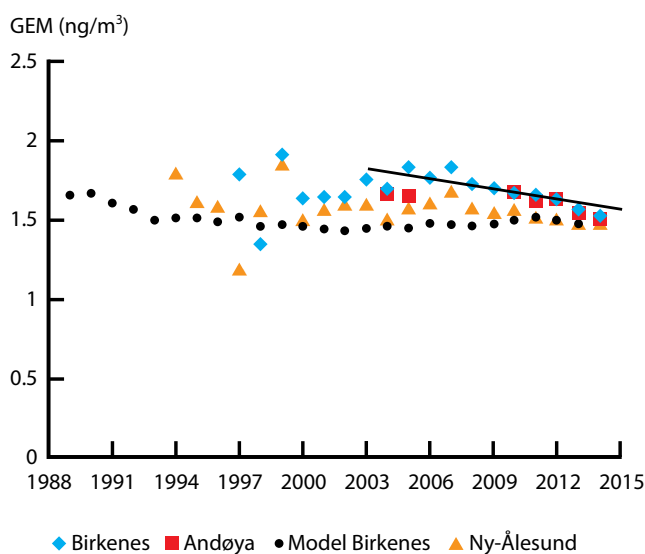


Figure 4.1. Annual mean GEM concentrations in Birkenes, Andøya and Ny-Ålesund. Source: NILU.

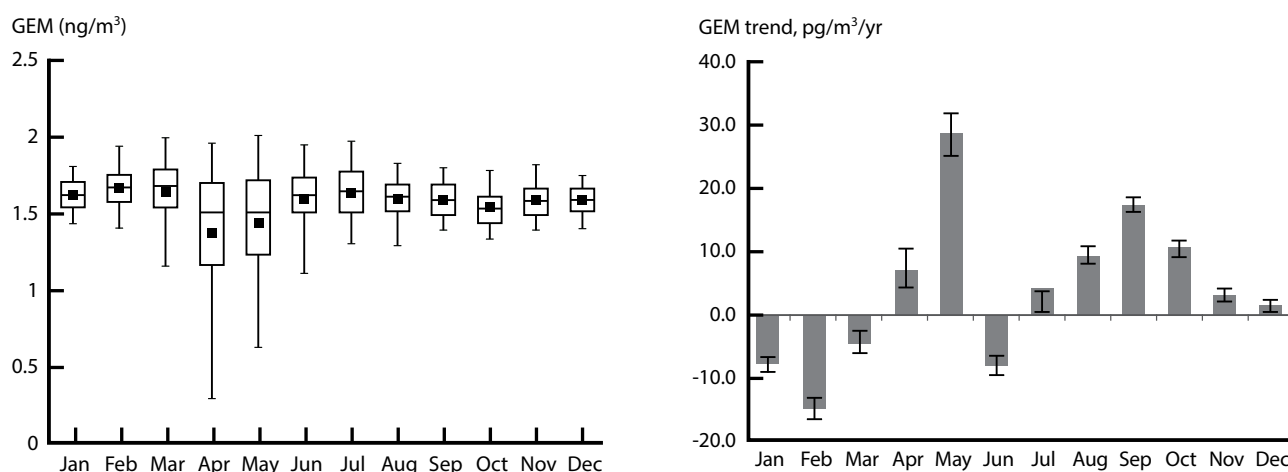


Figure 4.2. A) Box-and-whisker plot presenting the concentration distribution of GEM measurements at Zeppelin, Ny-Ålesund. The middle line in the box shows the median concentration, while the box indicates the range between the 25th and 75th percentiles. Whiskers above and below the box indicate the 90th and 10th percentiles, respectively. The filled square shows the arithmetic mean. B) Seasonal (monthly) trends in GEM based on daily average measurements from 2000 to 2009. Error bars represent 95% confidence limits. Source: Berg et al. (2013).

at the Zeppelin station (Fig. 4.2). Based on annual concentration changes calculated using the Sen's slope estimate (a statistical method for robust linear regression that chooses the median slope among all lines through pairs of two-dimensional sample points), no significant overall annual trend between 2000 and 2009 was found. The lack of an annual trend is in agreement with the earlier trend analysis for Hg at this station; however, it contrasts with what has been reported for other sampling sites (Cole and Steffen, 2010; Ebinghaus et al., 2011). Also, the seasonal Kendall test (a statistical method that accounts for seasonality by testing each period separately) was applied to the data and this showed significant trends in every month. The Sen's calculation of slope was performed to give an estimate of the linear slope for each month and this showed trends of both different magnitude and direction for each month but no homogeneous seasonal trend. In winter and early spring, the Hg concentration at Zeppelin is controlled by direct transport of polluted air from strongly industrialized countries in continental and eastern Europe. The declining trend in these months over the sampling period may be a response to declining emissions from European mid-latitude source regions. In spring the monthly mean Hg concentration is controlled by AMDEs. During the ten years of sampling, AMDEs occurred less frequently, which resulted in an increasing statistical concentration trend in spring (Berg et al., 2013). During summer, GEM concentrations are controlled by re-emission from surfaces, especially by evasion from the ocean. The increasing trend in summer may be due to enhanced evasion from the ocean as sea-ice cover declines and the summer sea-ice minimum shrinks (Hirdman et al., 2009).

Intensive measurement campaigns designed to understand the cycling of atmospheric Hg provide much information, but they apply only to a snapshot in time. Care should be taken when scaling up such measurements to longer periods and wider geographical areas (Pfaffhuber et al., 2012). Long-term measurement programs can provide valuable information towards

understanding the processes involved in Hg cycling in the polar atmosphere. These observations are also important for addressing the potential effects of climate warming on the Hg cycle. Long-term observations are also needed to identify possible effects of changes in global anthropogenic Hg emissions on Hg deposition in the polar regions (Steffen et al., 2008).

4.2 Geochemical processes in the snow and ice cover

Concentrations of gases in the Arctic atmospheric boundary layer are affected by chemical reactions on ice surfaces and in snowpacks. These processes are the result of a combination of factors: snowpacks are permeable, allowing exchange between snowpack interstitial air and the atmosphere (Domine et al., 2008); snowpacks strongly forward scatter radiation, resulting in sunlight penetration well below the surface (Simpson et al., 2002); and deposition to snowpacks concentrates many compounds. Since the discovery of AMDEs in the late 1990s (Schroeder et al., 1998), research has focused on Hg in the Arctic cryosphere because snow and ice are key receptors for atmospheric deposition, and chemical processes occurring at the snow-air interface affect how much Hg enters the environment. The amount of Hg retained in snow and ice reflects, in part, where it is found (Poulain et al., 2007a; St Louis et al., 2005). The high variability in total Hg concentrations reflects complex processes and transformations within the snowpack. Freshly deposited Hg on snow is highly reactive, and a large proportion of the Hg deposited during AMDEs is quickly returned to the atmosphere owing to the photo-reduction of Hg(II) to GEM within the surface snowpack (Lalonde et al., 2002; Poulain et al., 2004). The presence of halogens in snow may act to retain Hg and, indeed, elevated snow Hg concentrations are more often found in coastal environments where deposition of sea salt can be expected (Douglas and Sturm, 2004; Poulain

et al., 2007a; St Louis et al., 2007). Conversely, these higher concentrations may occur primarily because AMDEs are a result of photochemical reactions involving marine halogens and ozone. Snowpack reactions affect not only Hg, but also nitrogen oxides, halogens, ozone, and organic compounds. Snow plays a significant role in accumulating and redistributing Hg (Johnson et al., 2008; Steffen et al., 2008). Burial through snow accumulation, sublimation, condensation, and ice layer formation are other processes that promote retention of Hg in the snowpack (Douglas et al., 2008). While deposition occurs at the snow surface, Hg can also be redistributed within the snow. Migration of GEM within the snowpack and subsequent oxidation can result in accumulation of Hg within deeper layers (Dommergue et al., 2003a; Fain et al., 2006). Melt events may redistribute Hg towards deeper layers away from the photochemically active part of the snowpack. Concentrations are commonly highest in the surface layer (Dommergue et al., 2003b; St Louis et al., 2007), but the middle stratum or depth hoar—a layer of large crystals at the base of the snowpack—can sometimes also be dominant reservoirs of Hg. Therefore, post-depositional processes through the entire depth of the snowpack may influence Hg accumulation.

At two sites near Barrow, Alaska, surface snow Hg concentrations increased by 15 to 30 times during a nine-day AMDE (a period during which there is a sharp drop in GEM concentration in the lower atmosphere), and much of this Hg was retained until snow melt (Johnson et al., 2008). Upon snowmelt, snow impurities are released to meltwater-fed catchments, soil and aquatic systems, potentially delivering a pulse of highly concentrated solutes and contaminants. During the first flush, roughly 80% of solutes, including Hg, are removed from the snowpack (Dommergue et al., 2010). The Hg concentrations in surface water in a small creek near the sampling sites ranged from 10 to 15 ng/L (i.e., they were elevated due to the deposition event) during a ten-day peak snow runoff period, indicating that AMDEs can lead to an increased loading of Hg on aquatic ecosystems (Douglas et al., 2008). Changes in the size of the storage reservoirs, and in the processes driving these changes, could increase the delivery of heavy metals to the tundra and coastal ecosystems through changes in dry and wet deposition via rainfall and fog.

Analysis of the heavy metal content in snow provides useful information on aerosol composition and long-range distribution patterns of anthropogenic substances emitted into the atmosphere at lower latitudes (Colin et al., 1997; Walker et al., 2003). There are few data available on heavy metal concentrations in snow in the Russian Arctic, and what there are mostly show no evidence of heavy metal pollution except for areas near the large industrial centers of the Kola Peninsula (Monchegorsk, Nickel) and Siberia (Norilsk) and along the Northern Sea Route (Gordeev and Lisitsyn, 2005; Shevchenko et al., 2010). In the near future, new heavy metal pollution from exploration activities and the extraction of oil, gas and mineral resources is a possible scenario as accessibility through new shipping routes via the Northwest Passage and longer open water seasons may enable more economic development (AMAP, 2007; Prowse et al., 2009).

The volume of snow on sea ice is declining owing to the reduction in Arctic sea-ice extent at the end of the melt season in September (Bintanja and van der Linden, 2013) and changes in precipitation patterns (e.g., shifts from snowfall to rainfall) delay the onset of autumn ice formation and cause a reduction in multi-year sea ice (Harbeck et al., 2008). These environmental changes cause changes in the size of contaminant storage reservoirs. The Arctic is likely to become a more effective trap for heavy metals, in particular Pb, Cd and zinc, owing to the increase in precipitation projected under a warmer climate (Macdonald et al., 2005; AMAP, 2011c). A statistical study of the impact of environmental factors on Hg in the snowpack showed that snowpack preferentially retains oxidized Hg deposited through wet processes compared to oxidized Hg deposited through dry processes (Durnford et al., 2012). This preferential retention may reflect the importance of the burial of deposited Hg by fresh snowfalls (Witherow and Lyons, 2008; Dommergue et al., 2010). The preferential retention by the snowpack of Hg deposited through wet processes has important implications for climate change. If precipitation patterns change in a future climate, the spatial distribution of wet deposition will also change. As a result, the spatial distribution of snowpack Hg concentrations and the transfer of Hg to the underlying surface with snowpack meltwater will also change (Durnford et al., 2012).

4.3 Atmospheric deposition

Mercury species are removed from the atmosphere by dry or wet deposition. Dry deposition is defined as the direct transfer of gaseous and particulate species to a surface and proceeds without the aid of precipitation. Wet deposition encompasses all processes by which atmospheric Hg is transferred to the surface in aqueous form. The sum of dry and wet deposition is termed total deposition. Measurements of total atmospheric Hg deposition are complicated, and so the usual way of monitoring Hg deposition is by collecting precipitation, which indicates the amount of wet deposition. Dry deposition of Hg is often assumed to be negligible in most environments but may be underestimated in the monitoring of bulk total Hg deposition under some circumstances. Measurements of wet deposition are available and well documented. Several wet deposition studies have been conducted in the northern hemisphere for Hg (Iverfeldt, 1991; Hall et al., 2005; Cole et al., 2014). In areas with frequent precipitation, wet deposition dominates. In more arid climates, dry deposition is the dominant deposition process, as found in polar regions. Depending on location and time of year, dry deposition may be the most important component of the atmospheric Hg deposition (Zhang et al., 2012).

Mercury in wet deposition has been collected on a monthly basis in southern Norway (Birkenes) using bulk samplers from 1990 to the present. The measurements have been compared with results from DEHM model simulations using global historical AMAP Hg inventories for 1990, 1995, 2000 and 2005 as input to the Hg module of the model (Christensen, 1997; Christensen et al., 2004; AMAP, 2010; Brandt et al.,

2012). Comparisons of measurements and model output are shown in Fig. 4.3.

The measurements show good agreement with model results, and indicate the same overall trend. During the 25 years of data, there is a decline in the annual mean concentration of total Hg. This decrease averages 0.5 ng/L/y, which is comparable to measurements from northern North America (Prestbo and Gay, 2009; Cole et al., 2014). Simultaneously, annual total precipitation has increased and the net effect is that the annual total Hg wet deposition remained unchanged over this period. Increased precipitation results in a dilution of the Hg concentration.

Several groups have studied dry deposition as well as (re)emission of GEM either by micrometeorological methods (gradient or relaxed eddy accumulation) (Lyman et al., 2007; Steffen et al., 2008) or by enclosure methods (Carpi and Lindberg, 1998), but the fate of atmospheric Hg is largely determined by the deposition of GOM. There are few studies in the literature measuring GOM deposition (Lindberg and Stratton, 1998; Poissant et al., 2004; Skov et al., 2006). Measurement of GOM flux is difficult given the low concentrations and the analytical precision of current methods (Gustin and Jaffe, 2010). Several reports on estimated and modeled GOM deposition are available. The overall conclusion of these studies is that the chemistry within or at the surface layer is very important for the budget and global circulation of Hg. Several studies have looked at the specific reactions within and on various surfaces and have found that fast reduction reactions may reduce oxidized Hg (Hg(II)) to elemental Hg that is afterwards emitted as GEM (Dommergue et al., 2003b; Lalonde et al., 2003; Ferrari et al., 2004; Fritsche et al., 2008).

4.3.1 Atmospheric deposition as interpreted from the nationwide moss survey

Surface soil is an important sink for metals and other air pollutants. In the boreal zone, which comprises large parts of Canada, Fennoscandia, and Russia, the surface soil is generally rich in humic substances, which have a high cation exchange capacity and may bind some metals even more strongly by complexation. The organic surface soil is therefore an efficient integrator of metals from the atmosphere and may as such be a useful supplement to other methods used to measure metal deposition on shorter time scales. Comparing the spatial distribution of element concentrations in moss and the humic surface soil layer (Figs. 4.5 to 4.8), it seems evident that the sources of the elements are the same for both environmental compartments because the spatial concentration distributions compare very well (Meyer et al., 2014). Data analyses of soil samples from Norway show that as much as 80–90% of Pb in the humus layer is likely to be from air pollution, even in the far north, similar to the percentages for Pb in moss. Soil has a ‘memory’ because the concentration decrease observed in the moss data over time is not visible in the soil. There is no temporal information existing on Hg in soil.

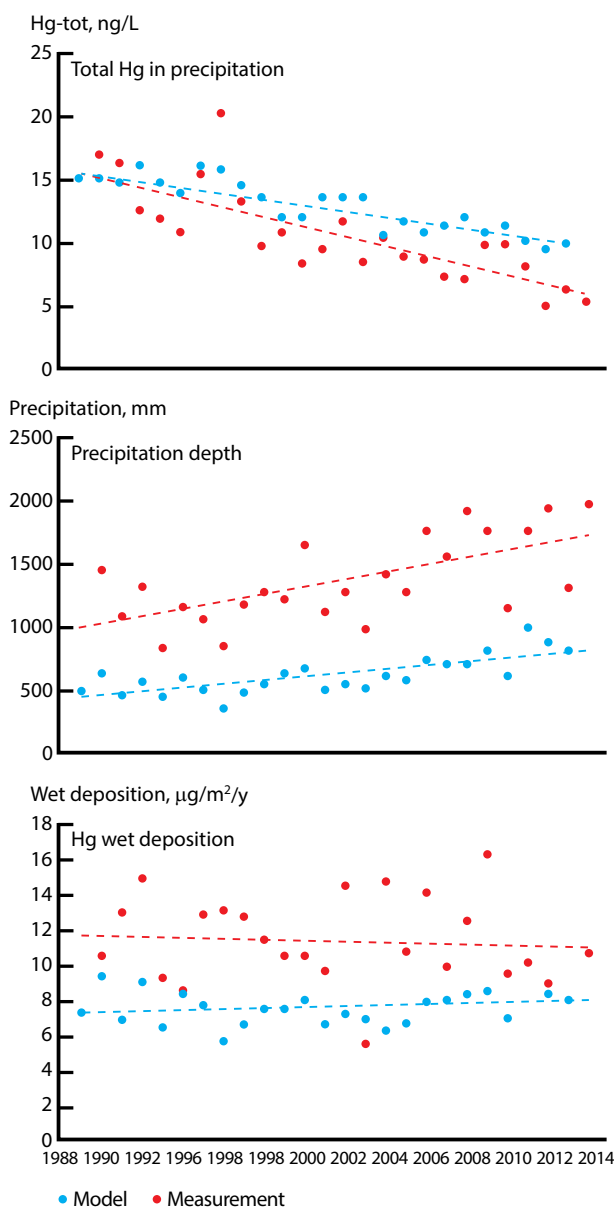


Figure 4.3. Comparison of DEHM model results and measurements at Birkenes in southern Norway. Annual mean concentrations of total Hg in precipitation (upper), total annual precipitation depth (middle), and annual wet deposition of total Hg (lower).

Source: NILU and Aarhus University.

Monitoring atmospheric deposition of metals is important to assess their influence on terrestrial and aquatic ecosystems and their transfer through the food web to humans. To assess the temporal and spatial trends in heavy metal deposition to Norway, results were used from the previous 35 years of the moss study. The heavy metals in moss survey was originally established in 1980 as a Swedish initiative (Rühling and Tyler, 1970). The idea of using mosses to estimate atmospheric heavy metal deposition was developed in the late 1960s by Rühling and Tyler (1968) and the aim was to map patterns of heavy metal bioaccumulation in terrestrial ecosystems.

Mosses, especially the carpet-forming species, obtain most of their nutrients directly from wet and dry deposition; there is little uptake of metals from the substrate and so they are particularly suitable for monitoring airborne pollutants. In Norway, the terrestrial moss *Hylocomium splendens* (Fig. 4.4) has been used as it is found throughout the country, is easily recognizable and grows in annual shoots, which allow sampling and analysis of annual accumulation. Monitoring the heavy metal concentration in mosses provides a surrogate, time-integrated measure of element deposition from the atmosphere to the terrestrial system. It is easier and cheaper than conventional deposition analysis as it avoids the need for deploying large numbers of deposition collectors with an associated long-term program of routine sample collection and analysis. Therefore, a much higher sampling density can be achieved than with deposition analysis (Gusev et al., 2009). Although the heavy metal concentration in mosses provides no direct quantitative measurement of deposition, this information can be derived by using regression approaches relating the results from moss surveys to measured deposition monitoring data (Berg and Steinnes, 1997; Berg et al., 2003). Comparison of the metal concentration in mosses with total deposition simulated across Europe using the European Monitoring and Evaluation Programme (EMEP) atmospheric transport model MSCE-HM showed that the modeled and measured spatial and temporal trends are similar for Cd and Pb but less similar for Hg (Gusev et al., 2009; Harmens et al., 2010).

As Hg in ambient air is predominantly found in the vapor phase and has a residence time of about one year, it must

be considered a global pollutant (Schroeder and Munthe, 1998) without distinct spatial deposition patterns. The lack of correlation between modeled deposition values for Hg and observed concentrations in moss may relate to the specific chemistry of Hg and corresponding interactions with the moss. In Scandinavia, the Hg deposition as measured by precipitation analysis in the 1980s showed a steep increase from north to south, similar to that for other metals predominantly supplied by long-range atmospheric transport (Iverfeldt, 1991). In moss collected in Norway during the same period, however, the Hg level was fairly uniform with no distinct north-south gradient (Steinnes and Andersson, 1991). This geographical distribution of Hg has been confirmed in more recent moss surveys and indicates that wet deposition of Hg(II) alone cannot be responsible for the distribution observed (Steinnes et al., 2011). The moss must also be able to retain dry-deposited gaseous Hg(0) to a significant extent. Indeed, several studies have shown the importance of the dry-deposited gaseous Hg(0) contribution to Hg concentrations in vegetation (Lodenius et al., 2003; De Temmerman et al., 2007, 2009). In addition, AMDEs might contribute to the Hg deposition in the Arctic regions of Europe and possibly explain part of the elevated Hg concentrations observed in moss in northern Norway (Schroeder et al., 1998; Berg et al., 2008). It could be that the deposition pattern depicted by the moss survey is a better measure of net Hg supply to the terrestrial ecosystem than indicated by EMEP modeled calculations.

Based on results from the moss surveys, Pb (Fig. 4.5) and Cd (Fig. 4.6) show the highest deposition in the southernmost



Figure 4.4. *Hylocomium splendens* is a species of moss used for monitoring heavy metal deposition in the terrestrial environment. Source: Katrine Aspmo Pfaffhuber, NILU.

Figure 4.5. Lead in moss in Norway (mg/kg) collected between 1977 and 2010. Source: Steinnes et al. (2011).

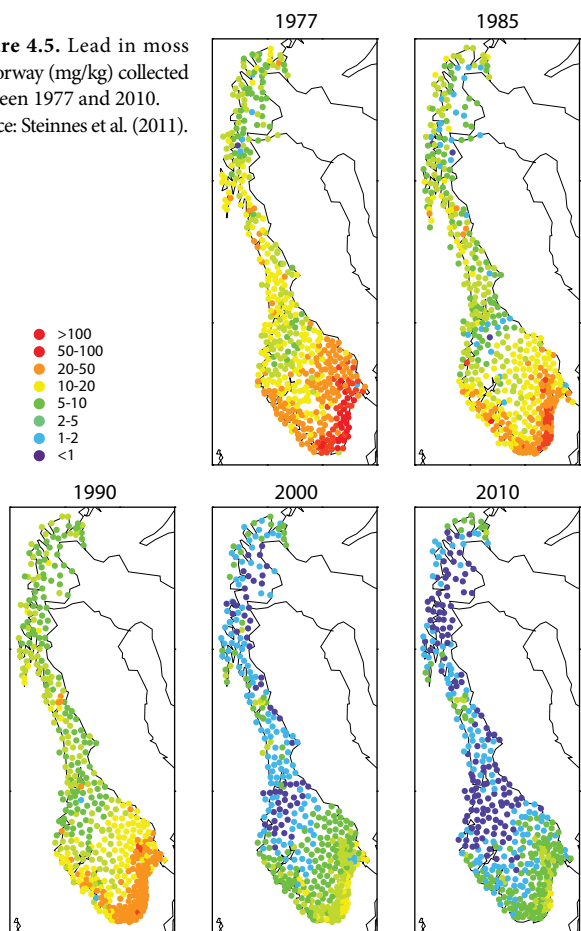


Figure 4.6. Cadmium in moss in Norway (mg/kg) collected between 1977 and 2010. Source: Steinnes et al. (2011).

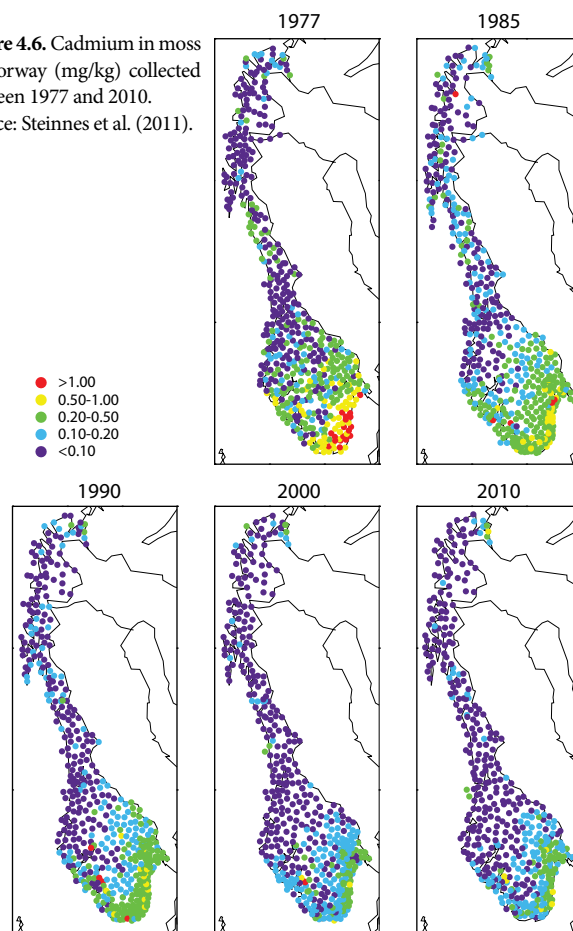


Figure 4.7. Mercury in moss in Norway (mg/kg) collected between 1985 and 2010. Source: Steinnes et al. (2011).

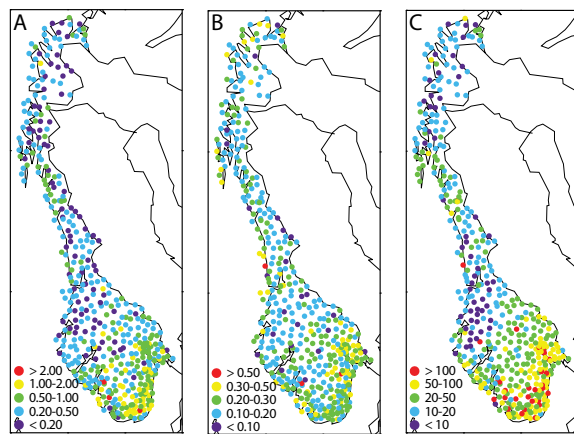
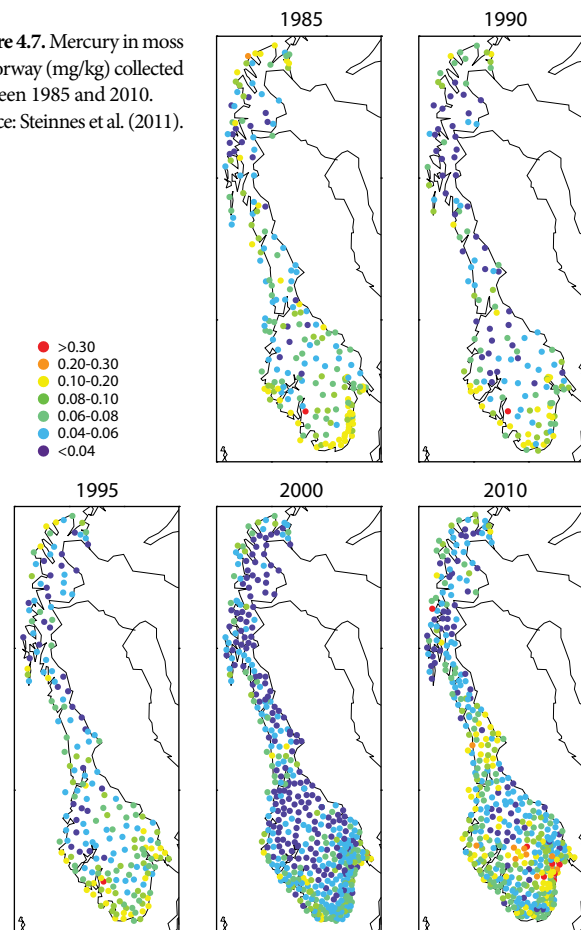


Figure 4.8. A) Cadmium concentrations (mg/kg) in soil collected in 2005. B) Mercury concentrations (mg/kg) in soil collected in 2005. C) Lead concentrations (mg/kg) in soil collected in 2005. The color code is different on each map. Source: NILU.

part of Norway, whereas Hg (Fig. 4.7) shows a more uniform deposition distribution. For Pb and Cd, this is in accordance with monitoring data for precipitation in different parts of Norway. Data analysis has shown that the substantially higher deposition of these elements in southern Norway compared to the rest of the country is clearly due to long-range atmospheric transport from highly populated and strongly industrialized countries elsewhere in Europe. As much as 90% of the Pb deposition in the south may be due to transboundary pollution. Moreover, deposition of Pb and Cd has strongly decreased over the past 30 years. For Pb, this is due to the introduction of unleaded fuel, with the concentration found in moss in 2005 only about 5% of the 1977 levels. For Cd, the deposition contribution from long-range atmospheric transport decreased significantly until 2000, and since then further changes appear to be small. The change in deposition is also evident when considering atmospheric concentrations. Long-term observations (47 years) of the chemical composition of total suspended particles from Kevo in the Finnish Arctic show significant decreasing trends for most components, including Pb and Cd. This is consistent with the reported trends in European emission inventories (Laing et al., 2014b). Nevertheless, concentrations were significantly higher than for other Arctic locations due to the relative proximity to Euro-Asian pollution sources (Laing et al., 2014a).

In contrast, Hg showed a uniform deposition distribution as derived from moss sampling and analyses. There seem to be no temporal deposition trends and no correlation with monitoring data for precipitation, pointing towards dry deposition as an important contributor to Hg in moss. Source region analysis of Hg sources to Norway and the Norwegian Arctic has shown that the heavily populated and strongly industrialized countries in continental and eastern Europe are also the main sources of Hg deposition, particularly in winter (Hirdman et al., 2009). Although Hg emissions from these parts of Europe have decreased over recent decades, this decrease is not evident from Hg concentrations in moss.

On the basis of current knowledge it is not yet possible to quantify the relative contribution of Hg dry deposition in Norway or to say whether this changes with latitude or time. The climate is projected to become wetter and warmer in the northern hemisphere, including in Scandinavia (AMAP, 2011c). Wet deposition of Hg is projected to remain unchanged, while re-emission is likely to increase as temperature is a key factor in this process. The overall result could be a smaller net Hg input to the terrestrial environment from this contributor. Changes in Hg cycling in the terrestrial system under a warming climate are complex, however, and deposition followed by re-emission is only one contributor.

4.4 Impact of global climate change on mercury

The climate in the Arctic is changing rapidly; however, these changes are not uniform across this vast and diverse

region. Climate change could affect the delivery of Hg to Arctic environments by altering atmospheric dispersion and deposition, hydrological transport, release from permafrost, and local sources from human activities (NCP, 2012). Previous research has indicated that current and future climate change will influence the environmental fate of Hg (Stern et al., 2012). Climate change has the potential to affect the transport of Hg in Arctic environments owing to close links between the Hg cycle and temperature, the cryosphere, and organic matter cycling (Macdonald et al., 2005). Changes in food webs driven by climate warming are also expected to affect Hg biomagnification to higher predators. Some impacts on food webs may enhance Hg bioaccumulation while others may reduce it, and the net effect is likely to vary due to regional differences in the structure of aquatic food webs and their response to environmental change. Potentially, the most important role of climate change concerns the net formation of MeHg in Arctic aquatic ecosystems (Outridge et al., 2008).

The biochemical Hg cycle in the Arctic is sensitive to climate variability and changes in the cryosphere. Higher Arctic air temperatures will accelerate the loss of the sea-ice cover, which will lead to a warmer upper ocean and consequently to a change in the atmospheric connections between the Arctic and lower latitudes (AMAP, 2011c; IPCC, 2014). Atmospheric Hg chemistry, including AMDEs, may also be affected by changing temperatures and precipitation (AMAP, 2011b). Atmospheric Hg deposition during AMDEs is driven by the oxidation of Hg(0) with bromine atoms, and this reaction is likely to be temperature dependent (Goodsite et al., 2004; Cole and Steffen, 2010). Theoretical calculations have shown that rising temperatures in the Arctic could slow the oxidation and deposition of atmospheric Hg in marine environments. The supply of reactive bromine for Hg(0) oxidation may also be affected by changes in sea-ice formation and by temperature effects on bromine speciation (Cole and Steffen, 2010).

For terrestrial systems, the rise in temperature has led to a shorter period of ice and snow cover, reductions in glacier size and mass, thawing permafrost, changed vegetation and river discharges, and increasing incidents of forest fires. In addition, the rise in temperature has also led to a rising water table that influences the methylation and changes migration pathways (AMAP, 2011b). A shorter period of ice and snow cover means that inorganic Hg is transformed to MeHg through greater productivity in wetlands and freshwater (AMAP, 2011b).

Thawing permafrost and a deepening of the active layer in summer are expected to increase particulate and dissolved organic carbon transport to Arctic lakes and rivers (Prowse et al., 2006; Xu et al., 2009). This means that watershed export of Hg, which is associated with organic carbon transport, may also change. Permafrost slumping and subsidence may result in the export of soil-bound Hg to freshwater (Klaminder et al., 2008; Rydberg et al., 2010). Thawing of

the vast areas of northern frozen peatlands may release globally significant amounts of long-stored Hg (including Hg of natural origin) and organic matter into Arctic lakes, rivers and the ocean. Warm and microbially-active thaw ponds receiving inputs from adjacent slumping permafrost soils may be sources of MeHg in the Arctic environment. Greater hydrological connectivity from permafrost thawing may potentially increase transport of MeHg from thaw ponds to neighboring aquatic ecosystems (MacMillan et al., 2015). Rivers are a significant source of Hg to marine environments such as Hudson Bay (Hare et al., 2008). Fluxes of Hg from rivers could be altered by long-term changes in hydrology owing to the positive relationship between discharge and aqueous Hg concentrations (Leitch et al., 2007; Graydon et al., 2009). Snowmelt is the dominant source of Hg to many Arctic lakes (Semkin et al., 2005). Changes in the amount of snow, the timing of snowmelt, or the importance of rain in the hydrological cycle could have implications for Hg delivery to these ecosystems (Prowse et al., 2006). Higher annual precipitation could increase wet deposition rates of Hg to Arctic environments.

Reductions in summer sea ice could stimulate commercial shipping through the Northwest Passage and economic development throughout the Canadian Arctic (Prowse et al., 2009). Greater accessibility through new shipping routes and a longer open water season may allow for more exploration activity and greater extraction of oil, gas, and mineral resources during the 21st century (Prowse et al., 2009). These activities present potential risks to the Arctic environment including increased fossil fuel combustion, mineral and gas/oil processing as well as waste generation that could result in new local sources of Hg.

Climate warming may affect Hg levels in biota through changes in food-web structure, growth rates, or ecosystem productivity (Braune et al., 2014). A change in the amount of Hg available for uptake in food webs may also affect bioaccumulation. Owing to climate change, the structure and dynamics of marine food webs are undergoing change (Wassmann et al., 2011). Arctic marine ecosystems are further affected by the reduction in the extent and thickness of sea ice. The ability to adapt to ecosystem shifts depends on the individual species of marine animal, so the effect on Hg bioaccumulation depends on the food-web structures and their response to climate change. Climate warming will increase the productivity of freshwater ecosystems which may, in turn, reduce Hg bioaccumulation in fish through growth dilution processes at lower trophic levels in the food web. Trophic structures vary geographically and changes may occur in two ways, either from the 'bottom-up' or from the 'top-down'. Bottom-up changes are linked to primary or secondary productivity, such as plankton and other organisms, and are associated with changes in such conditions as stratification, nutrient supply, light intensity or ice cover (AMAP, 2011b). All of these changes can have major effects on biological production at a higher trophic level. A good understanding of ecosystem dynamics along with a baseline of Hg trends is required to test for climate change impacts on

food-web structure and Hg processes. Predators and other higher-trophic level species exert a top-down influence on food-web Hg levels owing to their feeding ecology and dietary preferences. Changes at the top of the food web are related, for example, to the loss of sea-ice cover and subsequently to a shift in diet, habitats and/or migration pathways. Gaden et al. (2009) found that Hg levels in seal muscle were significantly higher during periods following both a relatively short and a relatively long ice-free season, because of ice-related changes in prey availability.

Available information on the global Hg cycle shows that Hg is in a relatively rapid equilibrium between the atmosphere and ocean surface waters (tens of years) but slower with deep-ocean waters (hundreds of years) (Lamborg et al., 2002; Mason et al., 2003, 2012; Sunderland and Mason, 2007). Sunderland and Mason (2007) showed that anthropogenic emissions had enriched the atmosphere by 300–500%, all surfaces by 25%, and deep-ocean waters by 11%. The lag between changes in atmospheric deposition and seawater Hg concentrations varied from decades in most of the Atlantic Ocean to centuries in parts of the Pacific Ocean (Sunderland and Mason, 2007). Many of the processes in the Hg cycle are temperature-dependent or involve parameters that might alter in a changing climate. Temperature changes may affect rates of reactions involving Hg species or reactions that determine the composition of possible reactants. In addition, changes in vegetation, land use or, for example, the populations of bacteria in the sea may affect the Hg cycle. Thawing Arctic tundra could liberate large reservoirs of Hg, while a change in humidity or precipitation patterns could lead to more wildfires, which are known sources of atmospheric Hg. AMDEs are dependent on the presence of refreezing leads, and their geographical extent is likely to change with higher temperatures in the Arctic and Antarctic (Macdonald et al., 2005; Cole and Steffen, 2010). Higher ocean temperatures are likely to increase the (re)emissions of volatile Hg species, and changed temperatures may also alter the chemical reactions that occur in the atmosphere.

In relation to anthropogenic emissions, future choices regarding energy sources also need to be taken into account (Sundseth et al., 2015). If coal combustion replaces oil, it will lead to an increase in anthropogenic Hg emissions to the atmosphere. Conversely, if fossil fuel combustion is replaced by 'clean' energy sources such as wind, wave or solar energy, Hg emissions will decrease.

The potential effects are many and can drive the environmental Hg cycle in various directions. As a result, it is not possible to say whether the combined effect of all these changes will be to increase or reduce the concentrations of Hg in the environment. Large gaps in knowledge concerning the processes controlling Hg behavior in the different environmental compartments mean that there are large uncertainties associated with any assessment of future Hg concentrations and fluxes in the environment. The response of the biogeochemical Hg cycle to future climate change is thus a priority issue for the scientific community and for society in general.

4.5 Metals and human health

4.5.1 Northern Norway

Table 4.1 shows metals data from the most recent sampling of pregnant women in the MISA study in northern Norway (2006–2008) juxtaposed with concentrations measured in pregnant women in various regions of northern Norway in 1994–1995, which were reported by AMAP (1998). This comparison using past levels in pregnant women in these cities in northern Norway is geographically appropriate because the MISA study populations lived in Nordland, Troms and Finnmark counties (Hansen et al., 2011).

Levels of Cd, Hg, and Pb have declined since 1994–1995 in pregnant women, which indicates that fetal exposure *in utero* has also been declining. However, recent selenium (Se) levels are also lower than those observed in 1994–1995, which could indicate a reduction in the consumption of marine food. Fish consumption was found to be a positive predictor for Se and Hg levels, along with multivitamin intake for Se (Hansen et al., 2011). Also, age reached significance only for Hg, while an inverse association was observed between parity and Hg. The observed concentrations for Pb and Cd suggest exposures expected from hunting traditional foods (Pb) and smoking (Cd). Moreover, smoking may increase the body burden of Pb, reinforced by the association between Pb levels and Cd concentrations for smokers (Hansen et al., 2011).

4.5.2 Northern Russia

Rylander et al. (2011) reported levels of Hg, Pb, Cd and Se in whole blood in men and women from the Komi Republic in Izhma and Usinsk (Table 4.2). Statistical analysis of these data indicated that Hg and Pb concentrations increased with age, and that men had significantly higher concentrations of Pb than women. The study group from Izhma had significantly higher concentrations of Cd when controlling for age and sex, and the study group from Usinsk had higher concentrations of Se than the others. Parity did not affect metal concentration.

Table 4.3 shows that maternal blood levels of Pb decreased by 21% between 2001–2002 and 2007, although the range broadened to include much lower and much higher levels in 2007. Maternal Hg levels remained essentially the same between the two sampling periods. This suggests a continuous exposure to Hg, probably through fish and seafood. Blood Pb levels in children have not changed, whereas concentrations of Hg in blood decreased by 31%. The decrease in Hg concentrations in 2001–2003 cord blood compared with 2007 infant blood indicates that Hg exposure during early life was less than that *in utero*. There may be differences in Hg exposure between mothers and their children; however, due to the relatively short half-life of MeHg in the body (Yaginuma-Sakurai et al., 2012), it cannot be determined from these results whether the potential differences are the result of long-term behavior or whether there were more recent changes in exposure prior to the 2007 sampling.

Table 4.1. Concentrations of metals ($\mu\text{g/L}$ whole blood) in pregnant women (early pregnancy) from northern Norway, in 1994–1995 and 2006–2008. Data are presented as geometric mean [standard deviation] or geometric mean (range) (AMAP, 1998; Hansen et al., 2011).

Metal	N	Kirkenes 1994	N	Hammerfest 1994	N	Bergen 1994	N	Tromsø 1995	N	Northern Norway ^a 2006–2008
Cd	40	0.5 [± 0.8]	57	0.5 [± 0.6]	50	0.5 [± 0.5]	15	0.4 [± 0.5]	282	0.2 (0.04–2.7)
Hg	40	3.4 [± 1.2]	57	2.5 [± 0.8]	50	3.4 [± 1.1]			282	1.2 (0.1–6.6)
Pb	40	12.4 [± 6.2]	57	12.4 [± 6.2]	50	14.5 [± 6.2]	15	10.4 [± 6.2]	280	7.4 (2.22–25.8)
Se	40	124 [± 15]	57	106 [± 17.4]	50	107 [± 18.2]	15	93.2 [± 18.2]	281	84.7 (58.2–128)

^aAverage age = 30.6, age range = 18–43, parity = 0.9.

Table 4.2. Concentrations of metals ($\mu\text{g/L}$ whole blood) in study participants from the Komi Republic, Russia in 2009–2010. Data are presented as geometric means (range). Source: Rylander et al. (2011).

Metal	Men		Women	
	Izhma (n = 25)	Usinsk (n = 25)	Izhma (n = 25)	Usinsk (n = 25)
Hg	2.3 (1–8.1)	2.2 (1–10)	2.3 (1–10)	2.3 (1–6.4)
Pb	33 (14–88)	32 (11–63)	27 (11–57)	23 (11–63)
Cd	0.43 (0.1–1.4)	0.28 (0.1–1.0)	0.40 (0.1–1.5)	0.23 (0.1–1.3)
Se	88 (61–130)	99 (61–134)	87 (57–119)	100 (63–139)

Table 4.3. Concentrations of metals ($\mu\text{g/L}$ whole blood) in maternal (n=17), cord and child blood (n=17), 2001–2002 and 2007, from coastal Chukotka (Lavrentia and Lorino, Russia). Data are reported as geometric means (range). Source: Dudarev et al. (2010).

Contaminant	Maternal		% ^a	Cord		% ^a
	2001–2002 24.6 y (15–33)	2007		2001–2002	2007 5.5 y	
Hg	1.6 (0.5–3.9)	1.6 (0.5–4.8)	+1	1.4 (0.5–3.3)	0.93 (0.5–2.7)	-31
Pb	37.5 (18.3–76.8)	29.6 (4.9–137)	-21	37.6 (14.3–78.3)	38.2 (6.9–102)	+2

^aGeometric mean values for 2007 relative to geometric mean values for 2001–2002 (100%).

5. Education of students within the project

A proportion of the funding by the Nordic Council of Ministers for this project provided support for the work of three students on Master of Science degrees and one student on a Bachelor of Science degree, while another two students were able to initiate MSc studies under funding for this project. This project has successfully integrated their education with the research within this project as well as several ongoing studies financed by the Svalbard Environmental Fund. The Annex to this report provides brief summaries of the results presented in the completed theses.

6. Future perspectives

The study reported here, together with the first stage of this project (AMAP, 2011a) belongs to the first integrated research effort to investigate the combined effects of environmental pollutants and climate change in the Arctic. This integrative and strongly interdisciplinary approach can serve as an example for similar studies in other regions of the world. The study is built on an interdisciplinary research foundation that acknowledges and integrates the complementary expertise of the participating research groups. Strong linkage between modelers, environmental biologists, environmental chemists, and human health experts has provided a fruitful and inspiring scientific environment in which the success of the study is founded.

This project facilitated the collaboration and integration of scientific work between researchers modeling the transport and environmental distribution of contaminants in the Arctic and researchers investigating uptake and biomagnification of contaminants in Arctic food webs, and the potential influence of climate change on these processes.

Based on the interdisciplinary nature of the project, strong thematic linkages were established between the theoretical and empirical aspects of the study; investigations were also conducted covering both organic and inorganic components in relation to environmental and human subjects. The most important overarching research topics are:

- Seasonal versus climate change influences on the transport and cycling of POPs and Hg to and within the Arctic
- Transfer processes for POPs and Hg between the atmosphere and water and ultimately to biota, and the potential influence of climate change on these processes
- Intercompartmental transfer of particles from local sources in the Arctic emitted into the atmosphere, their deposition to the marine environment and the sorption of POPs onto these particles, and ultimately their influence on the bioavailability of these particle-bound organic contaminants, including 'emerging' contaminants, POPs and PAHs to marine organisms
- Factors influencing the deposition of Hg on the terrestrial environment and particularly the effect of increased precipitation and temperature

- Concentrations of environmental contaminants in food items, particularly wildlife species consumed as part of a traditional diet in Arctic Indigenous communities, ongoing changes in dietary habits of Arctic residents, and risk assessment of dietary contaminants for humans
- Potential climate-change related changes in dietary POPs exposure for humans.

The scientific results will support regional regulatory efforts to establish reliable and sustainable regulation strategies in order to minimize the potential hazardous effects of anthropogenic pollution on the region.

Although the scientific understanding of climate change impacts on metal and POPs cycling and impacts in the Arctic has increased dramatically in recent years, it is still not sufficient to enable predictions of future environmental concentrations and impacts. Long-term monitoring programs are thus necessary to provide information-rich data and early detection of trends, and for understanding the processes involved in atmospheric cycling of Hg, POPs and 'emerging' contaminants in the Arctic and how climate change will impact on these processes. These monitoring programs are also needed to track the effect of changes in global anthropogenic Hg emissions on Hg deposition in polar regions. Results from long-term monitoring programs are also crucial for assessing and understanding trends and fate of legacy and emerging organic contaminants. Such programs are of high priority for providing policy makers with the best information available as well as for contributing to effectiveness evaluations of the Stockholm Convention on Persistent Organic Pollutants and the Minamata Convention on Mercury.

Long-term age-period-cohort studies are very important for understanding the fate of POPs and Hg in humans and should be given high priority for research in the future.

Although not covered in this project, more research is needed on biovector-mediated contaminant transport and transfer across ecosystems in the Arctic. Understanding of the environmental effect of animals as biovectors for contaminant transport to and within the Arctic is still poor. Empirical data need to be collected and combined with modeling to understand the potential impact of climate change on invasive species as biovectors. It is thus important to evaluate migration and invasive behavior of populations as a potential early warning signal for effects on ecosystems caused by ongoing global climate change (Kallenborn et al., 2012). Current transport models should be expanded to include biovector transport processes.

7. Conclusions

7.1 Persistent organic pollutants

DEHM model results indicate that, in a future warmer climate, there will be higher volatilization of POPs, which favors greater atmospheric transport to the Arctic. However, higher temperature, higher precipitation and declining snow/ice cover may mitigate this increase or, as was the case for the PCB congeners studied in this project, may decrease total transport to the Arctic and decrease concentrations in Arctic environmental compartments. Changes in POPs emissions, for example, due to changes in temperature or reduced use of the chemicals, are likely to have a much greater effect on POPs transport to the Arctic than climate change.

Climate change will, however, influence the distribution of volatile and semi-volatile POPs between environmental media (i.e. air, water, soil), thus impacting long-range transport of these contaminants to the far north and the Arctic from source regions further south. For POPs regulated under the Stockholm Convention, changes in distribution will be offset by increased degradation in a warmer climate.

Overall, model studies of POPs do not show clear agreement on whether climate changes will reduce or increase the concentrations of POPs in the Arctic. Further refining work is needed to understand this issue. This work will, among others, need to be based on the physical-chemical properties of the various groups of POPs occurring in the environment.

Arctic ecosystems are changing in response to climate change and some Arctic food-web structures are being affected in ways that may have potential consequences for the biomagnification of environmental contaminants. The ability to adapt to ecosystem shifts depends on the individual species, so the effect on Hg bioaccumulation depends on the food webs' structures and their response to climate change. Some climate change impacts on food webs may enhance Hg bioaccumulation while others may reduce it; the net effect is likely to vary geographically due to regional differences in the structure of aquatic food webs and their responses to environmental change.

Models of food-web biomagnification need to be refined to understand the full impact of climate change on bioaccumulation. Particulate organic carbon (POC) should be included in trophic magnification studies as it affects bioavailability and the uptake of contaminants at the base of the food web.

The impact of POC, sedimentation, primary production, lipids and a mixed ocean layer in modeling the bioavailability of contaminants in the water column/food web needs further elucidation. In particular, there is a need to identify all such factors relevant to food-web modeling and to determine their influence as quantitatively as possible on trophic magnification of

contaminants in the food web under investigation. Some factors increase trophic magnification while others decrease it. As was shown in the first phase of this project, elucidation of such factors is very important for understanding the trophic magnification of MeHg and the impacts of climate change on this process.

In the future Arctic climate scenario investigated in this project, simulated food-web biomagnification of POPs was only marginally different to that at present, with the magnitude and direction depending on the substance modeled; the average change from 2007 to 2107 was 0.4% for γ -HCH, -3.1% for PCB52 and -1.7% for PCB153. Thus, seasonal variation in bioaccumulation is greater than alterations in response to projected climate change. The observed seasonal variation in food-web biomagnification was explained by including seasonality in lipid content. As the best-fit seasonal scenario included the annual cycle in lipid content, this physiological factor is also crucial for making firm predictions of contaminant food-web bioaccumulation for the future. Seasonality in contaminant exposure, temperature or variations in diet did not significantly affect food web-biomagnification. Thus, changes in abiotic drivers alone are not sufficient to explain food-web bioaccumulation on a temporal scale. The seasonal variation in food-web biomagnification was greater than any variation forecasted in the future climate scenario.

Furthermore, including physiological (lipid) and ecological (food web) variations suggests that lipid is a driving factor in the seasonality in food-web biomagnification, rather than variations in trophic interactions. However, the food-web variations were based on a minor tweak in the relative feeding relationships of the same species in all seasons. Under climate change, it is anticipated that new, more southerly species will migrate into the Arctic, and there are no investigations yet of how species replacement and altered trophic structure due to climate change would affect the food-web biomagnification of contaminants.

Although climate-related changes in POPs uptake may occur at lower trophic levels of the Arctic marine food web, they may not be as large as present and future seasonal changes in contaminant uptake and bioaccumulation that occur naturally.

7.2 Trace metals including mercury

Monitoring heavy metals in mosses is very useful for understanding the deposition of metals to the terrestrial system. Moss is a useful medium for investigating the spatial distribution and temporal trends in toxic trace metals transported to the north via the atmosphere and deposited onto the terrestrial environment. Monitoring throughout Norway has shown a significant decrease in the atmospheric deposition of Pb and, until 2000, Cd, but no decrease in Hg deposition despite increased emission controls in some countries. However, moss surveys are uncertain indicators of Hg deposition. Metal concentrations in moss also show clear regional distributions for Pb and Cd, with higher levels in southern Norway, while

Hg levels in moss are more-or-less evenly distributed over the entire country.

Atmospheric Hg concentrations are decreasing in the northern hemisphere owing to reduced anthropogenic Hg emissions in North America and Europe since the 1990s. However, the Norwegian long-term monitoring stations do not pick up such a trend. Recycling of earlier emissions of anthropogenic Hg in the environment is the most plausible explanation for this.

Long-term observations also indicate reduced levels of Hg in aerosols over the central part of the Russian Arctic. However, if the decreased atmospheric levels are a result of increased deposition in sub-Arctic river catchment areas, rivers can become an important sink for Hg. The seasonal Arctic snow cover is changing due to changes in precipitation patterns, rising temperature and earlier snowmelt, which may change the contaminant storage reservoirs and the delivery of heavy metals to the tundra and coastal ecosystems.

Mercury reaches the biological systems via dry and wet deposition. Dry deposition is the more common in arid, polar climates. In areas where wet deposition is more important, increased precipitation will have a 'dilution effect' on Hg delivered to the surface, given that the amount of Hg is not increasing. While wet deposition of Hg has increased over recent decades from northern to southern Scandinavia, concentrations in mosses have remained evenly distributed. Dry deposition onto mosses 'levels out' geographical differences in wet deposition.

Climate-driven changes in precipitation patterns, river discharges, lake and sea ice, permafrost, vegetation and atmospheric connections between northern and southern latitudes are the most important environmental factors with regard to the fate of Hg in the Arctic. Changes in food webs owing to climate change can also affect the bioaccumulation and fate of Hg.

There are diverging results regarding the fate of MeHg in the literature. Increased temperatures could lead to higher microbial activity and thus a higher methylation rate for Hg. However, more rapid bacterial growth as a result of higher temperatures and increased demethylation of MeHg could result in lower levels of MeHg. It is most likely that the net methylation of Hg will increase with higher temperatures, but more research, especially on food-web changes and the role of lipids, reproduction and POC, is needed to understand the future fate of MeHg in the Arctic.

Current scientific information gives no clear indication as to whether climate change will increase or decrease the risk of exposure to Hg in the Arctic. Research indicates that climate change may act in both directions. As shown in this report, there is no linearity in relationships between future changes in Hg emissions and changes in concentrations of Hg in fish and seafood. More research is needed on the issue of climate change impacts on the effects of Hg on human health in the Arctic.

Further reductions in Hg emissions from anthropogenic sources worldwide would need to be introduced as soon as possible in order to lower the adverse impact of climate change on human health.

7.3 Contaminants and human health

Levels of most of the POPs measured in people from northern Norway have declined significantly since 1979, paralleling the implementation of measures during past decades to reduce or eliminate the production and use of these POPs. Trends in PFAS concentrations in this population have also tended to follow overall production and usage trends, and concentrations of PFOS and PFOA in blood are now declining. However, levels of several unregulated PFASs are continuing to rise. The differences observed between the compounds detected are also likely to be influenced by variations in global transport mechanisms, bioaccumulation potentials and consumer exposure.

The study of male and female volunteers from Nelmin-Nos, Izhma, and Usinsk indicated that older people in northwestern Russian communities may have significantly higher blood levels of many contaminants that accumulate in people over time, and this was also the case for PCBs and Pb in men compared with women. Mercury concentrations also increased with age, suggesting that older people from these communities might be consuming higher amounts of fish and marine mammals than younger people.

The major source of POPs exposure for people in the Russian Arctic is the contamination of traditional food items, including marine mammals, from global and local sources. POPs concentrations in blood from mothers in Chukotka, Russia decreased between 2001–2003 and 2007. For example, *p,p'*-DDE levels in blood have decreased in mothers from Chukotka and women and men from Nelmin-Nos.

Concentrations of metals declined in pregnant women in northern Norway from 1996 to 2008, but marine food intake may also have declined. The concentrations are considered relatively low and not of clinical importance for women of childbearing age and their children in northern Norway.

Parity and breastfeeding were associated with lower body burdens of PFASs in Norwegian women. Consumption of more marine food was associated with increased levels of PFOS, PFNA, PFDA and PFUnDA, while the consumption of beef was associated with increased levels of PFOA. Consumption of a large amount of game was associated with increased levels of PFHxS, PFHpS and PFNA. Seven of ten PFASs were measured in more than 80% of women during 2007–2009, suggesting that ongoing monitoring of these compounds is warranted to determine long-term trends for PFASs in pregnant women following the decline in production and use of these compounds.

Long-term studies of a specific human population cohort show evidence of the positive effect of international regulations and bans on the production and use of chemicals that end up in

the environment: decreased exposure over the years since regulation are reflected in decreased concentrations of these contaminants in human blood. Such decrease is not seen for unregulated environmental contaminants.

The impact of new and emerging contaminants on human health in the Arctic needs further research. These contaminants will, to some extent, have a different usage, distribution pattern and fate compared to legacy POPs. Increased human activity in the Arctic, such as shipping, industries and tourism, will create local sources of pollutants that may affect human health.

Further investigations are needed on the influence of diet (especially from marine food items) on the levels of legacy and emerging contaminants in humans, and assessments of health outcomes in relation to known and predicted contaminant levels.

7.4 Attribution of sources of environmental contaminants

Increasing population and further development of industrial activities in the Arctic in the coming years will increase local pollution, which is additional to the long-range transported contaminants. Local sources of pollution in Fennoscandia and the European Arctic need to be evaluated (in addition to contaminants arriving by long-range transport) because pollutants in power plant emissions and municipal/industrial wastewater discharges directly affect local populations, often in a more direct way than long-range pollutants. The environmental impact of local sources needs to be evaluated and understood. Measurements relevant to sustainable development of Arctic societies should be retained where they are ongoing and implemented where needed.

Characterization of atmospheric particles can be used to differentiate between local, regional and long-range transported environmental contaminants, particularly metals and organic pollutants that possess a high molecular weight. With the increasing number of local sources of emerging contaminants in the Arctic, this measure can provide important information about the sources of pollution.

Rapid changes in particle composition for the fine and coarse fractions immediately after high-temperature release from the smokestack have been identified. Condensation processes have been found to be mainly responsible for the rapid changes observed in composition. These changes, however, have considerable consequences for the particle association (absorption and adsorption) of combustion-derived organic pollutants, namely, PAHs, to these atmospheric particles. Distinct differences in PAH distribution profiles were found between samples obtained near emission sources and those collected far away. These distinct profiles may be used for characterizing sources and distinguishing between long-range transport and nearby source emissions in Arctic environments.

This project concerns several 'legacy' contaminants; that is, older chemicals that are either banned or highly regulated under the Stockholm Convention on Persistent Organic Pollutants. However, there are many unregulated or newer ('emerging') contaminants that are now being detected in Arctic ecosystems. Concentrations and the bioaccumulation potential of emerging contaminants need further investigation in the Arctic environment and biota. The impact of potential local sources should also be evaluated.

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Annex: Summaries of theses supported by this project

Selected perfluorinated compounds in the sediment of an Arctic freshwater lake: Case study at Kapp Linnè

Stig M. Lunde, 2016. Summary of the Master of Science thesis; Norwegian University of Life Sciences (NMBU), University Centre in Svalbard (UNIS)

Perfluorinated alkylated substances (PFASs) were found in Arctic char (*Salvelinus alpinus*) from Lake Linnévatnet, Svalbard and were reported in the Master of Science thesis of Maren Garsjø (2013). Short-chained PFASs are not known to bioaccumulate, and their elevated concentrations in Arctic char might therefore be caused by high concentrations in the sediment.

The aim of this thesis was to determine the concentrations of PFASs in the sediment of Lake Linnévatnet, with a focus on short-chained PFASs. Possible point-source locations on Svalbard were investigated and discussed for comparison. The field work was conducted in June 2015. Samples were extracted and cleaned by the same method that was applied in the work of Maren Garsjø, although modified for sediment instead of fish tissue. The laboratory work was conducted at UNIS and NMBU. Samples were analyzed using high-performance liquid chromatography and tandem mass spectrometry (HPLC-MS/MS).

Selected perfluorinated compounds in the water of an Arctic freshwater lake: Case study at Kapp Linnè

Jøran Solnes Skaar, 2016. Summary of the Master of Science thesis; Norwegian University of Life Sciences (NMBU), University Centre in Svalbard (UNIS)

Based on the results of the MSc thesis of Maren Garsjø in 2013, high levels of certain short-chained perfluorinated compounds were found in Arctic char (*Salvelinus alpinus*). These compounds are not known to bioaccumulate, and high concentrations in the fish might indicate high concentrations in the water as well. The aim of this thesis was to determine the levels of perfluorinated compounds in the water of Lake Linnévatnet with an emphasis on the short-chained compounds, and to examine possible point-source locations on Svalbard for comparison. Sediment and water samples were collected during fieldwork in April and June 2015 at Kapp Linnè. The samples were processed and extracted using solid-phase extraction (SPE) at UNIS during autumn 2015. Water samples were also taken from Lake Linnévatnet in March 2014, and were included in this MSc work and analyzed at NMBU using high-performance liquid chromatography and tandem mass spectrometry (HPLC-MS/MS).

A quantitative analysis of organochlorine pesticides in Svalbard reindeer (*Rangifer tarandus platyrhynchus*)

Kirsten Husby Melien, 2014. Summary of the Master of Science thesis; Norwegian University of Life Sciences (NMBU), University Centre in Svalbard (UNIS)

This study focused on the analytical challenges of analyzing selected organochlorine pesticides in muscle, liver, and fat of Svalbard reindeer (*Rangifer tarandus platyrhynchus*) in Svalbard, Norwegian Arctic. A brief comparison with Svalbard rock ptarmigan (*Lagopus muta hyperborea*) and pink-footed goose (*Anser brachyrhynchus*) is also included. All samples were collected by local hunters and stored frozen until they were cleaned-up by cold column extraction, acid treatment, and silica gel, before gas chromatography - mass spectrometry (GC-MS) analyses. Hexachlorocyclohexene (HCB) was quantified in most samples and the majority of the results were lower than in other studies, ranging from 0.1 to 1.3 ng/g ww for muscle, 1.7 to 7.0 ng/g ww for fat, and one result of 10.2 ng/g ww for liver in Svalbard reindeer. At even lower values, *p,p'*-DDE was quantified in three samples only, ranging from 0.1 to 1.8 ng/g ww. Other target pesticides (e.g., aldrin, chlordanes, *p,p'*-DDT and *o,p'*-DDT) were below the quantification limits. Results from this study have a degree of uncertainty as the linear range of the internal standard and the quantification standard had concentrations that were too low and the recovery standard did not demonstrate optimal performance. Because pesticides are a long-term problem for humans, wildlife and the environment in the Arctic, it is important to conduct continuous monitoring of wildlife to understand the sources and pathways causing change, whether increases or decreases, in legacy POPs, as well as detection of new and emerging compounds. This study targeted a small geographical area around Longyearbyen in Svalbard, and all samples were from the same year. Although such research cannot alone show temporal and spatial trends, results can be compared with those of other studies to obtain broader knowledge of organochlorine pesticides in Arctic terrestrial herbivores.

Perfluoroalkyl substances in Arctic birds – A comparison between glaucous gulls and black guillemots from Svalbard

Siri Axelson, 2014. Summary of the Bachelor of Science thesis; Swedish University of Agricultural Sciences (SLU), University Centre in Svalbard (UNIS)

Perfluoroalkyl substances (PFASs) are ubiquitous in the environment and have even been detected in remote areas such as the Arctic because they undergo long-range transport via the atmosphere and ocean currents. PFASs may be harmful to organisms owing to their persistence in the environment, bioaccumulation potential and toxicity. Studies have shown that PFASs can cause adverse effects on metabolism as well as on endocrine and reproductive systems. In this study, PFASs were investigated in glaucous gull (*Larus hyperboreus*) and

black guillemot (*Cephus grylle*) from Svalbard. Mean Σ PFAS concentrations were 147 ng/g ww in liver and 15 ng/g ww in muscle from glaucous gulls. In black guillemot, mean Σ PFAS concentrations were 36 ng/g ww in liver and 2.5 ng/g ww in muscle. Perfluorooctanoic sulfonate (PFOS) was the most abundant compound and contributed on average 72% of the Σ PFAS. The mean PFOS concentration was more than four times higher in glaucous gull than in black guillemot. This can be explained by the glaucous gull's high trophic level, migration patterns to more industrialized areas, and omnivorous feeding patterns. Concentrations were higher in liver than in muscle for both species, which is in accordance with other studies.

As PFASs are produced in industrial areas far from Svalbard, the concentrations detected in the species studied give reason to further investigate the fate and transport of PFASs, as well as their effects on wildlife in the Arctic region.

Handling and storage of the samples to avoid contamination is very important for PFAS analyses. In this study, several PFASs were detected at low levels in the laboratory blanks. Nevertheless, the PFAS concentrations in the samples could be appropriately quantified. Generally, the recoveries in this study were very good (mean: 83%).

There was a large concentration variation between individual organisms in this study, but the number of samples was too small to identify these variations as statistically significant or natural variations. Moreover, the age and sex of the birds in this study were unknown and these factors might influence PFAS levels. The time frame of this study was limited and for future studies a larger number of samples should be analyzed and more information about the age and sex of the birds investigated should also be provided.

Perfluorinated alkylated substances (PFAS) in Arctic char (*Salvelinus alpinus*): A case study from Svalbard

Maren Garsjø, 2013. Summary of the Master of Science thesis; Norwegian University for Life Sciences (NMBU), University Centre in Svalbard (UNIS).

Perfluoroalkylated substances (PFASs) have been used globally and are persistent enough to be transported to the Arctic. Owing to their toxicity, long-range transport potential and potential for bioaccumulation, PFASs are of concern for Arctic wildlife and humans living in Arctic areas. Because the Arctic is not expected to be a source, the main contribution of PFASs comes from long-range transport via ocean currents and atmospheric transport from lower latitudes. However, there are some local sources of PFASs in the Arctic which might contribute to the total PFAS load there.

The aim of the study was to increase scientific knowledge about PFAS levels and distribution in Arctic char (*Salvelinus alpinus*) from Lake Linnévatnet in western Svalbard, with regard to the potential exposure route for human consumers of these fish. In most of the High Arctic, Arctic char is the only resident

fish species in freshwaters, and it was therefore chosen as a biomonitoring species in this project. Arctic char were caught in March 2013 and September 2010. Analyses of liver and muscle samples were conducted at the Norwegian Institute of Air Research (NILU) in Tromsø. Principal component analysis (PCA) was used to examine the relationship between liver and muscle samples and PFAS.

The results showed that perfluorobutanoic acid (PFBA) and perfluorohexanoic acid (PFHxA) were the most abundant PFAS compounds detected in all samples. In 2010, PFBA in muscle samples showed the highest concentration (8.3 ng/g ww). The fluorotelomer 6:2 FTS showed the highest concentration (5.3 ng/g ww) in a liver sample from 2013. The short-chained PFAS compounds (four to six carbons) contributed 78% of the Σ PFAS in the 2010 sample measurements, while the long-chained PFAS (seven to 13 carbons) together accounted for 56.8% of the Σ PFAS in 2013 samples. In general, the concentrations were relatively low. There was a decrease in levels of PFASs in muscle samples from 2010 to 2013, while the highest levels were detected mainly in liver samples from 2013. This is consistent with previous studies indicating that PFAS binds to proteins in the blood and bioaccumulates in the liver. Because of the location of Lake Linnévatnet and the low concentrations of PFASs, long-range atmospheric transport is the main pathway for these contaminants instead of local sources. Based on previous studies, perfluorooctanoic sulfonate (PFOS) and perfluorooctanoic acid (PFOA) were expected to have higher concentrations and to be the dominating contaminants, but this was not the case in the present study. This indicates that temporal trends and spatial distribution of PFASs should be continually monitored. The present study is, to our knowledge, the first to report levels of PFASs in freshwater fish in Svalbard.

Organic pollutant and particle characterization from primary atmospheric emission sources in the Arctic

Katharina Schütze, 2013. Summary of Diploma thesis; Norwegian University for Life Sciences (NMBU), University Centre in Svalbard (UNIS), Technische Universität Darmstadt, Germany

Atmospheric particle samples were collected from two settlements (Longyearbyen and Barentsburg) on Svalbard. The samples were analyzed for PCBs and PAHs and the composition of the particles was characterized. This thesis presents concentrations as well as fingerprints of PCBs and PAHs originating from the power plant in Longyearbyen and other local sources.

The concentrations of PAHs in Longyearbyen town center (663 ng/m³) were comparable to concentrations on the European mainland, which can be explained by heavy snowmobile traffic during the sampling period (March 2012). Five kilometres east of Longyearbyen, the levels had decreased but were still comparable to PAH concentrations measured in ambient air samples found, for example, in Stockholm, Sweden.

A fingerprint of the PAHs in the exhaust of the coal power plant in Longyearbyen was also assessed. The dominant PAHs emitted were indeno[1,2,3]pyrene, benzo[ghi]fluoranthene, benzo[bjk]fluoranthene, benzo[a]fluoranthene, benzo[e]pyrene and benzo[a]pyrene. This PAH fingerprint was not completely reflected at the sampling locations in the settlement owing to wind direction and other local sources (e.g., traffic) present.

PCBs were only detected at a few locations. The range in Σ PCB concentration at those locations was 170–673 pg/m³. The highest concentration was measured at the power plant in Longyearbyen.

Different sources influencing the contaminant pattern in the settlements can be assessed by characterization of particles. The anthropogenic fraction of the emitted particles $\geq 0.5 \mu\text{m}$ was mainly aluminum silicates and iron fly ashes, often found to be covered by a layer of sodium sulfate. Hence, these particles originated from the coal combustion process of the power plant. A further investigation by transmission electron microscope (TEM) revealed differences in heavy metals as heterogeneous inclusion internally mixed with secondary aerosols and soot. Lead was found at all sample sites, especially in the plume of the power plant. Hence, lead occurring as heterogeneous inclusion internally mixed with soot can be regarded as originating from coal combustion at the power plant. At UNIS, other heavy metals such as zinc, zirconium, barium, manganese and titanium were also observed that had not originated from the power plant.

The sample sites differed with regard to their concentrations of PAHs and PCBs, as well as in the particle types observed. The PAH pattern and heavy metals associated with particles found

in the fine particle fraction might be a very good indicator for source assessment. Because the further analysis of very small constituents of particles on the fine fraction with TEM exceeded the frame of this thesis, further analyses should be carried out to characterize emissions and pollution from different local sources in the sensitive Arctic environment on Svalbard.

Thesis links

Garsjø, M. Perfluorinated Alkylated Substances (PFAS) in Arctic char (*Salvelinus alpinus*): a case study from Svalbard. Master thesis. Issue date: 2014-02-03. Norwegian University of Life Sciences, Ås, Norway.
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Lunde, S. M. Selected perfluorinated compounds in the sediment of an arctic freshwater lake: A case study at Kapp Linnè. Master thesis. Issue date: 2016-08-10. Norwegian University of Life Sciences, Ås, Norway.
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Melien, K. H. A quantitative analysis of organochlorine pesticides in Svalbard reindeer (*Rangifer tarandus platyrhynchus*). Master thesis. Issue date: 2015-02-27. Norwegian University of Life Sciences, Ås, Norway.
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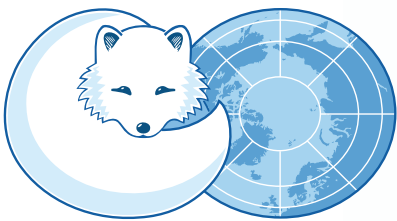
Skaar, J. S. Occurrence of selected poly- and perfluoroalkyl substances (PFAS) in Arctic freshwater: a case study from Svalbard. Master thesis. Issue date: 2016-08-10. Norwegian University of Life Sciences, Ås, Norway.
Permanent link: <http://hdl.handle.net/11250/2398613>

Acronyms and Abbreviations

6:2 FTSA	6:2 Fluorotelomer sulfonic acid
AMAP	Arctic Monitoring and Assessment Programme
AMDE	Atmospheric mercury depletion event
ArcRisk	Arctic Health Risks: Impacts on health in the Arctic and Europe owing to climate-induced changes in contaminant cycling
Cd	Cadmium
DDD	Dichlorodiphenyldichloroethane
<i>o,p'</i> -DDD	1,1-dichloro-2-(2-chlorophenyl)-2-(4-chlorophenyl)ethane
<i>p,p'</i> -DDD	1,1-dichloro-2,2-bis-(4-chlorophenyl)ethane
DDE	Dichlorodiphenyldichloroethylene
<i>o,p'</i> -DDE	1,1-dichloro-2-(2-chlorophenyl)-2-(4-chlorophenyl)ethene
<i>p,p'</i> -DDE	1,1-dichloro-2,2-bis-(4-chlorophenyl)ethene
DDT	Dichlorodiphenyltrichloroethane
<i>o,p'</i> -DDT	1,1,1-trichloro-2-(2-chlorophenyl)-2-(4-chlorophenyl)ethane
<i>p,p'</i> -DDT	1,1,1-trichloro-2,2-bis-(4-chlorophenyl)ethane
DEHM	Danish Eulerian Hemispheric Model
ECMWF	European Centre for Medium-Range Weather Forecasts
EMEP	European Monitoring and Evaluation Programme
FOSA	Perfluorooctane sulfonamide
GEM	Gaseous elemental mercury
GOM	Gaseous oxidized mercury
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
Hg	Mercury
IPCC	Intergovernmental Panel on Climate Change
LOD	Limit of detection
MeHg	Methylmercury
MISA	The Northern Norway mother-and-child contaminant cohort study
MM5v3.7	Fifth-generation mesoscale model
NCEP	National Centers for Environmental Prediction
PAH	Polycyclic aromatic hydrocarbon
Pb	Lead

PBDE	Polybrominated diphenyl ether
PCB	Polychlorinated biphenyl
PFAS	Perfluorinated alkylated substance
PFCA	Perfluoroalkylated carbon acid
PFDA	Perfluorodecanoic acid
PFDoA	Perfluorododecanoic acid
PFHpA	Perfluoroheptanoic acid
PFHpS	Perfluoroheptane sulfonate
PFHxS	Perfluorohexane sulfonate
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFTTrA	Perfluorotridecanoic acid
PFUnA	Perfluoroundecanoic acid
PFUnDA	Perfluoroundecanoate
POC	Particulate organic carbon
POP	Persistent organic pollutant
SRES	Special report on emissions scenarios (IPCC)
TMF	Trophic magnification factor
ww	Wet weight

AMAP



ARCTIC COUNCIL

AMAP
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