Arctic Health Risks: Impacts on health in the Arctic and Europe owing to climate-induced changes in contaminant cycling (ArcRisk)

Executive Summary

Global climate change has the potential to remobilize environmental contaminants and alter contaminant transport pathways, fate, and routes of exposure in human populations. The ArcRisk project, with a consortium of 21 partners, studied this potential in relation to the long-range transport of contaminants to the Arctic, transfer processes of contaminants between Arctic environmental compartments and into food webs, and contaminant exposure and its effects on human health in the Arctic, in comparison with other exposed populations in Europe.

The main objectives of the ArcRisk project were to describe and quantify to the extent possible: 1) the transport routes of selected groups of contaminants to and within the Arctic and the potential influence of climate change; 2) environmental transfer processes of contaminants, their uptake into food chains and transfer to organisms consumed by humans, and possible influence of climate change; and 3) health outcomes related to exposure to contaminants in selected Arctic populations and exposed local populations in the EU and the potential influence of climate change on future contaminant exposures.

ArcRisk used modelling tools to study the transport of several persistent organic pollutants (POPs) and mercury to the Arctic via the atmosphere and oceanic currents under both present climate and projected future climate scenarios. Models were used to compare the influence of climate change on contaminant cycling in the Arctic as well as European regions, with a particular assessment of climate influence on contaminant transport and fate in the Baltic Sea region for comparison. A bioaccumulation model describing the food-web transfer of organic contaminants in an Arctic marine food web was developed to determine the possible role of climate change on future dietary exposure to pollutants.

Field/laboratory work in the Arctic included studies on: 1) contaminant deposition and behavior in snow and subsequently during melt periods; 2) pesticides in a west Greenland fjord affected by glacier meltwater; 3) input of PCBs via major Arctic-draining rivers and PCB loads in coastal Arctic seas; and 4) contaminant levels in seawater/ice/snow and fluxes from sea ice to water near Svalbard. A wide range of biota including marine invertebrates, fish, and reindeer from the Svalbard area and coastal areas of northern Norway and market-basket food samples from Greenland were analysed for organic contaminants to determine dietary levels for consumers.

Contaminant levels and health outcomes were studied in several human cohorts, mainly mothers and children, in the Arctic and in Mediterranean areas. A database was developed containing results of studies of contaminant levels and health outcomes in people in the Arctic and these results were combined with a review of contemporary literature to investigate the association between exposures to various POPs a number of health outcomes. A study was made of dietary sources of environmental contaminants and geographical differences in human exposures.

Case studies were prepared reviewing PCBs and Hg from emissions to potential health effects. Key findings and recommendations arising from the scientific work have been compiled in a synthesis report and in a web-based legacy ódatabaseô containing an overview of project results. Many scientific papers have been published, with more still in preparation. A glossy, layman-style overview of the results for policy-makers and the public was published and distributed at the final conference, which was held at Arctic Frontiers 2014 in Tromsø, Norway in January 2014, with the full range of scientific results presented in oral presentations and posters.

Summary of the context and main objectives of the ArcRisk project

All living organisms including human beings are exposed to a number of chemical contaminants that persist in the environment for long periods of time. Some chemicals, such as mercury, occur naturally in the environment, but are also mobilized by or used in human activities. However, most of these chemicals have been manufactured for various purposes, such as for a large variety of industrial and household products, flame retardants, pharmaceuticals, insecticides and other pest control chemicals, or they occur as by-products in the manufacturing process. Environmental contaminants are chemicals that enter the environment as a result of intentional use (such as pesticides) or unintentional releases (such as dioxins released during incomplete combustion of wastes). Chemicals may enter the environment through poorly controlled emissions during manufacture, during their ordinary use, and from poorly controlled waste disposal conditions. Although many of these chemicals serve a useful purpose in their original uses, their subsequent dispersion and accumulation in the environment is highly undesirable, and has the potential to negatively affect the health of humans and ecosystems. In some cases, exposure to such environmental chemicals is high enough to raise concerns about possible effects on human health. Climate change has the potential to alter the pathways by which these harmful chemicals cycle through the environment and enter food chains.

Following their release, environmental contaminants persist in the environment, either in their original form or as breakdown products; they degrade to harmless chemical forms only very slowly. Concern is greatest for chemicals that as well as being environmentally persistent also find their way into food chains (i.e., they bioaccumulate) and are toxic. These characteristics give rise to abbreviations that are commonly applied to groups of these chemicals: PTS (persistent toxic substances) and POPs (persistent organic pollutants).

Owing to their persistence, many of these chemicals are widely dispersed around the globe by winds and ocean currents. The chemicals used in high volumes are found almost everywhere, but as the properties of individual compounds differ so does their ćpartitioningĉ. This means whether the chemicals in the environment associate with air, soil and sediments or water, and in animals whether they accumulate in particular tissues and organs (many POPs concentrate in fatty tissues).

Although substantial research has been carried out on the mechanisms and effects of environmental contaminants, it is still unclear to what extent and/or in what situations and population sub-groups environmental contaminants may represent a significant, long-term health risk, particularly for populations living in the cold northerly fringes of Europe, where the cold conditions combined with a traditional diet of fish and marine mammals serve to enhance human exposure to contaminants.

The Arctic is contaminated by chemicals released as a result of both human activities and natural processes. Despite the fact that most of these substances have few, if any, sources within the region, these chemicals reach the Arctic from distant sources via the atmosphere and via northerly flowing rivers and ocean currents.

Although no longer pristine, Arctic abiotic environments generally exhibit lower levels of contamination than those found in regions closer to major sources, such as most of Europe. However, certain characteristics of the Arctic (cold, ice and snow cover, extended periods of darkness) mean that the Arctic has the potential to accumulate certain globally transported environmental contaminants, including a number of persistent organic pollutants (POPs) and mercury. Processes that affect contaminant movement within and between different environmental compartmentsð and thereby determine to a large degree their transport pathwaysð are complex. These processes and pathways are influenced by a number of physical factors, such as temperature, precipitation, winds, ocean currents, and snow and ice cover, all of which are subject to climate-related perturbations.

At the same time, the Arctic possesses unique food webs, including a number of key species that utilize fat reserves for energy storage and insulation. Throughout history, Arctic human populations have relied on these natural resources for foods that form the basis of their ótraditional dietsô This combination of factors has resulted in a paradox whereby, despite generally low levels of contamination in air, soils or water, Arctic species high in the food chainsð polar bears, seals and toothed whales, certain seabirds, and predatory fishð accumulate very high concentrations of certain contaminants. As these species also form part of the traditional diet of Arctic peoples, certain Arctic populations receive greater exposures to specific contaminants than people anywhere else on the Earth. Many of these contaminants are toxic, with known potential to adversely affect the health of animals and humans. In some cases, evidence exists of direct health effects resulting from exposure to contaminants in general (i.e., non-occupationally exposed) in population groups in some Arctic countries. As with the physical factors that influence contaminant transport pathways, ecosystems are also subject to climate-related perturbations that can alter food webs through a range of mechanisms and thus alter contaminant exposure in various parts of the system.

Given the particular sensitivity of the Arctic to climate change and the clear impacts of climate change that are already being observed, the ArcRisk project was established to investigate the potential climate-mediated changes that may occur in the long-range transport of pollutants, including air pollutants, to the Arctic from regional and global sources and the ways in which climate variability and global climate change will affect the mobilization and transfer of pollutants in the Arctic environment and their uptake into living organisms, particularly those in food webs leading to species consumed by humans. These potential climate-induced changes in bioaccumulation and bioavailability of contaminants in human food species, such as fish, marine mammals, and reindeer in the Arctic, further may affect the health of their human consumers, in addition to the other potential health impacts of climate change. To provide a broader perspective, comparisons were also made with the potential impact of climate change on contaminant exposure and effects on human health in several areas of Europe with exposed local populations.

Accordingly, in the ArcRisk project, the influence of climate change on contaminant transport and the resultant risk to human populations in the Arctic and other areas of Europe were studied with special focus on:

1) the ways in which climate change will affect the long-range transport and fate of selected groups of contaminants, and possible implications for the re-distribution of contaminants (geographically and between relevant environmental media), involving modelling utilizing the existing information base on the distribution of relevant contaminants in the Arctic and other areas of Europe;

2) the impacts that climate change will have on contaminant transfer and fate in aquatic and terrestrial environments and on contaminant uptake and transfer within food webs, leading to foods consumed by humans, as determined in experimental work and process studies;

3) evaluating present indications of human health outcomes resulting from dietary and other exposure to environmental contaminants in Arctic populations and exposed populations in selected areas of Europe, as based on meta-data analysis of large numbers of health studies and several relevant cohort studies, and determining how climate-mediated changes in the environmental fate of selected groups of contaminants will result in changes in exposure of human populations, in the Arctic and in selected areas of Europe, and the implications of this for the direct and indirect effects of contaminants on human health.

The main objectives of ArcRisk were to:

- Use models to describe, using selected climate change and chemical usage scenarios, the changing routes and mechanisms by which persistent chemical contaminants are delivered to the Arctic, including via long-range atmospheric transport, ocean currents, and Arctic-flowing rivers, and the possible role of climate variability and global climate change on the processes influencing pollutant transfer and distribution;
- Quantify the deposition and accumulation of selected persistent chemical contaminants on snow/ice and on ice-free surfaces, their geochemical fate in the seasonal snow pack, and their transfer to aquatic food chains with melt-water runoff;
- Determine the transfer of contaminants from the abiotic Arctic environment into the base of food chains and to higher trophic level organisms (e.g., fish, marine mammals, reindeer) consumed by humans and the possible role of climate variability and global climate change on these processes, including bioaccumulation of select contaminant groups in specific food webs and organisms relevant to human diet;
- Compare the role of climate change on the transport, fate and food-web transfer of contaminants in the Arctic to the situation in relevant selected areas with exposed local populations in the EU;
- Through the development of an extensive database containing comprehensive data from recent and on-going assessments regarding health outcomes in relation to contaminant exposure, combined with data from publications and on-going research projects in the Arctic, identify and quantify the current main health outcomes in relation to exposure to ólegacyô contaminants in selected populations in the Arctic and exposed local populations in the EU;
- Based on this database and the results of recent and on-going cohort studies of health outcomes in relation to contaminant exposure, establish geographical and temporal trends in the distribution of health impacts associated with contaminants in several areas of the Arctic and selected areas of Europe;
- Develop projections for the effects of climate change on the ólegacyô contaminant exposure and health outcomes in the selected populations in the Arctic and in exposed local populations in the EU;
- Develop a legacy project results database containing a summary and synthesis of information resulting from the project on the influence of climate change and climate variability on emissions and transport of selected contaminants, and exposure of humans to these contaminants that can be used by policy-makers, scientists, and the public.

Describing the full sequence from emissions to exposure for contaminants with varying chemical properties is extremely complex and currently available scientific tools and methods do not allow detailed quantification of all the steps involved nor of the potential influence of all aspects of climate change on these steps. The ArcRisk research was thus designed to investigate a number of potentially important processes via field experiments and to perform model simulations for climate change scenarios for selected compounds using a variety of modelling tools. For most contaminants, the information needed to follow this step-wise approach was lacking. Sufficient information was, however, available for PCBs and mercury, and they were therefore chosen for the preparation of case studies to illustrate how climate change can be expected to influence contaminant behavior, fate and human health impacts. In these case studies, literature reviews and results from other national and international research activities were used as sources of information, along with the results of ArcRisk activities focusing on these specific contaminants.

An overarching goal of the ArcRisk program was to generate scientific results that are relevant to policies dealing with contaminants and health on European, pan-Arctic and global scales and to convey scientific information to policy-makers, stakeholders and other interested parties about how climate change might alter our exposure to those environmental contaminants that can affect human health. This was based on a policy evaluation of EU policies and directives relevant to risk assessment and human exposure to environmental contaminants as well as policies and international agreements whereby contaminants including POPs are controlled owing to their potential for long-range atmospheric transport. Wide communication of the results of the project was also provided, primarily through regional and international scientific conferences but also at the national and university level, and a large number of peer-reviewed scientific papers have been published.

Main Scientific Results of the ArcRisk project

1 Introduction

A core set of substances was selected for study in all parts of the ArcRisk project based on the existence of an adequate amount of high-quality scientific data available on their emissions and presence in the environment, their significance as environmental contaminants and in relation to human health, and the expertise of the partner institutes. The main substances chosen were dichlorodiphenyltrichloroethane and related chemicals (DDTs), polychlorinated biphenyls (PCBs), hexachlorocyclohexanes (HCHs), perfluoroalkyl substances (PFASs) and mercury, although other contaminants were also considered, particularly in relation to human health.

With a few exceptions, the modelling work in ArcRisk that addressed climate scenarios was carried out using the IPCC Fourth Assessment Report (AR4) Special Report on Emissions Scenario (SRES) A1B climate scenario and, when output from an external climate model was needed, the ECHAM5/MPI-OM and/or CCSM models were selected, as recommended in ArcRisk Deliverable D7. A database of physical-chemical properties and emission estimates of chemicals was prepared as Deliverable D11; however, in many cases, the emission estimates were not entirely suitable for the modelling applications and were enhanced or augmented later in the project, as needed for each modelling task.

2 Overall results of ArcRisk modelling work

The effect of climate change on the fate and transport of organic contaminants was investigated using a range of modelling tools. All modelling tools showed that climate change can affect future levels of concentrations of organic contaminants (both increase and decrease) if compared to baseline óno climate changeô scenarios, with projected changes around a factor of two, i.e., by 2100 concentrations are projected to be half to double those in runs with no climate change. The models typically consider the effects of climate change (in varying degrees of sophistication) on temperature and precipitation, as well as on air and ocean currents. Changes in organic carbon cycling, catchment hydrology, land use, extreme events (which are on the rise in frequency) and ice cover, which are more complex to model and have important effects on contaminant levels, were often not considered or modelled in a simplified way. Increasing temperature under a future climate will cause a shift in environmental distribution, which causes an increasing level of some chemicals in air and decreasing concentrations in surface media. These increasing air concentrations are expected to be offset, however, by decreases due to expected reductions in the emissions of many chemicals. Changes in sea-ice cover and thickness have also been modelled in the Arctic Ocean. A shift from multi-year sea ice to first-year sea ice under a future climate is expected to result in more efficient delivery of contaminants from melting ice into the underlying surface water and thus increase contamination in the surface water of the Arctic Ocean.

The effect of climate change on the atmospheric deposition of mercury in the Arctic was investigated in a Danish modelling study. Under climate change scenarios, there was a significant decrease of mercury deposition over the Arctic Ocean and a significant increase over the continents. The main reason for these changes in deposition patterns is the decreasing ice cover leading to decreased mercury oxidation in the marine boundary layer over the Arctic Ocean and increases in the ozone concentrations in the troposphere over the continents leading to increased mercury oxidation.

The comparison with the Baltic region showed that the effect of climate change on the fate and transport of organic contaminants in this region was similar to other regions previously modelled, including the Arctic (i.e., projected changes around a factor of two in either direction). The state-of-the-art contaminant fate modelling tool for the Baltic region, POPCYCLING-Baltic, was identified as being overly simplistic to accurately model the effects of climate change on the fate and transport of organic contaminants in this region.

A food-web model was developed to simulate the bioaccumulation of selected PCB congeners in a marine food web located near Svalbard. The model was evaluated with monitoring data for the food web collected within ArcRisk field studies and the model was shown to largely under-predict the concentrations of PCBs in the food web. There are, therefore, problems with our ability to estimate food-web bioaccumulation in Arctic marine food webs with state-of-the-art modelling tools under existing climate conditions. Even if modelling tools can be developed to reliably predict bioaccumulation in Arctic marine food webs, the further challenge of estimating how climate change will affect bioaccumulation is limited by the lack of understanding of the effects of climate change on primary production, speciesô distributions and trophic interactions.

2.1 Global and Arctic abiotic modelling and the influence of climate change

The global distribution and fate of *p,p'*-DDT was studied for 1950 to 2100 using the MPI-MCTM under a model-generated climate that follows the SRES A1B scenario. The results emphasize the increasing role of the ocean in the large-scale distribution and fate of DDT. Volatilization is the main sink for DDT in surface seawater, and because of the earlier peak of emissions, DDT export to the deep sea is ahead of that of PCBs.

The fate and transport of PCBs in the global environment was studied using three models: MPI-MCTM, BETR Research, and DEHM. The models are in good qualitative agreement with regard to the expected impacts of climate change on PCB concentrations in the Arctic. For low-chlorinated PCBs, modelled concentrations in Arctic air are only slightly sensitive to climate change, while for mid- and high-chlorinated PCBs higher concentrations are expected under a climate change scenario. Concentrations of PCBs in Arctic Ocean water are also higher under a climate change scenario in BETR Research and MPI-MCTM results that considered the changes in PCB emissions over their product life cycle. In model experiments with DEHM that assumed the same emissions but different climate conditions, modelled concentrations of PCBs in Arctic Ocean water were lower under the climate change scenario.

HCHs were modelled using BETR Research and DEHM. The BETR Research modelling considered the life cycle of use of HCHs from 1945 to 2050, and evaluated model results against measurements in air and ocean water in the period 1985 to 2010. Modelled concentrations of HCHs in the Arctic were found to be only slightly sensitive to recent climate variability, and forecast concentrations in Arctic air and ocean water were lower by factors of two and five, respectively, under a climate change scenario compared to present-day climate. In DEHM model experiments that assumed the same emissions under two climate scenarios, concentrations of HCHs were higher in atmosphere of the Northern Hemisphere under a climate change scenario.

Oceanic transport of perfluorooctanoic acid (PFOA) to and within the Arctic Ocean over the period 1950 to 2010 was modelled using NAOSIM and atmospheric transport and oceanic transport were modelled using MPI-MCTM. Modelled concentrations of PFOA in the Arctic Ocean show clear changes due to changes in ocean circulation that occurred during the mid-1990s and 2000s during a prolonged positive phase of the Arctic Oscillation. These model results for PFOA are very similar to the behaviour of iodine-129, a radionuclide tracer for which there is a considerable amount of empirical evidence consistent with model simulations.

Mercury modelling using DEHM indicates higher mercury deposition over continental areas in the Arctic and near-Arctic under a climate change scenario compared to present-day climate and lower deposition rates directly to the Arctic Ocean. The changes in mercury deposition are driven by changes in the atmospheric chemistry of mercury that result from reduced sea-ice cover and increased concentrations of ozone in the troposphere that are forecast under the climate change scenario.

2.2 Regional contaminant fate modelling in the Baltic region and influence of climate change

The regional multimedia chemical fate model POPCYCLING-Baltic was used to make estimates of climate change-induced effects on the environmental concentrations of persistent organic chemical contaminants in the Baltic Sea region. The focus was on óperfectly persistentô chemical contaminants that are not degraded in the environment. The property combinations studied here encompass the entire plausible range for non-ionizing organic substances.

The effects of single or multiple climate projections for temperature (T), wind speed (W), precipitation (P) and particulate organic carbon (POC) in the Baltic Sea water column on the environmental concentrations of persistent chemicals in the Baltic Sea region were investigated. Climate data for T, W, and P for two scenarios were extracted from the Rossby Centre coupled regional climate model which was driven with data from two general circulation models, HadAM3H and ECHAM4/OPYC3. The two climate scenarios used are the IPCC SRES A2 (2071-2100) and baseline (BL) climate scenarios (1961-1990). Three hypothetical and illustrative emissions scenarios were created to compare and contrast the climate change-induced effects on chemicals that are emitted in the Baltic Sea region to each of air, water and soil.

Media-specific concentration ratios were calculated to quantify the changes in modelled concentrations under the climate change scenario (A2; 2071-2100) versus the baseline scenario (BL; 1961-1990). Modelled concentration ratios between the SRES A2 climate change scenarios and the BL scenario ranged from factors of 0.5 up to 2.0 for chemicals with property combinations similar to the legacy persistent organic pollutants listed by the Stockholm Convention. These moderate effects of climate change on projected concentrations are similar to those predicted in the Arctic region. These property combinations cover most of the chemicals that have been selected as research chemicals for the ArcRisk project in Deliverable D3, including PCBs, HCHs and DDTs.

For many chemicals with different property ranges, either counteracting or reinforcing effects occurred when multiple climate parameters were considered simultaneously. These effects are dependent on the emission mode. Among the four variables investigated, temperature has the greatest influence on the predicted concentrations regardless of emission mode or climate change scenario. Changes in POC are generally the second most influential parameter affecting the fate of persistent organic chemicals in the climate scenarios used. Changes in precipitation have less influence on modelled chemical concentrations than temperature and POC, while wind speed exerts the least influence in comparison to the other three climate variables.

2.3 Contaminant food-web modelling and the influence of climate change

A steady-state fugacity-based marine food-web bioaccumulation model (the Svalbard Marine Food Web Model) was developed to simulate the bioaccumulation of selected PCB congeners. Field sampling of marine species was carried out in the Svalbard region in the summer of 2010. The samples collected were for amphipods *Onisimus glacialis* (OS) and *Apherusa glacialis* (AG), and polar cod

(*Boreogadus saida*, BS), which compose part of the Svalbard marine food web that has been modelled. Nine PCB congeners were regularly detected in the samples collected, and their concentrations were used to evaluate the food web model. Unfortunately, the seawater in the Svalbard region was not sampled and analysed for PCBs at the same time as the organisms were collected. To overcome this problem, the concentrations of selected PCB congeners in seawater were estimated based on measured data in 2001 in combination with modelled data in 2001 and 2010, assuming a constant ratio between modelled concentrations from these two years.

Sensitivity and uncertainty analysis identified the six most important parameters controlling variance in model-predicted concentrations in food-web species. The digestion factor is the most important parameter for the modelled PCB concentration in polar cod and *Onisimus glacialis*, which indicates high importance of biomagnification as a result of consumption of more lipid-rich food items relative to *Apherusa glacialis*. The water concentration of PCB congeners is generally the second most important parameter and is most important for *Apherusa glacialis* at the base of the food web. Water concentrations are important as they are the source of exposure for all three species. This is especially the case for the herbivorous *Apherusa glacialis*, which feeds entirely on algae whose concentration of PCB congeners is assumed to be in equilibrium with water. The octanol-water partition coefficient (K_{OW}) and the particulate organic carbon content in water (C_{POC}) are also important as they determine the bioavailable fraction of PCBs in water.

A comparison between modelled and measured PCB concentrations showed reasonable (within a factor of 10) agreement for *Apherusa glacialis*. However, for *Onisimus glacialis* the food-web model underestimates the concentration of PCB28 by a factor of up to about 180. Possible causes for underestimation include: (i) oversimplified parameterization of the food-web model, (ii) erroneously high concentration of PCB28 measured in *Onisimus glacialis*, (iii) erroneously low concentration of PCB28 measured in seawater in 2001, (iv) erroneous value for the particulate organic carbon content, and (v) low estimated concentration of PCB28 in seawater in 2010.

The food-web modelling work in ArcRisk has determined that there are problems with our ability to estimate food-web bioaccumulation in Arctic marine food webs with state-of-the-art modelling tools under existing climate conditions. A large limitation is the quality and quantity of field data needed for model evaluation. For example, in this study concentrations of PCBs in water were lacking for the food web studied. Future progress on estimating how climate change will affect the food-web transfer of persistent organic pollutants (POPs) is limited therefore by questions concerning the adequacy of present modelling tools for estimating PCB bioaccumulation under current climate conditions. It is recommended that more efforts be devoted to generate high quality datasets for model evaluation, in which multiple species and abiotic media are simultaneously and continuously sampled and analysed for PCBs. Even if modelling tools can reliably prediction Arctic bioaccumulation, the further challenge of estimating how climate change will affect future bioaccumulation is also not currently achievable due to deficits in current understanding of interactions between climate and ecology. In particular, an understanding of the effects of climate change on the distributions of species and trophic interactions is needed.

2.4 Comparison of modelled global climate change impacts in the Arctic and the Baltic Sea region

ArcRisk Deliverables D38 and D40 contain a more thorough report on the various modelling studies that have assessed selected chemicals including *p,p'*-DDT, PCBs, HCHs, per- and polyfluoroalkyl substances (PFASs), and mercury under climate change scenarios. The modelling study carried out for the Baltic Sea region assessed hypothetical persistent organic chemicals with a wide range of physical properties. This provided the possibility to conduct a comparison of modelling studies that took into account climate change and relevant organic pollutants in the Arctic and Baltic Sea regions.

For *p,p'*-DDT, the results based on the MPI-MCTM model suggested that oceanic waters in all tropical and subtropical regions, including the Mediterranean and the Baltic Sea regions, had become net-volatilizational with respect to *p,p'*-DDT around the year 2000. However, this was forecast not to occur in the Arctic Ocean until the end of the 21st century (ArcRisk Deliverable D38). The results suggest that global climate change (GCC) will lead to increases in the atmospheric concentration of *p,p'*-DDT regardless of emission mode. These increases are up to a factor of 1.6 in magnitude, and attributable to intensified volatilization from the Baltic Sea water. Unless emissions occurred to air, the Baltic Sea water is predicted to act always as a source. In summary, GCC will not play different roles in the Arctic and in the Baltic Sea region, i.e., GCC may intensify the volatilization of *p,p'*-DDT from both the Arctic and Baltic Sea waters. However, GCC may play an additional role for the Arctic region by redistributing more *p,p'*-DDT to the Arctic region.

For PCBs, the BETR Research, MPI-MCTM and POPCYCLING-Baltic models all suggest a similar role of GCC on the atmospheric concentrations of highly chlorinated PCBs. The model results all suggest increases in the atmospheric concentrations of these contaminants in the two regions. These increases are within a factor 1.8 and are attributable to intensified volatilization from surface compartments. The POPCYCLING-Baltic and DEHM models also predict decreases in the seawater concentrations of some highly chlorinated PCBs in the two regions. These decreases have been attributed to increased adsorption to POC and deposition to the sediment.

Regarding HCHs, model predictions suggest both increase and decrease in the atmospheric concentrations of three HCHs as a result of GCC. The factor of increase or decrease is up to 2.0. The concentrations of Ŭ- and ɓ-HCH in Arctic atmosphere and seawater are predicted to decrease by the BETR Research model. In the Baltic Sea region, the modelled GCC-induced effects are in general the same on the environmental concentrations of Ŭ-, ɓ- and ɔ-HCH, although Ŭ- and ɔ-HCH have different transport modes than ɓ-HCH. The long-term steady-state concentrations of the three HCHs in coastal and open-ocean water in the Baltic Sea are predicted always to decrease regardless of emission mode. In contrast, the atmospheric concentrations of the three HCHs will be increased due to climate change (except when ɓ-HCH was emitted to air; in this case, its concentration was predicted to decrease in the Baltic atmosphere). Therefore, GCC plays a similar role in the two regions with respect to the HCHs studied.

Oceanic and atmospheric transport of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) was simulated using the NAOSIM and MPI-MCTM models. Results suggested that changes in ocean circulation in the Arctic Ocean had caused changes in the surface seawater concentrations of PFOA. However, no attempt was made to quantitatively assess the GCC-induced changes in the transport and fate of PFOA and PFOS into and within the Arctic region. In the environment, PFOA and PFOS exist mainly as anions because they are very acidic, water soluble, and have very low vapour pressure. The results in ArcRisk indicate that the concentrations of PFOA and PFOS will increase in air, but decrease in water as a result of temperature-driven volatilization if emissions occurred to water and soil. These results may be questionable, however, because the volatilization of PFOA and PFOS should approximate to zero as a result of their ionizing nature.

With regard to mercury, based on the IPCC SRES A1B scenario, the DEHM model forecast decreases in the deposition of mercury over the Arctic Ocean and increased deposition over terrestrial areas in the

Arctic and near-Arctic. The predicted increases have been attributed to a significant increase in the ozone concentrations in the troposphere. The deposition of mercury could increase by a factor of up to 2.9 in the Baltic Sea region. The results based on the POPCYCLING-Baltic model cannot be extrapolated and compared for the two regions studied.

2.5 Summary of conclusions on modelling work

In other work, it has been suggested that the impact of GCC will vary with the environmental contaminant, organism, human population, and geographical region of interest. The comparisons presented here confirmed this finding. For example, modelling results suggest that in both the Arctic and Baltic Sea regions GCC will have a similar impact on the atmospheric concentrations of volatile chemicals, i.e., GCC will cause greater volatilization of volatile chemicals such as Ǔ- and γ-HCH and lower-chlorinated PCBs. Correspondingly, the concentrations of these volatile chemicals in water may decrease as a result of GCC-induced stronger volatilization. The limited number of studies suggests that GCC may have a similar effect on the bioaccumulation of organic contaminants in aquatic organisms in the two regions. For example, GCC could possibly cause decreases in bioaccumulation through causing decreases in bioavailable fractions of organic contaminants, and cause increases in growth dilution and digestive elimination.

These conclusions on the potential influence of GCC on the bioaccumulation of organic contaminants in aquatic food chains in the Arctic and Baltic Sea are subject to large uncertainties because they were based on a limited number of studies mostly focused on GCC-induced direct effects. Studies that have so far considered the effect of GCC on bioaccumulation have not considered, for example, indirect GCC effects on the structure of food webs. However, it is argued that GCC has influences on a large number of processes and variables which can play a role in bioaccumulation. Unless mechanistic models that can holistically account for the multitude of different processes are developed, it will be extremely difficult to assess the GCC-induced changes on bioaccumulation. Many important effects, especially indirect effects, induced by GCC were either not considered or oversimplified in the existing studies.

3 Results of ArcRisk field studies on processes of contaminant transfers in the environment and uptake into food webs

3.1 Transfer processes across the wider Arctic

The Arctic cryosphere is undergoing substantial change most notably through the reduction in summer sea ice and changes in glacier ice mass owing to increased warming. Predicted changes in weather patterns for different regions of the Arctic include altered precipitation and changes in the frequency of weather extremes. These perturbations are now borne out by observations and they will affect contaminant pathways both into and within the Arctic. Field work conducted in ArcRisk has established some of the baseline measurements with which to assess the effect of these perturbations on contaminant transfer, with particular focus on snow and ice for which contaminant measurements are sparse (ArcRisk Deliverable D43).

Work in both the European and Canadian Arctic has investigated the marine cryosphere with regard to its role in the storage and processing of chemical contaminants, with ice and snow serving as important links between the atmosphere and the upper ocean. Observations of PCBs, organochlorine (OC) pesticides and perfluoroalkyl substances (PFASs) in the marine environment have attempted to put the role of sea ice and associated snowpack into context with regard to the chemical load present in the surface Polar Mixed Layer (PML) of the Arctic Ocean. Scaling up of snow and sea-ice chemical loads (pg/m^2) (derived through snow/ice water equivalents) from field work conducted in the Canadian Arctic yielded loads of 42.1 ± 21.2 kg of Ŭ-HCH, 6.17 ± 3.34 kg Ɇ$_{10}$PCBs and 123.9 ± 58 kg of perfluorooctanoic acid (PFOA) within the ice system of the entire Beaufort Sea. Assuming complete release to seawater during spring melt, then these loads account for less than 1%, about 5% to 18% and 3% to 4% of the total Ŭ-HCH, Sum_{10}PCBs and PFOA, respectively, present in the PML (40 m deep) of the Beaufort Sea.

It is likely that this will also be the case for sea ice across the entire Arctic Ocean, in that only a small fraction of the organic contaminant burden will be held in the snow/ice at any given time relative to the load present in the PML. However, ArcRisk research shows that the sea-ice compartment experiences significant enrichment of these chemicals, marked by greatly elevated concentrations in ice relative to the beneath-ice seawater. This enrichment appears to be dependent on the age of the ice, with new ice showing markedly lower concentrations compared to single-season (one-year) ice. This enrichment of chemical concentrations in ice and the overlying snowpack was observed for hydrophobic chemicals such as PCBs but also for perfluoroalkyl substances, most notably the perfluorocarboxylates (PFCAs). $C_6$to C_{12} PFCAs were present in sea ice at concentrations which exceeded those in the underlying seawater (and often the overlying snowpack) by factors of approximately three to ten, resulting in bulk ice concentrations for PFOA greater than 1 ng/L in very remote parts of the Arctic. This enrichment has been observed in ice of varying ages (fresh ice, single-season and two-year ice), but appears to be most pronounced in single-season ice (the predominant ice form in the current, warmer Arctic).

The accumulation of contaminants during ice growth is not fully understood but is probably related to the presence and dynamics of brine (high-salinity water) within the ice. First-year sea ice, the dominant ice type in a warmer Arctic, has a high proportion of brine relative to multi-year ice. This brine will serve to transport contaminants within the ice during periods of melt with the potential for increased exposure of ice-associated biota during the late winter and early spring. Furthermore, concentrations of OC pesticides in bulk first-year sea ice were significantly higher than those observed in the late winter season snowpack, highlighting contaminant entrainment or accumulation from seawater during ice growth. PCB concentrations, however, were lower in ice relative to snow, suggesting the predominance of the atmosphere in supplying these compounds to the sea ice rather than the underlying seawater.

Ship-based campaign work across the Arctic has resulted in the first contemporary assessment of the current PCB burden in surface waters of the coastal Arctic seas, and the contribution played by the major Arctic-draining rivers in supplying PCBs to coastal sediments and water. The contemporary flux of PCBs (sum of 13 PCB congeners) from the six largest Arctic rivers was estimated at approximately 400 kg/yr. This is consistent with the estimated PCB inventory in the surface polar mixed layer of the entire Arctic Ocean of approximately 390 kg, indicating the importance of the major Arctic rivers in supplying these pollutants to the Arctic marine system, along with inputs from the atmosphere. Regionally, the coastal marine waters on the Russian side of the Arctic show a PCB profile that is more influenced by the heavier penta- and hexachlorinated PCB homologues in accordance with the major Russian PCB formulation ŚSovolû. Whereas the tri- and tetrachlorinated PCBs contribute about 75% to the PCB inventory of the Bering, Chukchi and Beaufort Seas, suggesting a predominantly atmospheric source. The hexachlorinated PCBs were proportionally more abundant in the European Arctic (Barents and Greenland Seas), implying the influence of waterborne transport from regions with previous heavy PCB consumption such as northern Europe and North America.

Accelerated ice-cap and glacier melt with climate change provides a mechanism by which previously deposited contaminants (currently stored in ice and perennial snowpacks) are released with meltwater to pose a secondary, ŕrecycledô source of these chemicals to freshwater and coastal marine systems. The deployment of passive water samplers to measure the organic contaminant signal in coastal fjord water, on a transect from the inner fjord to the open ocean, revealed the presence of both ocean-derived and glacier-derived contaminant end members affecting the chemicals present in surface waters of Godthåbsfjord in southwest Greenland. The pesticide distribution indicated that glaciers and snow caps around the fjord were secondary sources of these contaminants to the coastal marine environment, with chlordanes identified as potential indicator compounds for meltwater runoff. Other OCs such as Ǖ-HCH and hexachlorobenzene (HCB) were associated more with an oceanic influence rather than snow-cap/glacier melt. The levels of chlorinated pesticides in Godthåbsfjord were comparable to those in Svalbard fjord waters, although lower than in earlier studies from the Canadian Arctic. Clearly the release of previously deposited contaminants with perennial snow and ice melt is complex and evidence for contaminant remobilization is likely to be determined from sediment surveys in glacier-fed fjords rather than found in the water column.

3.2 Atmospheric processes

An overview of current atmospheric concentrations of ICES Ʃ$_7$PCBs (PCB28, PCB52, PCB101, PCB118, PCB138, PCB153, PCB180) at different sites in European and Arctic areas is presented in ArcRisk Deliverable D48. The highest atmospheric concentrations of Ʃ$_7$PCBs are present at various locations in central Europe and farther east, indicating the presence of hot spot areas. The highest PCB levels at background sites were also found in central Europe and east of Europe. There is an obvious decrease in the PCB concentrations from central Europe and southern Scandinavia to the Arctic areas.

The atmospheric PCB concentrations in the Arctic have shown a continuous decreasing trend over the past decades, after the international regulation of PCB usage was enforced as a part of the UNEP Stockholm Convention. A general decline in the concentrations at the atmospheric monitoring stations in the High Arctic (Alert and Zeppelin) and the European sub-Arctic (Pallas in northern Finland) was observed from 1998 to 2006.

A combined source apportionment, positive matrix factorization of atmospheric pollutants, and trajectory analyses applied for Pallas data for the period 1996 to 2009 showed that approximately half of the PCBs at Pallas originated from contaminated soil, with a systematic summer maximum. According to the trajectory sector distribution plot, the air masses related to this factor originated mainly from south and west. Observed increases in PCB concentrations in air from 2004 to 2009 have been attributed to the occurrence of forest fires in Siberia and North America.

Deposition from the atmosphere is an important route of entry of PCBs to both terrestrial and marine environments in the Arctic. However, deposition data from long-term monitoring in the High Arctic are not available.

3.3 Processes in snow, ice and water

Field measurements of PCBs in air, snow, ice and water performed in ArcRisk have provided a number of results of relevance to the assessment of the influence of climate change.

Evaporation of light congeners from the marine snow/sea-ice system such as di- and tri-PCBs from the snowpack is facilitated when the temperature rises. The PCBs are lost from the melting snowpack according to the physical-chemical properties of the respective congeners. Fresh precipitation and percolation from below the snowpack make it difficult to predict the input from a melting snowpack to the receiving area.

PCBs are also accumulated in sea ice, thus providing a chemical “store”. During periods of melting ice, these contaminants will be released to the water, providing a mechanism for exposure to the marine food web. The contribution of PCBs from melting snow and sea ice to Arctic seas is low in comparison to the input via Arctic rivers, although rapid thawing processes could impact the contaminant load of the ice-associated algae and fauna during the spring algal bloom

3.4 Contaminants in the terrestrial snowpack and their fate during melt

Winter snowfall and subsequent contaminant accumulation in the seasonal snowpack is considered a significant source of atmospherically derived pollution to spring meltwater. Fresh snowfall yielded higher concentrations of PFAS and halogenated flame retardants compared to older snow layers in the seasonal snowpack. Depositional snow fluxes of $C_{7\text{-}9}$ PFCAs ranged from 0.3 to 2.4 $ng/m^2/d$ and in broad agreement with fluxes derived for dated snow layers from the mid-2000s in the Canadian High Arctic. This indicates a wide array of well-mixed sources to be affecting the Arctic as whole for these chemicals. Snowfall events that occurred at warmer air temperatures (-2 to > 0 °C) provided approximately two-fold higher depositional fluxes for $C_{7\text{-}9}$ PFCAs than those derived during colder conditions (< -2 °C) even though the prevailing air mass direction was the same. This increase in deposition for snowfall events occurring during warmer air temperatures may be due to liquid water on the surface of snowflakes enhancing the scavenging ability of chemicals in the vapor phase. Particle-bound PFAS concentrations were generally low relative to the snow-sorbed or dissolved fraction, with snowfall particle-bound washout ratios about 10^5. The low particle-bound concentrations indicate that the PFAS arise in the remote snowpack due to their vapor transport in air and/or the photochemical oxidation of volatile perfluoro-precursors (e.g., fluorotelomer alcohols), rather than via particle-bound transport, with important implications with regard to the key PFAS sources affecting the Arctic.

In contrast, particle-bound concentrations dominated for several halogenated flame retardants including dechlorane-plus and bromodiphenyl ether (BDE)209 in the surface snow of remote northern Norway, with the most abundant novel flame retardants being tetrabromocyclohexane, pentabromotoluene and pentabromobenzene at concentrations averaging about 20 pg/L (markedly lower than PFAS). Aerosol transport of BDE209 is a feasible mechanism for the occurrence of this chemical in high-altitude lakes in the Pyrenees and Tatra mountains across Europe and this is also the case in the northern Norwegian snowpack.

For the melting snowpack, field work conducted in both northern Sweden and Norway revealed that ΣPCB and polybrominated diphenylether (PBDE) loads showed a gradual decrease in the snowpack over a two-week melt period (e.g., ΣPCB: 120 to <20 ng/m^2; ΣPBDE: 30 to ~5 ng/m^2), with a significant inverse correlation with nearby stream flow. On the other hand, HCHs were not correlated with stream flow and instead showed a marked rapid decline in their snowpack loads during the first few days following the onset of melt (e.g., α-HCH, Day-1: >1000 ng/m^2 to Day-3: <50 ng/m^2). Similarly, *p,p'*-DDT/DDE and predominantly particle-bound chemicals did not reveal any trend with either changing snowpack depth or stream flow. Unlike higher molecular weight PCBs, low particle-bound fractions (< 0.4) for several of the higher-brominated PBDEs were observed in the late-season snowpack (i.e., BDE153, BDE138, BDE183). This suggests novel formation in the snowpack (possibly due to photochemical debromination of nona-BDEs and deca-BDE) rather than input through particle-bound deposition from the atmosphere. Similarly, snowpack loads for $C_{4\text{-}9}$ PFCAs decreased gradually as melting progressed, but actually increased for $C_{4\text{-}8}$ perfluoroalkane sulfonic acids (PFSAs) and the longer chain C_{10-12} PFCAs. This enrichment in the diminishing snowpack may be due to their retardation within the pack related to their physical-chemical properties, whereby there is a greater tendency to partition to accumulated particulate matter. However, novel formation processes such as the photochemical oxidation of gas phase perfluoro-precursors on snow surfaces cannot be discounted. The C_{4-9} PFCAs are therefore likely to be released into meltwater runoff and hence enter surface waters, whereas the other PFASs that are retained in the final melt layer are likely to be released directly into the ground at the base of the snowpack.

Chemical input to surface waters during snowpack melt follows an elution order, rather than being delivered as a single "pulse". However, rapid thaw of the snowpack associated with anomalous winter warming events (climate change) may alter this behavior markedly, allowing rapid transfer to meltwater runoff.

3.5 Contaminants in biota

PCBs have been measured in a number of biological matrices in the Arctic environment. A west-to-east gradient for PCB levels has been identified, with the highest levels occurring in the eastern Arctic environments. In particular, the top predator animals of the eastern Arctic are exposed to considerable PCB burdens in their lipid tissues.

Within the ArcRisk project, a number of Arctic food items were analysed for contaminants. Traditional Greenlandic food items were analysed for legacy OC pesticides, PCBs, PBDEs and PFAS. Food items included fish filets from salmon and halibut, seal and whale beef and narwhal mattak (i.e., skin and blubber). The results from this study showed that food products derived from marine mammal species are contaminated with legacy POPs, including PCBs. The highest PCB levels were found in narwhal mattak (mean 1146 ng/g lipid weight (lw)), while the concentrations were lowest in smoked halibut (mean 37 ng/g lw). The median concentration of ΣPCBs in fresh salmon was 227 ng/g lw, while the median concentrations in smoked salmon and smoked halibut were 135 and 41 ng/g lw, respectively. Whale beef, seal meat and narwhal mattak contained median concentrations of ΣPCBs of 526, 302 and 1147 ng/g lw, respectively. PCB153 and PCB138 were dominant in all samples, contributing up to around 30% of the total PCB concentrations, except in seal meat in which these two compounds contributed up to 51% of ΣPCBs. Together with PCB118 and PCB101, these congeners contributed up to more than 50% of ΣPCBs in all samples and more than 60% in the seal meat.

The distribution pattern for the PCB congeners was similar for smoked and fresh salmon, but the concentrations were twice as high (lipid weight basis) in the fresh salmon compared to the smoked salmon for ΣPCB and some of the individual congeners (e.g., PCB52: 12 and 7 ng/g lw, respectively; PCB99: 13 and 7 ng/g lw; PCB118: 23 and 14 ng/g lw). This indicates that food processing has a marked effect on contaminant concentrations and could be related to changes in the extractable lipid content during the smoking process; alternatively, salmon sources as wild versus farmed may also be playing a role.

The OC pesticides, including *trans*-nonachlor, oxychlordane, *cis*-chlordane and HCB displayed some of the highest concentrations in narwhal mattak and whale beef (~100 to >600 ng/g lw), but were below tolerable daily intake (TDI) threshold levels set by the Danish National Food Institute. A combination of relatively high levels of POPs and intake frequency makes seal the most important contemporary food source of POPs among Inuits in Greenland, even though other food items, such as mattak, have higher levels of POPs. The levels in the ArcRisk study were slightly lower than or comparable with levels in other available studies from Greenland. Only a few PBDE congeners were detected in the biota samples, with BDE47 being the predominant congener. The levels of BDE47 varied from less than the limit of determination in smoked halibut up to 18 ng/g lw in narwhal mattak and 21 ng/g lw in whale beef. PFAS were detected in marine mammal samples, but not in the fish samples. All fish samples had been processed by the fish industry (i.e., washed, smoked, packed), which may be an important factor for PFAS falling below the limits of detection in this study, and in agreement with fish filet samples collected elsewhere across the Arctic. Where PFAS were detected in the food items, perfluorooctane sulfonic acid (PFOS) was the dominant compound, followed by several longer chain carboxylic acids, with ɆPFAS concentrations highest in whale beef (2.9 ng/g ww (wet weight)) and seal beef (13.5 ng/g). PFOS levels were slightly higher compared to finwhale muscle (0.5 ng/g ww) from Iceland and narwhal blubber (0.4 ng/g ww) from Nunavut, Canada. This study shows that the exclusion from the diet of local food items such as intestines and blubber have a strong positive effect on the reduction of POPs levels in food, but without markedly reducing the health benefits of a traditional food diet.

Enantiomer-selective analyses of chiral pesticides (Ǖ-HCH, *trans-* and *cis*-chlordane) and a metabolite (oxychlordane) were performed to elucidate contaminant exposure for zooplankton sampled from different Svalbard fjords, each characterized by different water masses. Among the compounds, *trans*- and oxychlordane were found to be most impacted by biodegradation. The enantiomeric fraction (EF) pattern of Ǖ-HCH was associated with ice cover/break-up, whereby biodegradation associated with plankton blooms (a feature of seasonal ice break-up) results in a specific range of EFs present in the zooplankton.

Enantiomer-selective analyses of chiral contaminants in Greenlandic traditional food items showed non-racemic (Í 0.5) enantiomeric fractions for almost all samples and compounds, indicating that significant metabolic processes affect contaminants present in biota and raising the potential of biologically active metabolites. The fish samples (salmon and halibut) showed a preferred degradation of (+)-Ǖ-HCH, while the marine mammals (seal and whale beef and narwhal mattak) showed a preferred degradation of (ī)-Ǖ-HCH. *Cis*-chlordane was racemic and oxychlordane close to racemic in seal meat, while both whale beef and the local delicacy narwhal mattak showed non-racemic EFs for these compounds. Hence, species-specific distribution exists and can be an important factor in future human dietary advice, if and when more knowledge about the toxic effects of each enantiomer and/or important contaminant metabolites becomes available.

Shrimp and halibut were collected from selected coastal fjords (near Tromsø, Norway) and screened for PCBs, OC pesticides, PBDEs and PFAS. For most of these substances, concentrations were similar to those measured in shrimp and Arctic cod elsewhere in the High Arctic. However, PFOS concentrations in whole shrimp averaged 2.77 ng/g ww, approximately ten-fold higher than levels measured in shrimp in the Canadian Arctic and possibly indicative of local contamination associated with activities or discharges from Tromsø. The average concentration of PFOA, at 0.19 ng/g ww, was much lower than that of PFOS, although the longer-chain PFCAs (C_{12}-PFDoA and C_{13}-PFTrA) also displayed elevated concentrations (about 1 ng/g ww) relative to the seal and narwhal in the Greenlandic marine food items discussed above. Furthermore, the majority of PFAS was also detected in halibut filets, with concentrations of PFOS ranging from 0.61 to 1.72 ng/g (ww), markedly different from fish filets collected elsewhere in the Arctic which generally have low to non-detectable PFAS in muscle tissue. Again, this indicates the influence of local sources of these chemicals to the coastal areas of the far north of Norway.

Levels of PCBs in fish depend on factors such as trophic level, age and lipid content, as well as geographical origin. Cod, which is a lean fish, will therefore have higher levels of PCBs on a lipid weight basis than fatty fishes (e.g., salmon). A comparison between cod and salmon on a wet weight basis shows lower levels in the cod (3.96 ng/g ww) than in salmon (8 to 17.9 ng/g ww). As documented in various scientific studies, the transfer of legacy POPs including PCBs from lower trophic level organisms into the top predators of the Arctic (i.e., polar bear, glaucous gull, polar fox) along a typical marine and/or terrestrial food web is usually associated with lipid transfer.

National and circum-Arctic monitoring programs indicate that PCB concentrations in the various environmental compartments, including biota, have generally decreased continuously over recent decades.

4 Exposure and health effects

Individuals and populations are exposed in their everyday lives to a large number of environmental contaminants occurring in many different combinations, and they have complicated effects on physiological functions in the body. Body burdens of contaminants vary with the source and level of contamination, exposure paths (including life-style factors), and genetic background of individuals. Environmental contaminants may cause several adverse health outcomes, and in epidemiological studies increased risks for congenital abnormalities, chronic diseases, and cancers have been observed. The perinatal period is particularly critical for the developing fetus and developmental effects may be a result of the exposures of both before and during pregnancy; they also depend on the timing of the exposure during the pregnancy. The adverse effects of contaminants depend greatly on their fate in the human body, which is a function of contaminant toxicokinetics, such as metabolism and cell transporter functions; during metabolic processes, the xenobiotic chemicals may become even more toxic, mutagenic, or carcinogenic products.

Contamination of dietary intake provides the most extensive exposure to many POPs and most heavy metals, and is the main source of non-occupational exposure in Arctic populations. In particular, Arctic populations have a higher exposure to POPs than other European populations owing to the high levels of POPs in Arctic marine mammals and some fish species that are consumed as part of the traditional diet.

In the ArcRisk project, comprehensive data from recent and on-going AMAP assessments concerning adverse health outcomes induced by contaminants were combined with data from a large number of publications and on-going research projects in the Arctic into an extensive database to provide a picture of baseline health status in the Arctic. This database will serve as a longer-term legacy of the ArcRisk project for future use. This extensive database regarding health outcomes in relation to contaminant exposure will provide further opportunities to identify particular combinations of different contaminants that deserve future research in the light of their implications for human health. Results based on reviews and analyses of the data collected in this database are reported below, together with results from recent and on-going human health cohort studies in Arctic populations and, for comparison, in several exposed populations in Europe. Furthermore, the results of a study the dietary sources of contaminants in some ArcRisk cohorts and the development of a toxicokinetic model to estimate the health risks of contaminant exposure are also described.

4.1 Mercury

In this project, the health research on mercury focused on compiling and examining available information on exposure to and health effects of mercury in population groups in the Arctic and in the

Mediterranean region. Data generated within the EU FP6 project PHIME were also included in this evaluation as well as a review of data published from 2000 on.

Chronic exposure to methylmercury (MeHg) via consumption of fish and marine mammals is still a major concern for human health, especially developmental exposure that may lead to neurological alterations in the central nervous system, including cognitive and motor dysfunctions. Large-scale epidemiological studies concerning childhood development and neurological disabilities in relation to *in utero* exposure to MeHg have been conducted in various fish-eating communities around the world. Neurological disabilities including language, learning, and attention deficits and, to a lesser extent, motor and visual-spatial impairment have been reported due to exposure to MeHg. High dietary MeHg exposure is suspected to reduce birth weight and weaken the immune functions by altering the relative abundance of TH-lymphocytes, which play a role in determining an individualõs immune responsiveness.

For an assessment of current human mercury (Hg) exposure, concentrations of mercury in hair and blood samples are often used an indicator. In the ArcRisk cohorts (ArcRisk Deliverable D39), data on Hg in hair were obtained from samples from populations in Spain (Ribera dõEbre, Menorca), Slovenia, Croatia and Greece in addition to Arctic parts of Norway (MISA study: Northern Norway mother-and-child contaminant cohort study). Data on Hg concentrations in blood were obtained, in addition to the above populations, also in samples from cohorts in Italy, northern Finland and Norway.

Among the ArcRisk study groups, participants in the Greek part of the PHIME cohort studies appeared to have higher exposure than those in the Slovenian part of the study. The main reason for the difference is associated with the consumption of locally caught fish in Greece, while in Slovenia fish is mostly obtained from the fish market supplied by farmed fish. Participants in the MISA study also appeared to have lower exposure. People living in Northern Finland appeared to have lower exposure than the participants in the Norwegian Fish and Game (NFG) Study. Children living in Ribera dõErbe and Menorca appeared to have exposures in the same range as adults in Greece.

Other studies have shown that mercury exposure in Arctic Canada is variable and has decreased with time. Exposure in the Inuvik region is lower than in the Baffin region, which again is lower than in Nunavik. The exposure in Nunavik has previously been several-fold higher than in the ArcRisk study groups but appears to have declined to the same range as in the Norwegian high fish consumers and in Greece at the latest sampling in 2007. In a Russian study, Hg exposure among adult males and females was found to be in the same range but somewhat higher than in the NFG high consumers. However, the level in pregnant women was considerably lower, and in the same range as in maternal blood from Italy, Croatia and Arctic Norway, and lower than in maternal blood from Greece. The Greek participants were from islands of the Eastern Aegean where the population mostly consumes locally caught fish containing higher Hg levels than those obtained from markets (with mostly aquacultured fish). Similar values were reported also for the Spanish islands, where predominantly local fish is consumed. Living in inland or coastal municipalities in Norway also affected blood mercury levels.

In addition, a review of current studies of Hg exposure in Europe, taking into account the potential routes of Hg exposure, actual Hg exposure levels assessed by different biomarkers and the effects of Hg in Europe, was also made. All published studies from 2000 onwards were reviewed and exposure and effects studies were mirrored against known Hg levels in environmental compartments by mapping the various population groups studied and taking into account known sources of Hg. A study of the spatial distribution trends confirmed that the highest exposure levels to MeHg are found in coastal populations who consume more fish in their diet compared to inland populations. Fewer studies addressed exposure to other Hg compounds, mainly elemental Hg through inhalation of Hg in air and inorganic Hg in food, particularly in highly contaminated areas. Overall, at the currently low exposure levels of Hg prevalently found in Europe further studies are needed to confirm the risk to European populations taking into consideration exposure to various Hg compounds and mixtures of stressors with similar end-points, nutritional status, as well as a detailed understanding of Hg concentrations in fish present on European markets.

Health effects were investigated in the PHIME cohorts and it was concluded that the exposure to mercury was low and did not significantly affect neurodevelopment by the age of 18 months (ArcRisk Deliverable D36). Exposure of the foetus to higher Hg levels during gestation did not cause lower performances in cognitive, language and motor neurodevelopmental testing, because other correlated variables (e.g., socio-economic indicators, maternal IQ) predict these outcomes. Higher fish consumption in pregnancy was associated with higher cognitive and language (but not motor) neurodevelopmental performance at 18 months of age, thus emphasizing the benefits of including fish in the diet.

In conclusion, consumption of fish is a major source of Hg exposure to humans in Europe and in the Arctic. Consequently, the highest exposure levels to MeHg in Europe are found in coastal populations that consume more fish, particularly locally caught fish. Exposure to mercury did not have any negative effect on neurodevelopment at the age of 18 months in the ArcRisk studies, but further studies are needed to evaluate the potential threat to European populations taking into consideration exposure to various Hg compounds and mixtures of stressors with similar end-points.

4.2 PCBs

Several hundred papers dealing with toxicological consequences of exposure to PCBs have been published and detailed information about PCBs and health is also provided in various reports from internationally recognized organizations. Nonetheless, although numerous studies about health effects of PCBs have been published, discrepancies remain, for example, regarding neurotoxicity in children, which is partly due to the complex chemistry of the 209 different PCB congeners.

A limited number of health studies on Arctic populations have been performed focusing on different health end-points. Some of the studies have found evidence for effects of PCBs, e.g., hormone disruptive potential, whereas other results have been inconclusive regarding the effects of PCB exposure for the specific end-point studied.

4.2.1 Critical reviews of health outcomes

Large research efforts on human health effects and dose-response relationships for organochlorine compounds have been performed and published. Combining the wealth of data collected in these studies should provide an opportunity for a more detailed and accurate assessment of human health effects. In ArcRisk, a review and assessment of published research on PCB effects on humans (mainly the end-points changed sex ratio and weight of newborn infants) was performed. This literature review revealed that some research findings cannot readily be combined in a meta-analysis using established effect size statistics (ArcRisk Deliverable D41). These include findings generated by multivariable analysis, e.g., multiple regression, discriminant analysis, factor analysis and structural equation models. Their complexity and diversity across studies with regard to the selection of variables and reporting practices has made it difficult to combine and compare the original studies. Multivariable regression is a flexible method of data analysis that may be appropriate whenever a quantitative dependent variable is to be examined in relationship with any other predictor variables. The synthesis of regression coefficients has received increased attention in recent years due to increased use of multivariable methods in medical research.

The main problems with these methods include the use of different measurement scales of explanatory variables across studies and the lack of required information in the study reports. The presentation of the findings should not rely only on the coefficients of the estimated models. The reporting of the models needs attachments such as tables describing the basic data. Studies should focus more on reporting descriptive statistics about the distributions of response variables and explanatory variables. This would help other researchers to utilize the results in their approaches to summarize and meta-analyse the magnitude of the effects.

A meta-analysis of published studies of the influence of PCB exposure on sex ratios performed in ArcRisk did not indicate that parental exposure to PCBs has any effect on the sex of their children, while the meta-analysis on birth weight vs. PCBs showed that a high maternal PCB body burden is probably related to low birth weight. However, a dose-response relationship with statistical significance could not be constructed based on this meta-analysis.

In conclusion, studies of human health outcomes in relation to contaminants are seldom conclusive and it is difficult to link health effects to specific contaminants. If cohort studies addressing health effects of contaminants were conducted according to agreed protocols, this would increase their suitability for meta-analyses.

4.2.2 Health effects of PCBs in the Mediterranean region

Several health effects have been associated with exposure to organohalogen contaminants in Mediterranean study populations. Assessment of the possible health effects of long-term exposure to low concentrations of these POPs requires their analysis in a large number of individuals. Newborns are more sensitive to contaminant exposure because their organs and metabolic functions are under development. In the Mediterranean cohorts, exposure to organohalogens has been related to, for example, low birth size, low birth weight, low birth head circumference, poor social behavior, increases in the incidence of attention-deficit hyperactivity disorder (ADHD), decreases in cognitive skills, overweight, and alterations of porphyrin, thyroid and liver metabolism. Most of these effects have been observed in the early stages of life. These findings need to be confirmed by studies of cohorts from other geographical areas.

4.2.3 Temporal trends of PCBs in Arctic populations

PCBs, especially those that contain a greater number of chlorine atoms, are readily soluble in fats and thus tend to accumulate in fat-rich tissues, such as liver and adipose tissue in any organ. In humans, the average concentration of PCBs in fatty tissues is over a hundred times greater than in the food they eat. PCBs have been detected in maternal blood and serum, umbilical cord blood, foetal adipose tissue, placenta, infant blood, childrenôs blood and serum, and breast milk. In the blood of breast-fed infants, the concentration may be many times higher than in maternal blood and the mean concentration of PCBs has been shown to remain higher in the blood of breast-fed children than in the blood of bottle-fed children up to 5 years of age.

PCBs have a long half-life in humans due to their lipid solubility; for example, from blood levels it has been calculated that the half-life is 15.5 years for PCB170, 14.4 years for PCB153, and 11.5 years for PCB180. Eventually they are excreted, mainly through the faeces, urine and breast milk.

PCB153 is among the most prevalent PCBs found in human populations and thus has been used as an indicator for PCBs. Measurements were made of the concentrations of PCB153 in plasma lipids in Inuit women living in Disko Bay (Greenland) in 1994-2006, Nuuk (Greenland) in 1999-2005, and Nunavik (Quebec, Canada) in 1992-2007. These good resolution data covering the years 1992 to 2007 clearly show declining trends of the geometric means of PCB153 in all regions. Concentrations of PCB153 were lower in mothers from Nuuk than in mothers from Disko Bay. The differences could be due to lower consumption of marine mammals in the capital Nuuk than in the remote region of Disko Bay. The lower concentrations of PCB153 in pregnant women from the Disko Bay area may be due to an increase in the consumption of traditional food from the marine ecosystem that contains lower concentrations of PCB153.

In general, levels of PCBs measured in humans in ArcRisk study populations have declined over the past 20 to 30 years. However, concentrations of some POPs remain relatively high, with PCB153 concentrations averaging about 50 ng/g lipid weight in women living in Disko Bay and Nuuk (Greenland) and Nunavik (Quebec, Canada).

PCB153-associated human health effects and risks were assessed using data obtained from the AMAP biomonitoring programme noted above on pregnant Inuit women from Nunavik, Disco Bay, and Nuuk and a one-compartment toxicokinetic model. The aim was to extrapolate body burdens and exposure to the whole lifespan of the population covering birth cohorts from 1940, 1950, 1960, 1970, 1980, and 1990. The toxicokinetic model shows that while PCB levels are decreasing in most population groups, the level will remain elevated above the benchmark dose lower limit (BMDL) for the 90th percentile until around 2020.

4.2.4 Future risk prediction

To assess potential PCB153-associated human health effects and risks, it is necessary to model past exposure (ArcRisk Deliverable D41). PCB153 blood concentrations, obtained from the AMAP biomonitoring programme, in Inuit women covering the years 1994-2006 at Disko Bay, 1999-2005 at Nuuk, and 1992-2007 at Nunavik were used to extrapolate body burdens and exposure to the whole lifespan of the population by the one-compartment toxicokinetic model. The ratio between the actual exposure (average daily dose (ADD)) and the corresponding threshold exposure (Reference Dose (RfD)) for a specific health end-point results in a hazard quotient (HQ), which is a unitless number. An HQ less than 1 means that the hazard is not considered to be a threat to public health (including sensitive subgroups) based on the exposure scenario and toxicity data available. Likewise, if the exposure level to a chemical exceeds the corresponding threshold exposure (i.e., HQ greater than 1), the exposure may present a potential adverse, non-cancer effect. Based on the toxicokinetic modelling results, calculated Hazard Quotients were found to be higher than 1 between the years 1955 and 1987 for the 90th population percentile and from 1956 to 1984 for the 50th population percentile. Cancer risks were estimated based on an oral cancer slope factor (CSF) provided by U.S. EPA-IRIS for PCBs. The probability of carcinogenic effects was calculated with two cancer slope factors: 4×10^{-2} per mg/kg-day for lowest risk and persistence (central-estimate slope factor) and 2.0 per mg/kg-day for high risk and persistence (upper-bound slope factor). Cancer risk was determined as follows: CR = LADD × CSF. Cancer risk for overall exposure to PCB153 ranged from 4.6×10^{-5} to 1.8×10^{-6} for the 90th percentile and 3.6×10^{-5} to 1.4×10^{-8} for the 50th percentile between 1930 and 2049, when central estimates or upper-bound slope factors were applied. Cancer risk was below 1×10^{-6} for the same time period when a lower slope factor was applied. Significant future research requirements to improve health risk characterization include, among others, larger sample sizes, better analytical accuracy, fewer assumptions in exposure assessment and, consequently, a better choice of the toxicity benchmark used to develop the hazard quotient.

4.2.5 Effects of dietary recommendations in indigenous communities of Arctic Russia

A study of the change in PCB levels in blood between 2002 and 2011 was conducted with representatives of the cohorts of indigenous people who had participated in an earlier survey in 2001 and 2002 (Persistent Toxic Substances, Food Security and Indigenous Peoples of the Russian North). After the 2002 study, dietary and other health recommendations were implemented in a number of indigenous villages during 2003 to 2010. Temporal trends and structural exposure changes were evaluated based on measurement results of the PCB concentrations in blood in 2011 compared with results obtained in 2002. Identical approaches were employed to assess the impact of contaminants, diet, traditional activities and other social and behavioural risk factors. Blood sampling protocols, written consent, storage and transportation followed established AMAP guidelines. It is assumed that such a comparison of biomarkers of exposure, documented for two groups of indigenous communities (those involved and those not involved in the implementation of the recommendations), can be used to assess the overall effect of the implementation of rehabilitation measures, and potential consequences associated with other non-chemical risk factors, particularly climate change and social and economic conditions of people living in the Arctic indigenous communities.

The change in geometric mean serum concentrations of PCBs in children of followed-up indigenous cohorts, although not large, was reflected by the academic performance of schoolchildren, indicating the possibility of a known adverse effect of PCBs on the neuropsychological development of children. Such effects in children from those villages where rehabilitation activities were carried out apparently turned out to be somewhat less, according to the median estimate of their knowledge in elementary school. For example, in a cohort of children whose birth concentration of PCBs in blood was less than 1 µg/L, average school performance was 5% to 7% higher than in children with PCB concentrations at birth greater than 1 µg/L. Comparing a cohort of children living in villages where rehabilitation activities were carried out, the average of their performance in primary school was approximately 10% higher than the children of the villages where such activities are not conducted.

4.2.6 PCBs in blood serum of Arctic and European populations

The results from a number of surveys of PCB153 in blood serum in Arctic and European populations showed that the highest concentrations, and thus exposure, were found among participants in the Norwegian Fish and Game Study and the lowest were found in the Spanish INMA and Norwegian MISA study groups. However, although the same congener has been measured, higher serum concentrations in the Norwegian Fish and Game Study participants can be expected due to the higher age among the participants (median: 55 years, range: 21 to 80 years). Furthermore, participants in the high-consumer group were invited due to their high consumption of foods that generally contain higher levels of POPs compared to other food groups. In addition, the sampling year needs to be taken into consideration, because human PCB exposure is decreasing in several regions. The NFG study samples are from 2003, while the sampling period and ages are more comparable between birth cohort mothers from Spain (INMA) and Arctic Norway (MISA). Interestingly, the mean and upper range of exposure was lower among the MISA participants than in the INMA cohorts (both in maternal and cord blood). The sampling period was also in the same range (INMA 2004-2008, except Menorca which was sampled 1997-1999, and MISA 2007-2009). Thus, this might indicate that women in the fertile age living in Spain have higher PCB exposures than women in the northern part of Norway.

4.2.7 Temporal trends of POPs in Norwegian men

In a study of POPs in Norwegian men from 1979 to 2007, substantial declines were observed for all POPs with the exception of chlordanes (e.g., *trans*-nonachlor). Overall, decreases were observed from 1979 in concentrations of most pentachlorinated PCBs (PCB99, PCB101, PCB105, PCB118, and PCB123) and hexachlorinated PCBs (PCB128, PCB141, PCB149, PCB153, and PCB167). Concentrations of heptachlorinated PCBs (PCB170, PCB180, PCB187, and PCB194) initially increased from 1979 to 1986 and then declined in subsequent years. Overall, the findings suggested that POPs concentrations decreased between 1979 and 2007 in men from northern Norway, and the average summed POPs concentrations in 2007 were one third of the concentrations measured in 1979. Peak PCB153 concentrations were measured in 1979 and 1986, confirming this period as the years of highest human exposure. The downward trends in serum concentrations likely reflect declining environmental concentrations due to reduced emissions during the same time period. This is in accordance with previous findings for environmental and human POPs concentrations in Europe. The findings indicate that serum concentrations of DDTs peaked before those of PCBs, which is consistent with emission estimates for DDTs. The delay in decrease of global emissions of PCBs could be due to the long lifetime of PCB-containing products (e.g., transformers, capacitors). The findings suggested that regulatory measures to reduce the manufacture and use of POPs during the 1970s and 1980s had rapid impacts not only on environmental concentrations, but also on human exposures.

4.2.8 POPs in breast milk - a comparative risk assessment

POPs (PCBs, DDT, HCB, and ϐ- and ɔ-HCH) levels in breast milk in central European populations were used for comparative studies in the ArcRisk project (ArcRisk Deliverable D36). A new method for POPs risk assessment for breast-feeding mothers was developed. The method depends on a reverse model for life-long POPs exposure doses of mothers. In this toxicokinetic model (PBPK model), reverse computation of the chronic daily intake (CDI) of POPs from human milk was used based on a model for the accumulation of organic compounds in nursing mothers. The human body is considered as a flux-through system and the main reverse-model results are the dietary, maternal intake (CDI). Inside the body, phase equilibrium is assumed and the compoundôs elimination from the body by exhalation and excretion is taken into account. In this case, human health risk was predicted by using the reference dose (RfD) approach.

The results suggest that for the selected POPs, the highest risk is associated with PCBs. Based on the human biomonitoring of POPs concentrations in breast milk in the Czech Republic, it has been concluded that the new method presented here for human health risk assessment of breast-feeding mothers is a useful tool for interpretation of the results of long-term biomonitoring epidemiological studies and that biomonitoring of human milk is a useful tool to evaluate the internal exposure of humans to different chemical substances.

4.3 Perfluorinated and brominated compounds

4.3.1 Foetal exposure

That pregnant women and young children constitute critical groups for contaminant exposure is well recognized. Contaminants can be passed from the mother to the foetus through the placenta and to young children during breastfeeding and affect critical stages of development. At the same time, breastfeeding is very beneficial to the health of newborns and in establishing the bond between mother and child.

Mother-and-child health studies in ArcRisk investigated the relationship between the diet of pregnant women and contaminant levels in their blood, finding that fish consumption was an important factor. A study of the maternal transfer of polybrominated diphenylethers (PBDEs) showed that levels in the blood from umbilical cords of newborns were clearly associated with the levels in their motherôs blood serum; this study indicated a broader range of exposure sources than fish, however, as neonates from rural areas had statistically significantly lower levels of contaminants in their blood than those from urban areas. A long-term study of the trends of PCBs and several organochlorine pesticides in breast milk from women in Central Europe has shown continually decreasing levels over the 15 years of the study.

In order to reach foetal circulation, foreign chemicals must cross the placenta, the main interface between mother and foetus. Based on analyses made from cord blood, it is known that the foetus is exposed to environmental contaminants present in the maternal circulation (ArcRisk Deliverable D42). The important properties determining the placental transfer by passive diffusion are molecular weight, pKa (acid dissociation constant), lipid solubility and protein binding. In addition to passive diffusion, the placenta expresses a large variety of transporter proteins which modify placental transfer processes. Transporter proteins also transfer foreign compounds such as therapeutic agents, environmental contaminants and chemical carcinogens bearing structural resemblance to their physiological substrates. Thus, even if the placenta cannot prevent the transfer of foreign chemicals into foetal circulation, it modifies their transfer and toxicity.

During the ArcRisk project, toxic metals and perfluorinated compounds were studied. The study showed that the foetus is significantly exposed to perfluorinated compounds (PFOS and PFOA) during gestation. In addition, the findings suggest that environmental contaminants interact with placental transporter proteins, which may affect foetal exposure to xenobiotics. The clinical significance of these findings is still unclear. The data imply that transporter protein function may cause person-to-person variation in foetal exposure to environmental contaminants, which may affect the individual risk for adverse events after exposure to harmful compounds.

4.4 Other contaminants in Mediterranean populations

Several health effects have been identified as a consequence of exposure to organohalogen pollutants in the Mediterranean populations studied. Significant decreases in birth weight have been associated with higher cord blood serum concentrations of DDTs and, marginally, hexachlorobenzene (HCB) and β-hexachlorocyclohexane (β-HCH). Decreases in birth length have been related to high HCB concentrations and decreases in head circumference to increased DDT. Higher concentrations of β-HCH have also been significantly associated with higher levels of thyroid-stimulating hormone at birth.

The Mediterranean populations studied also show higher concentrations of polybrominated diphenylethers (PBDEs) than the European equivalents (ArcRisk Deliverable D39). Part of the difference is due to the high proportion of BDE209 in the Mediterranean cohorts. However, the Mediterranean values are clearly lower than those observed in the USA. According to the PBDE composition, deca-bromo and penta-bromo PBDEs seem to be the primary and secondary commercial mixtures responsible for the accumulation of these compounds in both adults and newborns from the

Mediterranean areas. Young individuals showed the highest concentrations, which is likely related to high exposure to these pollutants during their growth period.

4.5 Diet and future predictions

It seems clear that changes in human behaviour, in particular dietary transitions, will have an impact on future human POPs exposure in the Arctic. This includes the gradual displacement of traditional food items (especially marine mammals) by foods imported from other regions. The extent to which subpopulations shift away from the consumption of locally harvested food represents the extent to which these people become decoupled from any potential changes in exposure to POPs related to environmental changes in the local or regional environment. Human food-web model calculations suggest that such dietary transitions can result in POPs exposure reductions in excess of an order of magnitude over a period of several decades. The dietary transition may therefore overwhelm any other potential effects of global climate change on human exposure to organic contaminants in the North, especially if it involves a substantial reduction in the consumption of marine mammals.

The influence of climate changes on future contaminant exposure from food will to a large degree depend on the consumption of fish and the type of fish (wild or farmed fish) and other seafood. Overall, at this point of time it is not possible to draw final conclusions about the exposure trends matching health trends.

5 ArcRisk case studies on mercury and PCBs

ArcRisk is a multidisciplinary project covering research on the occurrence, transport and fate, exposure and health impacts of selected contaminants in Europe and the Arctic, and the projected influence of climate change on these processes; this has been illustrated by case studies on PCBs and mercury. ArcRisk results from measurements, models and studies on health effects have been used to obtain an overall picture of PCBs and mercury and to describe the links between emissions and health effects in the Arctic. These case studies (in ArcRisk Deliverable D48) have contributed to our understanding of the complex relationships between sources, transport, bioaccumulation, exposure and health impacts of PCBs and mercury in relation to climate change.

5.1 PCB case study

Although the production and use of PCBs have been banned under the Stockholm Convention, emissions of PCBs still occur from, e.g., construction and waste dumps in industrialized countries, albeit at decreasing rates. Secondary emissions of PCBs accumulated in environmental reservoirs (sediment, water, soil, snow and ice) are important for the continued ŕecyclingô of these chemicals and climate change will most likely increase these secondary emissions, but not to an extent that the overall decreasing trend is compensated.

Atmospheric long-range transport is considered to be a major route for the global distribution of PCBs to the Arctic and deposition from the atmosphere is an important pathway of PCBs to both terrestrial and marine environments in the Arctic. In addition to long-range transport via the atmosphere, transport via oceanic currents is important for the occurrence of PCBs in the Arctic. Climate change is likely to affect all of these pathways and the ultimate environmental fate. The climate parameters most relevant in relation to the transport and fate of PCBs, as for other POPs, include changes in temperature, precipitation, sea- and land-ice cover, and the global circulation of the atmosphere and the oceans.

Precipitation has been identified as an important factor in relation to PCB levels in the snowpack surface, where PCBs may also accumulate over time, particularly in areas with a perennial snowpack (e.g., areas of high-altitude glacier surfaces and ice-caps). Climate-related effects such as changes in precipitation patterns and snow-melt periods can thus influence the accumulation and release of PCBs from the snow. PCBs will also be accumulated in sea ice, thus providing a chemical ‘store’, and climate change will affect the pathways and mobility of PCBs in Arctic ice. During periods of melting ice, these contaminants will be released to the water thus providing a mechanism for exposure of the marine food web, especially during early spring. However, in total, PCB releases from sea ice and snow will not provide large inputs to the Arctic Ocean in comparison to those predicted for the major pan-Arctic rivers.

With regard to the influence of climate change, model results indicate relative increases in PCB concentrations both in the Arctic atmosphere and the Arctic Ocean due to the effect of global climate change. The modelled increases are mostly due to the effect of temperature on volatilization from primary and secondary emission sources.

The atmospheric concentrations of PCBs in the Arctic have shown a continuous decreasing trend over recent decades. Although the modelled concentrations of PCBs were found in some cases to be higher under the climate change scenarios (up to a factor of five), absolute concentrations by the end of the 21st century are forecasted to be several orders of magnitude below present levels in all scenarios.

Temperature changes resulting from global climate change will have a direct influence on the partitioning of PCBs in natural waters. The major impact of climate on bioavailability is likely to be due to changes in the distribution of PCBs in exposure media owing to increasing temperature or changes in primary productivity.

It is well-established that global climate change can affect the food-web transfer of PCBs through both indirect and direct effects. A food-web model was developed to simulate the bioaccumulation of selected PCB congeners in a marine food web located near Svalbard. The model was evaluated with monitoring data for the food web collected within ArcRisk. However, the model was shown to largely underpredict the concentrations and there are still problems with our ability to estimate bioaccumulation in Arctic marine food webs with state-of-the-art modelling tools under existing climate conditions. A large limitation is the inadequate quantity and quality of field data needed for model evaluation.

PCBs are present in a number of biological matrices in the Arctic environment and within the ArcRisk project a number of Arctic food products were analysed for PCBs. The results showed that food products are contaminated with PCBs, with narwhal containing the highest PCB levels.

Regarding effects of environmental PCB exposure on human health, future studies are necessary in order to reduce the uncertainties in the health risk estimation of PCBs by better identifying the sources of contamination and by improving the overall process of health risk assessment. It seems clear that changes in human behaviour, in particular dietary transitions, will have an impact on future human exposure to POPs. In the Arctic, this includes the gradual displacement of traditional food items (especially marine mammals) by foods imported from other regions. The influence of climate change on future contaminant exposure from food will to a large degree depend on the consumption of fish and the type of fish (wild or farmed fish) and other seafood.

5.2 Mercury case study

The potential impacts of climate change on mercury emissions, transport, transformations and exposure have been discussed extensively in the mercury case study. This report also includes information on future scenarios for emissions and how they will be affected by various policy measures mainly directed at reducing CO_2 emissions as well as mercury.

The environmental cycling of mercury is complex and involves a number of chemical and microbial processes in which atmospheric oxidation, evasion of volatile mercury from snow and other surfaces and methylation/demethylation are the most prominent. All of these processes are likely to be influenced by climate change via, for example, increased temperatures which may cause increased rates of chemical and microbial processes as well as melting of snow and ice. A shorter period of ice and snow cover means that inorganic Hg can be transformed to MeHg through greater productivity in wetlands and marine and freshwater environments. It is in these environments that most of the risks of Hg to humans and wildlife are occurring.

Modelling results from the ArcRisk project for two different decades, 1990-1999 and 2090-2099, showed a significant decrease of Hg deposition over the Arctic Ocean and a significant increase over the continents from 1990-1999 to 2090-2099. A change in ice cover during the period leads to a decrease, and its influence on Arctic Mercury Atmospheric Depletion Events in the model. The deposition of mercury directly to the Arctic Ocean is lower by 20% to 40% in the 2090-2099 time period compared to the 1990-1999 time period. The increase over the continents (higher by 20% to 40%) results from a significant increase in the ozone concentrations in the troposphere.

Based on effects from recent climate warming on the Arcticȏs physical environment, it is concluded that the most important impacts have occurred in precipitation rates and type of precipitation (i.e., rain or snow), river discharge and seasonality, lake-ice and sea-ice seasonality, thickness and extent, declining length and depth of snow cover, increasing active layer depth in permafrost soils, altered vegetation and drainage basins, and changing atmospheric connectivity between the Arctic and southern latitudes.

An assessment of the impact of future changes of climate on human health in the Arctic region is a very difficult task that depends on the quality of information on energy projections, related Hg emission scenarios, environmental transport and intercompartmental transfer of Hg leading to environmental and human exposures in the future. The quality and accuracy of this information differ from fairly adequate data for energy projects to very uncertain information on human exposures in the future.

Through bioaccumulation, Hg reaches higher concentrations in biota than are initially present in environmental media. This bioaccumulation process of Hg in biota is very complex, and therefore very difficult to predict under climate change as the structure and dynamics of marine food webs are being altered in response to climate change. Humans are exposed to methylmercury, the most toxic form, through their diet, mainly from consumption of fish and other seafood. Dietary exposure will be affected by climate change through changes in relevant food-web structures, which are thought to be the most important single factor for human exposure to methylmercury and are far more important than other environmental characteristics. Other societal factors, such as the consumption of traditional foods and ways of living (especially in the Arctic populations), may also be important with regard to Hg exposure. As the main exposure route is now, and will be in the future, the consumption of fish contaminated with Hg, information on fish distribution and Hg contamination in the context of climate change is crucial in the assessment of the climate change impacts on human health with respect to Hg.

Fish consumption is foreseen as a major supply for food and proteins in the growing population worldwide, migrating more and more towards coastal areas. This also applies to the Arctic populations.

There is no linearity in the relationship between the future changes of Hg emissions and changes in Hg concentrations in fish. Thus, while chemical recovery of the abiotic environment due to emissions reductions could be measured as a short-term process, the biological recovery of ecosystems will take much longer time. It is therefore of great importance that emission reduction measures are implemented as soon as possible. This is particularly true in the case when climate change will result in revolatilization of Hg already accumulated in soil and water (re-emission). This process is expected to emit more Hg to the atmosphere in the future compared with current re-emissions due to the estimated air temperature rise. This enhanced re-emission may counteract reductions of emissions from anthropogenic sources. Therefore, it is important that measures to reduce emissions from anthropogenic sources are introduced.

Potential impact of the ArcRisk project

Humans are exposed to environmental contaminants such as persistent organic pollutants (POPs) and toxic metals through ingestion of food and drinking water and also through inhalation of ambient and indoor air particles. A main objective of the ArcRisk project was to relate information on environmental exposure to information on human health impacts, and thereby to try to define human health responses to contaminant exposure. These relationships could then be analysed in the context of climate change influences on various steps in the path from use and releases of chemicals to dietary exposure. This type of analysis is necessary in order to inform policy-making and further develop and implement strategies to reduce the impacts of environmental contaminants on the health of populations living in the Arctic region and in Europe, both now and in the future under changing climate conditions. Major efforts in the ArcRisk project have been made to study the effects of dietary exposure to contaminants on human health in the Arctic, and to compare them with the effects of exposure to contaminants in other regions of Europe, particularly the Mediterranean region.

1 Implications regarding contaminant exposures and human health

The overarching aim of the ArcRisk project was to investigate the potential for climate change to increase exposure to environmental contaminants, and the implications of this for future risks to human health. By focusing the project on the Arctic, where rapid changes are already under way, it is possible to gain insights into possible future changes in other regions. Comparisons between the Arctic and other areas of Europe therefore provide a basis for enhancing our knowledge in these subject areas. The research under ArcRisk considered a number of chemicals including substances that are already banned or regulated due to recognized negative impacts on human health and the environment, such as mercury (Hg), polychlorinated biphenyls (PCBs) and pesticides such as DDT, termed ðlegacyô contaminants. A number of compounds that entered use in society more recently,or which are only now emerging as environmental contaminants, such as perfluorinated and brominated compounds, were also studied. Human exposures and body burdens of contaminants have been assessed via biomonitoring (blood, hair and breast milk) as well as sampling and chemical analysis of some main food items.

ArcRisk studies confirm that for the contaminants studied exposure in the general population is mainly related to the consumption of fish and shellfish, as well as marine mammals in the Arctic. The ArcRisk project confirmed that most population groups in Europe and the Arctic are subject to a widespread low-level exposure to environmental contaminants. Consumption of certain species of fish and, in the case of the Arctic, marine mammals is of particular significance in this respect. Analyses of Arctic marine foods (fish, shrimps, marine mammals) revealed a range of concentrations of legacy contaminants as well as chemicals that have been used in industry, agriculture or household products more recently. They were generally at concentrations well below levels of concern, but were found at high concentrations in some marine mammals such as seals and toothed whales that accumulate mercury in their muscle and potentially large concentrations of organic contaminants in their fatty tissues and liver. Much of the contamination in the Arctic is due to long-range transport; however, Arctic towns and settlements may be important sources of contaminants to local coastal fisheries. Similarly, in other areas of Europe, fish and shellfish consumption is an important route of exposure. The highest exposure levels to methylmercury occur in populations living near the sea and consuming more fish compared to inland populations.

In one ArcRisk study in a northern area, it was determined that oily fish, including salmon, trout, mackerel, halibut, herring, monkfish and catfish, contributed the most to the dietary intake of PCBs, dioxins, and the brominated flame retardants PBDEs and HBCD in the study population, which included consumers of both average and large amounts of fish. For several perfluorinated compounds, the combined intake of fish and shellfish was the largest contributor to the total intake. Seafood contributed on average 95% to the estimated dietary exposure to mercury.

However, seafood is also a rich source of healthy nutrients such as marine omega-3 fatty acids, vitamin D, iodine and selenium. Thus, the risks associated with contaminant intake and the benefits of nutrient intake must be balanced when developing dietary recommendations in relation to seafood consumption.

ArcRisk studies provide new insights into contaminant transfer during foetal development and further confirm that health risks associated with contaminant exposure are greatest among certain vulnerable groups, especially the developing fetus and very young children. Contaminants can be passed from the mother to the fetus through the placenta and to young children during breastfeeding and affect critical stages of development. Laboratory studies in ArcRisk looked at the transfer of several contaminants, mainly perfluorinated compounds, in newly delivered human placentas; the overall findings indicate that the foetus can be significantly exposed to perfluorinated compounds in the womb. These studies also suggest that environmental contaminants may interact with placental transporter proteins, which may affect foetal exposure to chemical contaminants. The clinical significance of these findings is still unclear.

Mother-and-child health studies in ArcRisk investigated the relationship between the diet of pregnant women and contaminant levels in their blood, finding that fish consumption was an important factor. A study of the maternal transfer of polybrominated diphenylethers (PBDEs) showed that levels in the blood from umbilical cords of newborns were clearly associated with the levels in their motherôs blood serum; this study indicated a broader range of exposure sources than fish, however, as neonates from rural areas had statistically significantly lower levels of contaminants in their blood than those from urban areas. A long-term study of the trends of PCBs and several organochlorine pesticides in breast milk from women in Central Europe has shown continually decreasing levels over the 15 years of the study.

Several health effects have been identified as a consequence of the exposure to organohalogen contaminants in the Mediterranean populations studied. Newborns are more sensitive to contaminant exposure because their organs and metabolic functions are under development. In Mediterranean cohorts, exposure to organohalogens has been related to, for example, low birth size, low birth weight, low birth head circumference, poor social behavior, increases in the incidence of attention-deficit hyperactivity disorder (ADHD), decreases in cognitive skills, overweight, and alterations of porphyrin, thyroid and liver metabolism. Significant decreases in birth weight have been associated with higher cord blood serum concentrations of DDTs and, marginally, hexachlorobenzene (HCB) and beta-hexachlorocyclohexane (β-HCH). Decreases in birth length have been related to high HCB concentrations and decreases in head circumference to increased DDT. Higher concentrations of β-HCH have also been significantly associated with higher levels of thyroid-stimulating hormone at birth. Studies of low-level mercury exposure have demonstrated no significant (or only minor) effects on child development by the age of 18 months in Mediterranean countries, and higher fish consumption during pregnancy was associated with better cognitive and language developmental performance.

A toxicokinetic model describing relationships between intake of contaminants, accumulation in body tissues and fluids, excretion and effects was developed and used to evaluate and extrapolate long-term monitoring data from human biomonitoring. The model provides insight on the lifetime of PCBs in the human body and how long it takes for highly exposed population groups to reach body burden levels below accepted risk levels. The model was also used to calculate average intake based on body burdens to allow comparison with reference doses of PCBs and for risk assessment.

The overall conclusion of the biomonitoring and toxicokinetic modelling for the chemical contaminants studied in the project is that no negative effects on human health are evident for general population groups but that evidence exists for negative impacts in sensitive groups such as the foetus and young children in both the Arctic and the Mediterranean regions.

The influence of climate change on fish supply is complex and unpredictable. It is a serious concern that climate change will cause changes that affect the supply of traditional food for people living in the Arctic. Marine mammals, fish and terrestrial animals have been central to the survival of Arctic indigenous peoples and eating them has also helped to avoid many of the health risks associated with a more European lifestyle such as coronary heart disease and diabetes mellitus. However, population groups in Arctic areas that consumed large amounts of marine mammals could at the same time be exposed to high levels of contaminants. Although the work in the project did not specifically include the issue of dietary guidance, any future guidance to Arctic indigenous populations in relation to chemical contaminants in traditional foods should be nuanced and sensitive to the fact that a diet based on a wise and balanced choice of traditional foods is nutritionally more complete and healthier than a market-based diet. Advisories to reduce the consumption of specific food items to avoid contaminant exposures have been shown in some cases to result in a complete avoidance rather than reduced consumption of a particular food, thereby also reducing the benefits of these traditional food items. Thus, care needs to be taken that advice to limit consumption of fish and seafood does not inadvertently do more harm than good by reducing the consumption of foods with health benefits and increasing the consumption of alternative foods that have potential health risks. However, recommendations should also include warnings about the intake of certain species known to have high concentrations of environmental pollutants. This is especially important for vulnerable groups including women of child-bearing age, pregnant women and young children.

Healthy diets play a key role in the prevention of cardiovascular disease, cancer, diabetes and other non-communicable diseases. At present, the nutritional transition from traditional diets to ówesternô diets including more processed, energy-dense, and unhealthy food is a larger threat to the health of Arctic populations than the potential increase in environmental contaminants following global warming.

Studies of human health outcomes in relation to contaminants are seldom conclusive and it is difficult to link health effects to specific contaminants. If cohort studies addressing health effects of contaminants were conducted according to agreed protocols this would increase their suitability for meta-analyses. Over the past few decades, many studies have been conducted that attempt to relate concentrations of contaminants such as mercury and organic chemicals in human blood and other tissues to health effects. These studies, conducted in many countries for different purposes and using various methods, have yielded interesting results but rarely give a clear picture of health-effect relationships. In the ArcRisk project, special analyses termed meta-analyses were carried out on a complete and unbiased collection of published results of studies of contaminant levels and their effects in exposed populations. The meta-analysis on sex ratio vs. PCBs did not indicate that parental exposure to PCBs has any effect on sex ratio of their children, while the meta-analysis on birth weight vs. PCBs showed that a high maternal PCB concentration is probably related to low birth weight. However, a doseï response relationship with statistical significance could not be constructed based on this meta-analysis.

This systematic assessment and evaluation of published research on several health effects in relation to PCBs revealed large discrepancies and deficiencies in reported methodologies and data presentations, such as different measurement scales of explanatory variables across studies and a lack of required information in the study reports.

In summary, POPs and mercury have their most significant effects on the developing foetus, on children, on women in the reproductive age and, following lifetime exposure, on the elderly. It will be essential to follow both the trends in human exposure levels to POPs and mercury in the Arctic and the effects of these contaminants in those subgroups most at risk.

2 Implications regarding the influence of climate change on the transport of contaminants to the Arctic and their transfer in the environment ultimate to animals consumed by humans

A set of models describing emissions, transport, transformations, and bioaccumulation of selected POPs has been applied to assess how climate change will affect environmental concentrations and thereby exposure and human health effects under scenarios of climate change. The model results provide insight into changes in the long-range transport of POPs and their concentrations in various environmental compartments, but no firm conclusions on future human health impacts could be drawn. The results suggest that other indirect effects of climate change, such as changes in food-web structure and changed availability of traditional foods, can potentially have a larger effect than the direct effects of increased temperature and changing patterns of precipitation, wind, etc.

All modelling tools employed in ArcRisk showed that climate change can affect future levels of concentrations of organic contaminants (both increase and decrease) when compared to baseline ñno climate changeô scenarios, with projected changes around a factor of two (i.e., from a halving to a doubling of concentrations). The models typically considered the effects of climate change on temperature and precipitation, as well as on air and ocean currents. However, changes in organic carbon cycling, catchment hydrology, land use, extreme events (which are becoming more frequent) and ice cover, which are more complex to model and have important effects on contaminant levels, were often not considered or were modelled in a simplified way.

Increasing temperature under a future climate will cause a shift in the environmental distribution of contaminants. It is expected that with rising temperature, POPs deposited in sinks, such as water and ice, will revolatilize into the atmosphere, which may cause increasing levels of some chemicals in air and decreasing concentrations in surface media. These increasing concentrations in air are expected to be offset, however, by decreases owing to expected reductions in emissions of regulated chemicals. For newer or ñemergingô organic contaminants which are affecting the Arctic, some of which possess very different properties compared to the ñlegacyô POPs, their behaviour may be quite different. Reduction in the amounts of snow and ice will serve to release these compounds to meltwater rather than re-release them back to the atmosphere. Detailed experimental studies of the behaviour of selected POPs in the snow-ice-water system in the Arctic illustrated impacts on POPs loading of water during the melting of ice and transport from glaciers to surface waters; thus, a warmer Arctic will affect these processes. Different chemical contaminants behave differently during ice melting and will be transferred to the surface water in a sequential manner based on specific chemical properties.

Increasing temperature in the Arctic area in the past decades has resulted in Arctic sea-ice retreat and snow melt. Changes in sea-ice cover and thickness have been modelled in the Arctic Ocean. A shift from multi-year sea ice to first-year sea ice under a future climate is expected to result in more efficient delivery of contaminants from melting ice into the underlying surface water, thus altering contaminant dynamics and possibly increasing concentrations in beneath-ice seawater in the surface Arctic Ocean.

The effect of climate change on the atmospheric deposition of mercury in the Arctic was investigated in another modelling study. Under climate change scenarios, there was a significant decrease in mercury deposition over the Arctic Ocean and a significant increase over the continents. The changes in mercury deposition are driven by changes in the atmospheric chemistry of mercury that result from reduced sea-ice cover and increased concentrations of ozone in the troposphere that are forecast under the climate change scenario.

A comparison with the Baltic region showed that the effect of climate change on the fate and transport of organic contaminants in this region was similar to other regions previously modelled, including the Arctic (i.e., projected changes around a factor of two in either direction). The state-of-the-art contaminant fate modelling tool for the Baltic region was identified to be overly simplistic to accurately model the effects of climate change on the fate and transport of organic contaminants in this region.

In Arctic populations that rely on traditional and local foods, the influence of climate change on contaminant levels in foodstuffs is expected to be small relative to future changes in dietary behavior. Contamination by PCBs and other legacy POPs can be expected to decrease over time as a result of actions that have been taken to reduce their emissions. Climate change may, however, influence the rate at which these reductions translate into decreases in environmental contamination. For example, temperature-controlled degradation may accelerate the breakdown of POPs in the environment. Conversely, temperature-driven re-volatilization may enhance re-mobilization of contaminants from soils and waters in response to warming. These climate change effects can vary on a chemical-to-chemical basis. For mercury, climate change is also expected to alter some of the basic environmental transport and transformation processes.

ArcRisk model results indicate that the influence of climate change on the fate of PCBs (and other POPs) in the physical environment will be relatively small, and are unlikely to counteract the decreasing emissions. The implication is, that by 2100, environmental levels of these contaminants will be far below their present levels, and on this basis, human exposure to PCBs and legacy POPs should also be much lower. However, climate change is also likely to cause changes in ecosystem structures. At present it is not possible to predict how this may affect food-web bioaccumulation and biomagnification, or what the implications of this may be for contaminant levels in fish and other species traditionally consumed by some Arctic populations.

Future risks to people associated with dietary contaminant exposure in the Arctic will, however, depend to a large degree on changes in diet and food supply. The results from ArcRisk show that based on current predictors of contaminant exposure, the influence of climate change on future contaminant exposure will to a large degree depend on the consumption of fish and other seafood. Future dietary exposure to contaminants may be influenced by climate change-related alterations in the distribution and availability of traditional foods, such as seals and fish, which cannot be predicted at this time.

3 ArcRisk science-based research policy recommendations

The assessment tools developed and used in the ArcRisk project with databases, models, and monitoring systems form a good basis for further quantitative analysis of impacts of climate change on variations in human health exposures through changes in contaminant origin, transport, fate and behaviour in the environment and uptake and transfer through relevant food chains. This analysis would need new input from future research, including the following:

1. Better characterization of primary and secondary sources of POPs and more accurate quantification of current and future releases of POPs from these sources are needed for prediction of environmental exposure to such contaminants and interpretation of monitoring data. More information is needed on transformation products of POPs that may be formed under climate change and on their impact on the health of ecosystems and humans. Assessing the input of newer POPs to the Arctic via environmental monitoring programs is required and research is needed on the effect of changing amounts and distributions of sea and land ice and snow on their fate and subsequent uptake into food chains.
2. Further improvement of fate and transport modelling in the physical environment is needed in order to consider in the models not only the direct effects of climate change (e.g., changes in temperature, ice and snow cover, precipitation, wind speed and ocean currents) on contaminant fate and behaviour, but also indirect effects, e.g., alterations in carbon cycling, food webs, catchment hydrology, land use, vegetation cover, etc.
3. Improved modelling of contaminant bioaccumulation in ecosystems is needed to reduce the large uncertainties in quantification and evaluation of human exposure to contaminants in the Arctic. More effort is needed to generate datasets in which multiple species and abiotic media are sampled at the same time and analysed for organic pollutants.
4. Long-term environmental monitoring of POPs listed under the Stockholm Convention and measurements of concentrations in human breast milk and blood plasma are needed. In addition, monitoring of social changes, such as human dietary changes, is also necessary in order to determine possible changes in human exposure.
5. More information is needed on the human health effects of more recently identified environmental POPs, such as per- and polyfluoroalkyl substances, PBDEs, hexabromocyclododecane (HBCD), and other substances with POP-like characteristics, particularly regarding effects on very young (including the foetus) and elderly subgroups of the population.
6. Based on initial results of meta-analyses and reviews of health outcomes in relation to POPs and mercury conducted in the ArcRisk project, it was concluded that currently published studies include too many variables to carry out coherent meta-analysis, which makes it difficult to estimate risk for human health. Harmonization of the data from large European bio-monitoring projects and AMAP may improve the control over such variables. The current ArcRisk database format, to be hosted by AMAP, will be able to hold such information.
7. New research should be undertaken to quantitatively assess the relationships between climate change impacts, changes in fish distribution, and mercury contamination. This is a drawback in current analysis of climate change impacts on human health in the context of mercury contamination in the Arctic.

4 Strategies to reduce human exposure to environmental contaminants

Avoiding future adverse effects on human health from exposure to contaminants requires an overall strategy that integrates policies and measures to reduce use and releases of contaminants, monitoring of environmental levels and human exposure, education and risk communication, and where necessary food consumption advice to critical groups. ArcRisk objectives include providing input for the development of strategies on how best to adapt to and prevent adverse health effects related to climate-mediated changes in exposure to chemical contaminants in human populations in the Arctic and in Europe. The research performed in the project has illustrated the complexity of contaminant cycling and effects and also the limitations in our capability to predict how climate change may affect these processes. Policy advice related to reducing future exposure and effects cannot therefore be formulated in a single straightforward statement but rather comprises a series of recommended actions that need to be incorporated in an overarching strategy to meet this situation.

- **Policies, restrictions and technical measures to reduce emissions of contaminants.** Long-term, comprehensive reductions in human exposure to harmful environmental contaminants can only be achieved by minimizing their use and eliminating as far as possible releases to the environment. For some contaminants, climate change may increase exposure, but climate change mitigation alone will not remove risks associated with chemical contaminants. European policy-makers need to act with industry stakeholders to further develop and strengthen implementation of EU directives concerning chemicals currently used in society, and to complement them by voluntary measures. This includes promoting climate and energy strategies that maximize co-benefits of reducing emissions of greenhouse gases and unintentionally produced POPs and mercury from the same sources. Europe can lead the way, raise global awareness, and build capacity to take advantage of these potential co-benefits.
- **International collaboration**. Given the vast amount of chemicals used in society and the number of chemical compounds involved, the task of identifying those chemicals that pose the greatest risks for human and environmental health is enormous, expensive, and requires international collaboration. Risk attribution is further complicated by potential interactions among the various contaminants in the human body. In addition, risk reduction measures must be adapted to the specific use and emission patterns associated with different groups of chemicals. The potential for long-range transport of many harmful contaminants means that international agreements such as the Stockholm Convention, the Convention on Long-Range Transboundary Air Pollution, and the Minamata Convention have a key role to play. In addition, potential synergies between existing international policies and agreements concerned with monitoring, risk assessment, and mitigation measures should be exploited. For legacy substances that are already banned or restricted, additional measures to reduce environmental dispersion from remaining sources, contaminated sites, landfills, buildings, etc., may be warranted.
- **Access to information**. Information on emissions, use and life-cycle of environmental contaminants and potential contaminants is largely lacking and improved access to such data is necessary to allow risk assessment and prioritization of measures.
- **Monitoring and assessment.** Monitoring of contaminants in the environment, foodstuffs and humans provides essential information to follow up the effectiveness of actions taken, to identify new threats and to develop the basis for guidance aimed at reducing exposure. Monitoring in combination with modelling and assessment can also provide knowledge on contaminant pathways in the environment which can be used for predicting future changes and identifying measures to be taken. Monitoring aimed at environmental compartments that are expected to be affected by climate change (snow, ice, glaciers) and that may increase availability of contaminants for uptake in food webs is especially important.
- **Future use of human biomonitoring data.** Existing human biomonitoring data representing individual internal exposure provide an excellent opportunity in long-term follow-up studies to pursue potential association of blood levels of contaminants with health outcomes in these populations. Umbilical cord and maternal blood levels give the possibility to link fetal exposure to later health effects according to the new concerns regarding developmental origin of adult onset diseases including cardiovascular diseases and cancers.
- **Advisories on human consumption of fish, seafood and mammals.** Many contaminants are persistent and continue to contaminate the environment for a long time after their sources have been reduced or eliminated. Food consumption guidelines and advisories can provide effective short-term solutions to minimize human exposure, in particular for critical groups. However, such

advisories must be developed in close cooperation with local health professionals and the target groups themselves, taking account of both risks and benefits of consumption of particular foods, including indirect benefits (such as cultural benefits). It will be necessary to update and complement existing food consumption guidance as knowledge about the effects of chemical contaminants and the associated impacts of climate change grows.

Specifically with regard to mercury pollution, it can be stated that:

- reductions of Hg emissions from anthropogenic sources worldwide would need to be introduced as soon as possible to ensure decreasing the adverse impact of climate change on human health in the Arctic. The forthcoming new UNEP global Minamata Convention on Mercury to reduce emissions is expected to contribute substantially to this reduction;
- new methods need to be developed for assessment of the implementation of emissions reduction impacts, including models integrating Hg transport within the abiotic and biological compartments of the environment, dose-response functions for Hg in the Arctic populations, and cost-benefit analyses for selecting the most efficient emission reduction measures; and
- new research should be undertaken to assess quantitatively the relationships between climate change impacts and changes in fish distributions and mercury contamination.

A number of strategies for adaptation and abatement for POPs and mercury in the Arctic have been elaborated by ArcRisk scientists and other research groups for UNEP and other international fora, such as the Stockholm and Minamata Conventions or European Commission units. The research within the ArcRisk project has further contributed to these strategies, confirming the need for implementation of Best Available Technologies (BAT) and non-technological measures to abate POPs and mercury, and the need for implementation of Best Environmental Procedures (BEPs) in the case of adaptation strategies.

5 Recommendations for future research and monitoring

The potential threat of exposure and health effects in some population groups calls for continued action and the complexity of this issue warrants further research and monitoring to provide scientific support for the development of health advice, mitigation measures and policies. ArcRisk research has led to a number of recommendations for future research and monitoring:

- To allow comparisons, generalization, and assessment over larger population groups as well as for future scenarios, harmonized and standardized methodologies for human health cohort studies are needed. The main components of this harmonization should include methods for identification and description of health endpoints, collecting and reporting data, and guidelines for taking into account confounding factors and statistical reporting of the findings.
- Toxicokinetic models need further development for better exposure characterization, especially in the case of results and concentrations in many matrices (mother milk, urine, hair, blood, etc.). The CDI (total chronic daily intake) can be a very useful key parameter in these case studies.
- Human biomonitoring studies of POPs can be used together with the expected exposure routes to parameterize toxicokinetic models. These models can then be further used for other similar chemicals or to evaluate effects of changes in exposure (e.g., change in the diet). This should be viewed in the context of the risk posed by contaminant mixtures (e.g., the burden of both legacy and emerging contaminants together) with a shift away from single substance assessments.
- Monitoring of contaminants in different environmental compartments such as air, water, and biota should be designed and maintained to provide information on temporal trends and geographical distributions of chemical contaminants.

- For emerging chemical contaminants such as per- and polyfluoroalkyl substances (PFASs), systematic observations are required, particularly in the marine environment relating to their occurrence in seawater and fate processes, e.g., the transfer of chemicals from air or from meltwater runoff and subsequent uptake into biota.
- A terrestrial deposition network to complement long-term air monitoring is required to assess the atmospheric input of new chemical groups over different seasons.
- Climatic aberrations are a growing feature of a warming Arctic, marked by an increased frequency of erratic weather events such as winter periods of unseasonably warm air temperatures that result in rapid thawing of the winter snowpack followed by ice formation or encasement once temperatures decline again. The effect of these events on contaminant fate in the snowpack and subsequent meltwater runoff is unknown and needs further research to assess risks for óshockô loading of an array of contaminants to terrestrial tundra surfaces and freshwater systems. The potential for the remobilization of contaminants previously deposited and stored in perennial snow and ice is high and a more thorough systematic assessment of these processes is required.
- Process-based studies are required in the marine cryosphere to examine chemical enrichment in sea ice and to quantify the role of sea ice as a source of chemicals to ice-associated biota, as well as to understand how ice-brine and the age of the ice affect chemical accumulation and fate.
- The use of enantiomers of chiral substances as markers of metabolic processes in marine biota is promising and could be used to distinguish climate change-related perturbations on specific marine food webs. More studies using this technique are needed.
- The influence on the concentrations and profile of contaminant residues in traditional, country foods by processing them prior to human consumption needs further assessment. The local influence of coastal Arctic towns on contaminant levels in locally caught marine biota also needs further investigation, particularly for chemical substances such as PFAS and other emerging substances that may arise in coastal fjord systems through local activities or runoff in addition to long-range transport processes.
- The various state-of-the-art multimedia environmental fate models used in the ArcRisk project have been shown to be capable of providing invaluable information when focus is on the direct effects of global climate change (GCC), e.g., changes in temperature, ice and snow cover, precipitation, wind speed and ocean currents. These modelled changes are anticipated to have small impacts on environmental concentrations of chemical contaminants relative to potentially important indirect changes, which can include alterations in carbon cycling, catchment hydrology, land use, vegetation cover, chemical emissions, etc. Our understanding of processes related to GCC-induced indirect effects is very limited and associated with large uncertainties which make those processes more challenging to be assessed.
- Information for quantifying, evaluating and improving food web models is often insufficient or associated with large uncertainties. This insufficiency is usually caused by the lack of good monitoring datasets for multiple species and the abiotic media. More effort is needed to generate datasets in which multiple species and abiotic media are sampled at the same time and analysed for organic contaminants. These more complete datasets will allow us to understand and more accurately model bioaccumulation in Arctic food webs. More basic research is also needed on how food webs and ultimately human diets will themselves respond to GCC.
- Finally, the Arctic is undergoing unprecedented changes through climate change allowing exploitation through new opportunities in areas such as shipping, mining, fishing, tourism and population change. These will bring new, possibly localized, pollution issues and scoping studies are required to assess the type of pollution and its risk to wildlife and the Arctic environment.