From Pole to Pole

Roland Kallenborn *Editor*

Implications and Consequences of Anthropogenic Pollution in Polar Environments

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Foreword

The International Polar Year 2007–2008 was the largest joint Polar research activity ever performed. Thousands of scientist participated and thousands of papers have been published, with some still on their way. Although pollution of the Polar regions—the environment and in the Arctic, its indigenous and non-indigenous populations, was not a priority area for the IPY, some projects did cover these important topics and these have provided important new knowledge. This book summarizes some of this new knowledge.

Pollution of the Arctic and Antarctic areas by chemicals originating from industrial, agricultural and transport activities in the tropics and at mid-latitudes has been documented since the 1980s; however, huge gaps in knowledge exist about these pollution sources, and related contaminant transport pathways, bioaccumulation, and biological effects on ecosystems, animals and humans. The IPY projects presented in this book address some of these gaps.

The atmosphere, rivers and ocean currents are the three main routes for transporting pollution as well as energy. The atmosphere is by far the fastest "highway" to the Polar regions. Two chapters in this book (Kallenborn et al.; Skov et al.) provide the reader with a good overview of the role of international scientific cooperation in documenting and explaining how atmospheric transport of contaminants such as persistent organics and heavy metals occurs. These chapters also provide new results obtained from related IPY projects documenting, for example, the contribution from sources in Southeast Asia, increasing understanding of some of the process involved in long-range transport, and discussing the transformation of the chemicals and their deposition in Polar regions.

Two other chapters (Shevchenko et al.; Grannas) describe processes involving the atmosphere and snow and ice. Snow and ice provide not only a temporary storage for particle associated and gaseous chemicals but also a means by which they can be transported. It also plays a central role in some photochemical process that can transform organic pollutants.

Black Carbon (BC) is a pollutant that affects human health, especially in indoor environments. It also contributes to climate change by absorbing heat while present in the air or when deposited on snow and ice. Some sources of BC around the world contribute to increasing temperature—also in the Polar regions. Sources of BC exist within the Arctic, including the emissions from industrial activities in northern Siberia. Chapter 10 also provides an overview of the observations and analyses made in Siberia and adjacent Arctic seas.

Persistent organics and mercury are contaminants that accumulate and in some cases bio-magnify in the marine food chain, from plankton, through fish and marine mammals, up to humans. Two chapters (Vorkamp and Muir; Dietz et al.) describe this bioaccumulation of environmental contaminants in the Arctic food chain, focusing on ringed seals and polar bear, the two key species in the Arctic marine ecosystem that are also used and as traditional food by some Arctic indigenous peoples. The chapters provide the reader with a good introduction to the subjects of contaminant bioaccumulation metabolism and toxicokinetics. They also present information on contaminant geographical and temporal trends, and biological effects of single compounds and mixtures on these animals. Concerns about new chemicals entering the Arctic with potential to further contaminate food chain are also discussed.

At the top of the Arctic marine food chain, we have Arctic indigenous peoples who for thousands of years have survived in the region by harvesting marine resources as part of their traditional diet. This diet is rich in nutrients, energy and vitamins, but in recent decades has become contaminated by chemicals that are seldom used or discharged in the Polar region. Since the early 1990s, several studies have investigated human health impacts of contamination of traditional foods in some Arctic areas and these topics are covered in two chapters in the book (Hansen et al.; Gomes and Roche). This material informs the reader about new results and new concerns—especially regarding some specific human health effects—and discusses the effectiveness of actions taken at local and global scales to address this important issue. In discussing the connection between food, diet, nutrition, contaminants and human health, Hansen et al. illustrate how risk communication must be addressed in a responsible manner to avoid unnecessary negative impacts on nutritional benefits, including post-natal nursing behaviour.

The final three chapters discuss a range of related subjects, including the effects of globalization on Arctic indigenous peoples based on investigations conducted in Greenland prior to the IPY (Sowa), security and the involvement of schoolchildren in sampling and analyses for environmental contaminants (Heimstad et al.). They illustrate the changes in the Greenlandic society over the last 40 years, from small hunting/gathering communities based on local food, to larger townships where western (Danish) food is increasingly found in the stores. The importance of local food as a significant source of nutrients and as a central part of traditional culture is discussed by Sowa, and in this context, the chapter also discusses the impacts of pollution and climate change.

The engagement of the youth in sampling and analyses of contaminants is important not just to stimulate recruitment of 'new Polar scientists'. Even more important is the role that such engagement has in transferring knowledge and understanding about how pollution from human activities within and outside the region can impact Polar areas and their unique ecosystems, and what this means for their human populations. More than 50 schools from 13 countries took part in the exercise that provides an example for similar educational activities, which should be conducted not only in Arctic communities but also all over the world.

The chapter written by Hoogensen Gjørv et al. introduces the subject of security, from military security to human security, viewed from a Northern perspective. It addresses energy and environmental security—including food and water security for Arctic indigenous peoples—and themes of increasing importance in a world where large groups of people are and will continue to migrate due to war and climate change.

This book is highly relevant for readers who would like to learn more than just the headline news. For the scientific community working on Polar ecosystems and for people living in the Arctic, the chapters provide important new information and insights. The impacts of pollution and climate change on Polar regions and world as a whole will continue for many years to come. Sound science is vital in order to underpin actions that need to be taken at global, regional and local levels. This book contributes to this science, but more is needed.

Enjoy your reading.

Lars-Otto Reiersen
AMAP Executive Secretary

Letter from the Editorial Team

The first two International Polar Years both failed to coordinate and distribute their assembled data adequately and to ensure its proper analysis, resulting in a less than satisfactory legacy from what had been considerable international efforts. Recognizing this, the participants in the Third International Polar Year (International Geophysical Year) made extensive plans to ensure that their contributions would be both accessible and used, establishing the World Data Centres as a major new initiative. In the early preparatory stages of the latest International Polar Year (IPY 2007–2009), the importance of providing the legacy of this demanding international research effort was made clear, with priority being given to planning for a well-organized dissemination and coordinated publication of the results, data evaluations and scientific findings. It was with this in mind that we proposed our publication project (IPY Project No. 79) in the form of the book series "From Pole to Pole: Environmental Research within the International Polar Year 2007–2009". With over 50,000 scientists involved in a myriad of projects, there was an obvious need for a guide to the principal findings and the key papers within environmental science fields.

The "From Pole to Pole" book series was conceived as a comprehensive publication framework for the documentation of environmental research activities performed during the IPY period. The book series is not intended to be a typical collection of original scientific project publications/chapters in the form of standard monographs. It is rather a bibliographic, a science-based information source and a starting point for interested scientists and public to access the summary information on the specific environmental research topics within the IPY activities. The volumes will provide scientifically sound general information on the concepts, findings and scientific motivation for the various relevant research activities and will direct the interested reader to more detailed scientific papers, web-based information and other publications which will provide the detailed data and their analyses. These compilations of citations and references within the book volumes will be important milestones for the assessment of progress in each area, and the scientific significance and value will grow as the series develops.

The volumes will also be available in e-book format which will allow continuous updating of references and information sources (including internet pages and databases) by the editorial team on an annual basis, thus keeping the works topical as living reference sources. It is expected that this documentation will provide a comprehensive picture of most of the environmental research performed within the IPY framework.

After the now ending initial phase, the "From Pole to Pole" book series will be transformed into a legacy component of IPY by widening the concept of the book series towards general coverage of recent scientific developments in environmental Polar research (Arctic and Antarctic).

With the initial published volume on the history of the International Polar Years (edited by Susan Barr and Cornelia Lüdecke), our concept has finally begun to be realized, and has been followed by the completion of other volumes. Marine biology has provided two volumes on "Adaptation and Evolution in Marine Environments—The Impacts of Global Change on Biodiversity".

The fourth volume for the "From Pole to Pole" book series "Implications and Consequences of Anthropogenic Pollution in Polar Environments" deals with anthropogenic pollutants, their environmental, toxicological and societal implications in the sensitive Polar environments. Many of the projects and chapters described here have developed into long-term commitments since IPY, and thus illustrate the impact this relatively short period of IPY research (2007–2009) is still having on the present international research activities in Polar environmental pollutant research.

The book project describes the development, continuation and scientific impact of 11 research projects initiated or associated with IPY. Scientific aspects and research in sociology, anthropology, chemistry, physics, medicine, human toxicology, glaciology, education and many more are covered, illustrating once again the strong interdisciplinary character of the IPY research endeavours. In total 68 authors contributed to this book representing a significant fraction of environmental scientists involved in this field of IPY research.

We thus congratulate the editor and the authors not only for their outstanding efforts to document all aspects of the research on anthropogenic pollutants undertaken during the recent IPY but also for developing these research priorities further in the present context of Polar environmental pollutants.

Ås, Norway	Roland Kallenborn
Naples, Italy	Guido di Prisco
Oslo, Norway	Susan Barr
Cambridge, UK	David Walton
January 2016	

Acknowledgement

The book project "Implications and Consequences of Anthropogenic Pollution in Polar Environments" had developed into a long-term commitment to which many colleagues, friends and family contributed in their own way. Without their support, understanding and active help, the book would not have been possible.

I, therefore, thank all authors of the 11 scientific chapters for their patience, their understanding and commitment which was highly essential for bringing this demanding project to a good end.

My work with the book was supported economically by my home institutions, the Norwegian University of Life Science and the University Centre in Svalbard (UNIS), allowing me to spend time apart from teaching and research on this book project. All my colleagues in the "chemistry group" at NMBU are thanked for their patience and understanding in times when I was not as accessible as I should have been during my daily work at the institute.

The Arctic Monitoring and Assessment Programme (AMAP) actively contributed advice as well as funding to this project. Thank you Lars-Otto Reiersen (AMAP secretary), Simon Wilson and Janet Pawlak for your support during the 2 years it took to complete this book project.

The project "Combined effects of Climate Change and Pollutant in the Arctic" funded by the Nordic Council of Ministers (NCM) as well as the FP7 project "Impacts on health in the Arctic and Europe owing to climate-induced changes in contaminant cycling" funded part of my work on this monograph.

Finally, but not least, I wish to express my heartfelt thanks to my family, Berit, Magnus and Mia who had to live with me through the times when I was completely occupied with the completion of this project, in addition to my other numerous duties at the institute and abroad. Thanks for your patience, understanding and keeping up pace with me during challenging times.

Roland Kallenborn

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About the Editor

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Chapter 1
Background

Roland Kallenborn

The International Polar Year 2007–2009 has been jointly established and organized by the International Council for Science (ICSU) and the World Meteorological Organization (WMO). The IPY 2007–2009 was also initiated as a marked follow-up of the previous IPY event: the International Geophysical Year 1957–1959, 50 years ago (Barr and Lüdecke 2010).

The coordinated efforts under IPY 2007–2009 are today acknowledged as one of the largest and most ambitious joint research efforts ever attempted by the international science community on research collaboration in the polar regions of the world. The coordination and priority strategies for selecting project initiatives for funding were in the hands of the nations joining IPY and their respective funding organizations. The coordination of the funded research, communication, education, and public relations were organized through a dedicated secretariat and program office (IPY International Programme Office; IPY-IPO, lead by Dr. David Carlson). In many countries, dedicated IPY research programs were established aiming at coordination the national IPY-related projects with international IPY activities.

From the early beginning, environmental research in all its facets was considered an important and priority research field for IPY 2007–2009 as stated by the international IPY secretariat in the initial documentation including the IPY Science Plan. Many coordinated research projects associated with environment-related research were conducted and funded under the frame of IPY 2007–2009. The IPY honeycomb chart, the visualization of all projects supported by IPY (http://ipy.arcticportal.org/images/uploads/linkedHoneycomb.pdf), identified as much as 60 endorsed project initiatives within environmental pollution research officially endorsed under the auspice of IPY (from a total of 231 project proposals according

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to Krupnik et al. 2011). From this list of 60 endorsed projects, almost half of the lists (25 endorsed projects) were proposed on topics related to atmospheric pollution issues. This is also reflected in the here presented book volume where almost half of the chapters deal with atmospheric pollution issues and associated research topics.

Implications of the International Polar Year (IPY) 2007–2009 for Research on Persistent Organic Pollutants (POPs)

Already from the early beginning, IPY 2007–2009 was considered an important potential research platform for coordinated Arctic–Antarctic research on pollutant fate and distribution (uptake and transformation). As important research focus within this broad scientific field, persistent organic pollutants (POPs) have been identified and investigated in the polar regions for almost half a century earlier. Therefore, the current science-based information status was considered as a good working platform for innovative interdisciplinary research on already known POPs ("legacy" priority POPs) as well as POPs of emerging concern (newly identified Arctic pollutants).

However, in light of the observed funding and research priorities leading to the finally conducted research projects during IPY, these expectations were obviously much to ambitious, and the international IPY funding did not provide substantially increased national funding priority for the suggested and endorsed projects within environmental pollutant research required and expected to facilitate the endorsed research program in organic pollutant research as documented in the IPY honeycomb chart.

Under IPY, only three project initiatives were directly funded from dedicated national IPY programs, with atmospheric POP distribution patterns and pathways as main research topic:

1. Fate, uptake, and effects of contaminants in the Arctic and Antarctic ecosystem (COPOL), funded by Norway (IPY 175).
2. INterContinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA), funded by Canada (IPY 327).
3. Greenland atmospheric pollution as a part of the International Arctic Systems for Observing the Atmosphere, funded by Denmark (IPY 196).

However, many research initiatives conducted on pollutants in the polar environment were indirectly initiated and/or jointly conducted in combination with IPY-funded research, although not directly funded from IPY programs. In many cases, also joint infrastructure was used or research priorities were tightly coordinated, leading to IPY-associated long-term programs with a considerable legacy component included for the long-term scientific commitment even with an implication for today's research priorities on organic pollutants in the pristine polar environments. Several examples of these research activities are highlighted in the here presented book in order to complete and complement the picture drawn of the research efforts undertaken during IPY 2007–2009 with a focus on environmental pollutants with an emphasis on organic pollutant research in the polar regions during IPY.

Format, Structure and Concepts

As earlier stated, from a total of 231 IPY-endorsed proposals, around 60 proposals were associated with pollutant research. From this endorsed IPY project portfolio, around 15 projects related to pollutant research (covering all the aspects of anthropogenic pollution) in the polar environments (including human populations) were funded, with 3 funded projects on persistent organic pollutants (POPs).

The aim of our book is to provide a first view on the direct and indirect achievements of the International Polar Years, as unique incentive for pollutant research in the polar regions. The here selected chapters will provide representative reports on the concepts, findings, and implication of pollution research performed under direct IPY-related funding or indirectly supported by IPY through infrastructure support or cobenefitting from ongoing IPY activities in the period 2007–2009. The bibliography of the research activities described herein will be continuously updated.

Both conventional project descriptions and overarching research concepts are represented in the chapters in order to illustrate the diversity of the scientific strategies' pursuit for environmental pollutant research during IPY.

The interdisciplinary of the here-documented integrated research is demonstrated by the implementation of findings spanning a variety of disciplines from analytical chemistry to social sciences and fate modeling.

Chapters 2–5 cover research activities and findings associated with the abiotic polar environment. Both transport processes in the regions (Chap. 2; Kallenborn et al. and Chap. 3; Skov et al.) and intraregional distribution processes including ice-associated transport (Chap. 5; Shevchenko) were investigated and reported here. The role of the cryosphere (ice-associated environmental compartment) as an important environmental feature of the polar regions was comprehensively investigated during the program period. As an important transformation process for all types of pollutants (anthropogenic as well as biogenic), photochemistry as an important seasonal feature is highlighted in a dedicated chapter (Chap. 4; Grannas).

A large section of IPY was dedicated to education and outreach activities. However, only one IPY-endorsed project was funded, dealing with educational concepts for introducing pollution aspects on high school level. This project is summarized in Chap. 6 (Heimstad et al.).

The “human component” of the book is also covered in Chaps. 7–10 from a scientific point of view. Since permanent circum-polar human indigenous populations only exist in the North, these aspects therefore focus entirely on the Arctic region. Research and assessment of pollutant impact on indigenous people of the North are channeled through the Arctic Monitoring and Assessment Programme (AMAP), a working group of the Arctic Council. Therefore, a comprehensive overview on health aspects in Arctic populations is provided based on the recent AMAP Human Health Report (Chap. 7; vanOostdam et al.). The specific effects and consequences of exposure to endocrine-disrupting chemicals (hormone-mimicking compounds) are summarized in Chap. 8 (Gomes and Roche). In addition, social aspects and the role of North indigenous people in a globalizing world are discussed in Chap. 9 (Sowa). Overarching societal aspects on Arctic-related “human security” issues are elucidated both from a social scientific and from a geopolitical perspective using petroleum exploration as a typical example of Arctic pollution. These scientific aspects are discussed in Chap. 10, concluding on the IPY GAPS project (Impacts of Oil and Gas Activity on Peoples of the Arctic using a Multiple Securities Perspective; Hoogensen Gjørv et al.).

The final chapters (Chaps. 11 and 12) deal with distribution, uptake, and effects of anthropogenic pollutants in Arctic marine biota with a focus on top predating marine mammals. Chapter 11 summarizes the findings of the IPY BearHealth project (Dietz et al.), and Chap. 12 represents a comprehensive review of different studies during the IPY period where the circum-Arctic ring seal populations were evaluated and investigated with a focus on persistent organic pollutants.

Chapter 2
Research and Monitoring of Atmospheric Persistent Organic Pollutants (POPs) in the Polar Atmosphere

Roland Kallenborn, Hayley Hung, Tom Harner, Pernilla Bohlin-Nizzetto and Susan Bengtson Nash

Introduction

Research and monitoring of persistent organic pollutants (POPs) in the Arctic atmosphere has a relatively long history within modern interdisciplinary environmental pollutant research. The first evidence of adverse effects of POP-like substances on ecosystems and higher organisms was already provided in 1962, when Rachel Carson's book "Silent Spring" summarized the scientific evidence on organic pollutants and opened the public discussions on adverse effects of POPs distributed through atmospheric transport over large distances (Carson 1962). Data on the occurrence of POPs in the Arctic atmosphere, however, date back to 1968, when Singer reported the contamination of the world's oceans by POPs including the Arctic oceans. The first campaign-based monitoring of POPs is reported in 1981 (Billings and Bidleman 1980). The first evidence of atmospheric POP transport to the Arctic based upon POPs monitoring was also reported in 1981 by the same group (Billings et al. 1981) followed by a monitoring report from Norway in 1984

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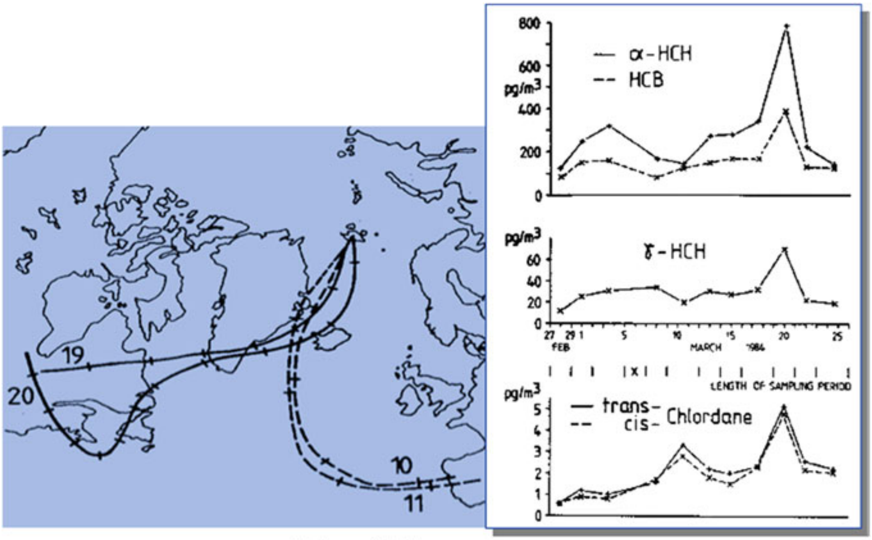

(Oehme 1991)

Fig. 2.1 Source region identification for air masses contaminated with elevated POPs. Figure modified according to Oehme and Ottar (1984), Oehme (1991) for compound abbreviation, see Table 2.1

(Oehme and Ottar 1984). For the first time, for this data set, meteorological modeling (back-trajectory calculations) was combined with POP monitoring date (Fig. 2.1).

Similarly, in 1981 and 1983, USA and Japanese research groups reported on the occurrence of POPs in the Antarctic atmosphere (Atlas and Giam 1981; Tanabe et al. 1983). In 1989, the first long-term monitoring programs provided the first convincing scientific evidence for the continuous hemispherical atmospheric long-range transport of POPs to the Arctic (Hung et al. 2010). These international research and monitoring activities were paving the way for today's international regulations on POPs usage and applications, documented in the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP) and its Aarhus Protocol on POPs (www.unece.org/env/lrtap/pops_h1.html) and the United Nations Environment Programme (UNEP) Stockholm Convention on POPs (www.pops.int).

During the initial stage of IPY 2007–2009, the atmospheric POP monitoring was mainly focusing on the Northern Hemisphere and the Arctic. However, since only campaign-based surveys on POPs in background Antarctic locations are reported earlier (Gambaro et al. 2005; Kallenborn et al. 1998), the establishment of pollutant monitoring in Antarctica was considered an important priority for POPs research in the environmental science community, especially during the initiation of IPY 2007–2009. IPY was consequently considered by the environmental experts as important initiative and opportunity to coordinate and harmonize the current efforts on global research and monitoring of atmospheric distribution processes for POPs, ultimately leading to accumulation and unwanted hazardous effects in the sensitive Arctic and Antarctic ecosystems. Therefore, a series of project initiatives were submitted and endorsed by IPY (ICSU and WMO) in order to establish an Antarctic–Arctic surveillance network based upon the already-established regional networks in the North (i.e., Arctic Monitoring and Assessment Programme, AMAP, and the European Monitoring and Evaluation Program, EMEP) and to identify knowledge gaps and future research priorities on coordinated POP monitoring in the polar atmosphere. Only one project initiative (*INterContinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic* = INCATPA) was dedicated fully to atmospheric POP research in the Northern Hemisphere. This project was funded by Canada under IPY [initially considered as an important Canadian link into the ATMOPOL network (IPY Project No. 79)]. Although several IPY initiatives on atmospheric POP research were endorsed as relevant scientific contributions by IPYIPO, science projects dealing with international research on atmospheric POPs were unfortunately not considered as sufficiently important research priorities for involved national funding authorities. However, although not directly funded by IPY, a series of IPY associated and inspired research projects were carried out under the frame of IPY 2007–2009 despite of the lack of direct funding. All IPY projects as well as associated research activities linked to IPY projects are presented and considered in this general overview of IPY-associated research activities under the IPY scientific umbrella.

Modern POPs Monitoring and Research

Modern atmospheric POPs research and monitoring in Polar remote environments has developed from a loosely mainly campaign-based linked collaborative network of national measuring stations into a tightly connected international coordinated monitoring network following jointly developed sampling routines and quality control procedures. In polar POPs monitoring, currently, the focus is still on the long-term atmospheric POP surveillance in the Arctic. However, during the past decade, the Antarctic atmospheric POP monitoring is gaining a significant role in globally monitored polar atmospheric POP monitoring. This development leading hopefully into a global grit of POP monitoring stations is especially supported by UNEP and UNEPs international convention for the global regulation of POPs (i.e., Stockholm Convention). Currently, the Stockholm convention identified a list of 26 priority compounds (isomer/congener groups), subject for international monitoring, restrictions, and regulations (Table 2.1). The below priority list represents the core group for all official national and international atmospheric monitoring programs regardless location and regions, in which the monitoring program is performed.

Table 2.1 Recent list of priority target POPs for international regulations according to the global convention on POPs regulations (Stockholm Convention); for information, see: www.pops.int

No	Compound/compound group	Pollutant group	Characteristics	Abbreviation
1	Aldrin	Pesticide	Cyclodiene pesticide	
2	Dieldrin	Pesticide		
3	Chlordane	Pesticide		
4	Endrin	Pesticide		
5	Chlordecone	Pesticide		
6	Hepachlor	Pesticide		
7	Hexabromobiphenyl	Brominated flame retardant		
8	Hexabromocyclododecane	Brominated flame retardant		HBCDD
9	Hexabromodiphenylether	Brominated flame retardant		
10	Heptabromodiphenylether	Brominated flame retardant		
11	Hexachlorobenzene	Unintended bi-product		HCB
12	α-Hexachlorocyclohexane	Pesticide		α-HCH
13	β-Hexachlorocyclohexane	Pesticide		β-HCH
14	γ-Hexachlorocyclohexane	Pesticide		Lindane
15	Mirex	Pesticide		
16	Pentachlorobenzene	Unintended bi-product		PeCB
17	Polychlorinated biphenyls	Industrial product	209 congeners	PCB
18	Technical endosulfan (incl. isomers)	Pesticide	2 isomers	
19	Tetrabromodiphenyl ether	Brominated flame retardant	Technical mixture	
20	Pentabromo diphenyl ether	Brominated flame retardant	Technical mixture	
21	Toxaphene®	Pesticide	Chlorobornanes and derivatives	
22	Dichlorodiphenyltrichloroethane	Pesticide		DDT
23	Perfluorooctane sulfonate	Industrial product	Perfluorinated alkylated substance (PFAS)	PFOS
24	Polychlorinated dibenzodioxins	Unintended bi-product	75 isomers/congeners	PCDD
25	Polychlorinated dibenzofurans	Unintended bi-product	135 isomers/congeners	PCDF

In addition to these internationally prioritized substances, the national monitoring authorities usually include additional national priority substances in accordance with national and regional regulations and the local monitoring priorities. Therefore, the national priority lists for atmospheric POPs monitoring may deviate from in the different countries contributing to the international research and monitoring of POPs. The actual lists can be found in the national monitoring reports. For details, please consult the respective national monitoring authorities.

Relevant Project Overview

For scientific research associated with POP-related atmospheric distribution processes and pathways, IPY 2007–2009 paved the way for important follow-up activities for many of todays' internationally coordinated Polar research initiatives. A series of campaign-based field studies with potential for long-term commitment were conducted during the IPY period both in Antarctic and in Arctic locations. The atmospheric POPs monitoring coordinated by AMAP and EMEP, as well as the associated atmospheric data repository (ebas.nilu.no), played an important role in channeling, shaping, and establishing innovative research in atmospheric pollution research (fate and distribution) including POPs associated with IPY 2007–2009. Already in the initial phase of IPY 2007–2009, the scientific assessment of already-available monitoring and research data indicated that distribution and fate of POPs including long-range atmospheric transport are potentially influenced by climate change (CC) processes (Hung et al. 2005). First signs for the influence of CC were reported in national reports from the atmospheric POP monitoring at the Norwegian Atmospheric research station "Zeppelin station" (Ny-Ålesund, Svalbard) and summarized in a comprehensive publication (Kallenborn et al. 2012).

Arctic Monitoring and Assessment Programme (AMAP)

Today, AMAP plays a key role in coordinating and harmonizing pollutant monitoring (POPs, metals, radionuclides) in the Arctic. AMAP is one of six Working Groups of the Arctic Council. AMAP has the official task and obligation to:

- Monitor and assess the status of the Arctic region with respect to pollution and climate change issues;
- Document levels and trends, pathways and processes, and effects on ecosystems and humans, and propose actions to reduce associated threats for consideration by governments;

- Produce sound science-based, policy-relevant assessments, and public outreach products to inform policy and decision-making processes.

AMAP's work is directed and supervised by the Ministers of the Arctic Council and their Senior Arctic Officials, who have requested AMAP to also support international processes that work to reduce the global threats from contaminants and climate change. These include the UNEP Framework Convention on Climate Change, the Stockholm Convention on POPs, the UNECE CLRTAP (EMEP), and the Global Mercury Agreement. More information on the AMAP activities can be found at the dedicated program Web page (www.amap.no) as well as in the comprehensive status reports, which can be downloaded from here.

Atmospheric Monitoring Network for Anthropogenic Pollution in Polar Regions (ATMOPOL)

The ATMOPOL project (IPY 76) received no direct national funding for the coordinative research and screening work planned from the national IPY funding programs. However, the INCATPA initiative, developed as a direct result of the ATMOPOL planning efforts, was supported by national Canadian research funds and is separately described below in this summarizing report. ATMOPOL planned to establish a comprehensive atmospheric station network on distribution of anthropogenic pollutants (metals and organic contaminants) in the Antarctic and Arctic environment.

The final ATMOPOL research during the IPY period was restricted to scientific work funded directly via several minor research projects supported by the Norwegian national research funds (RCN), AMAP, the Nordic Council of Ministers (NCM) as well as the 7th EU Framework Programme (as collaboration with the FP7 project "Arctic Health Risks: Impacts on health in the Arctic and Europe owing to climate-induced changes in contaminant cycling; ArcRisk"). The international research performed as contribution to the ATMOPOL studies encircled the influence of CC on atmospheric distribution patterns and the identification of new emerging contaminants in Arctic environments (Grannas et al. 2013; Kallenborn et al.2012a, b; Carlsson et al. 2012; Becker et al. 2012; Olsen et al. 2011; Bengtson Nash 2011; Barber et al. 2010).

A long-term Norwegian atmospheric monitoring program on POPs was established in 2007 at the Norwegian Troll/Trollhaugen research station (Queen Maud Land, Antarctic, see below). The first monitoring period (2007–2011) on POPs in Antarctic air has been initiated, funded by the Norwegian Antarctic Research Expedition (NARE) and published with contribution from the ATMOPOL co-operation (Hansen et al. 2009). The long term first trend analysis on POP distribution in atmospheric samples from the Trollhaugen monitoring program was published in 2012 (Kallenborn et al. 2013).

Inter-Continental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA)

The *INterContinental Atmospheric Transport of anthropogenic Pollutants to the Arctic* (INCATPA, IPY 327, http://www.ec.gc.ca/api-ipy/default.asp?lang=En&n=8EBD7558-1) project was endorsed by IPY in 2006 and established in order to measure toxic chemicals produced from human activity and carried through the air to the Arctic. INCATPA focused on the environmental risks associated with the emissions of POPs and mercury (Hg) in the Pacific region for the contaminant loads in the Arctic. Before the IPY, Arctic air monitoring of POPs and Hg was performed mainly at Alert (Canada), Zeppelin (Norway), Pallas (Finland), and Storhofdi (Iceland) since the 1990s under national monitoring programs, reported to and coordinated by AMAP. Hg has also been continuously measured in air at Whistler (BC, Canada) and Amderma (Russia). During the IPY, air measurements of POPs and Hg were initiated at Little Fox Lake (Yukon, Canada) and Waliguan (China), as well as Valkarkai (Russia) (POPs only), Barrow (Alaska, USA) (Hg only), Dillingham and Fairbanks (Alaska, USA) (POPs only), Mt. Changbai (China) (Hg only), Wudalianchi and Xuancheng (China) (POPs only), Hedo Island (Japan) (POPs only), and Ba Vi (Vietnam) (POPs only). Soil and air samples were collected along the Chilkoot Trail (Yukon/Alaska, USA) in summer 2007, at different elevations. The purpose was to investigate the atmospheric deposition of POPs and emerging chemicals on mountain ranges in the Kluane National Park (Yukon, Canada). Combined with the air concentration data collected at Little Fox Lake, this work has provided insight on the roles that mountains and forests play in intercepting POPs carried by trans-Pacific air masses.

IPY INCATPA is an international collaboration among Canada, Russia, China, Vietnam, Japan, and the USA. with linkages to international air monitoring networks such as AMAP. The project aims to provide information to help determine what relative risks chemical emissions from the Pacific region pose on the Arctic environment compared to emissions from other parts of the world. These chemicals were simultaneously measured in air in the Canadian, Russian, and American Arctics, as well as at potential source regions on the Pacific Rim. Global-scale multimedia transport models were used to forecast trans-Pacific and intracontinental transport to the Arctic. The impact of changes in emissions and climate on contaminant levels and trends in Arctic air were assessed. Moreover, under INCATPA, an active outreach and communication component enhanced the general public's understanding on contaminant transport and behavior in the Arctic.

The sampling and chemical analytical protocols applied for the INCATPA collaboration and the respective samples followed the sampling and quality control procedure developed for the Canadian National Northern Contaminants Programme which is in line with the AMAP quality requirements (Hung et al. 2005).

Global Atmospheric Passive Sampling (GAPS) Network

The GAPS Network was established already before IPY2007–2009. The GAPS program was initiated in December 2004 as an initial two-year pilot study before evolving into a long-term monitoring network. Today, the monitoring network consists of more than 50 sites on seven continents using polyurethane foam (PUF) passive air samplers to monitor POPs (Bengtson Nash et al. 2011).

The GAPS Network is a long-term international passive air monitoring program that supports the research and policy needs of Canadian and international programs on POPs. Internationally, GAPS helps to assess effectiveness of control measures that have been implemented for POPs under treaties such as the Stockholm Convention on POPs, and the POPs Protocol of the CLRTAP under the UNECE.

GAPS was the only global-scale program for air that reported to the first global monitoring plan (GMP) of the Stockholm Convention on POPs, adopted at COP4 in May 2009 in Geneva. Air is one of two core media (human tissues being the other) under the GMP. The GAPS approach to passive air sampling is promoted in the Guidance Document for the GMP as a feasible and cost-effective way to improve spatial coverage. For some of the UN regions and subregions, air measurements from GAPS represent the only available data for POPs in air. The GAPS Network promotes and assists in capacity building projects that aim to fill data GAPS and develop regional air sampling networks for POPs that employ passive samplers (Xiao et al. 2007).

Results from GAPS measurements of POPs in air are integrated with other information (Fig. 2.2) to assess temporal trends of POPs (effectiveness of control measures) and for investigating regional and long-range transport of POPs and other priority chemicals. The role of variable meteorology and climate has also been shown to be critical for this analysis. The GAPS Network is a today key program for producing comparable global-scale data for POPs in the troposphere. The GAPS Network conducts measurements of POPs and priority chemicals in air with the following objectives:

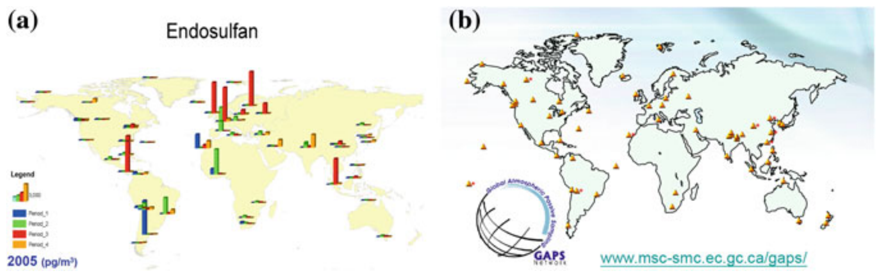

Fig. 2.2 **a** Spatial distribution of pollutants (example: endosulfan 2005) as well the station network **b** for the GAPS Network

1. Demonstrate the feasibility of passive air samplers (PAS);
2. Determine spatial and temporal trends in air;
3. Screen for and identify new chemicals in air; and
4. Contribute useful data for assessing regional and global long-range atmospheric transport.

Target chemicals include legacy POPs listed under the Stockholm Convention such as PCBs and organochlorine pesticides (OCPs) as well as emerging priority pollutants—e.g., brominated flame retardants, polyfluorinated chemicals, and current-use pesticides, to name a few. Under the GAPS Network, seasonality and long-term temporal trends can be investigated.

Pilot studies are currently undertaken in order to assess a modified PUF disk sampler comprising XAD powder impregnated onto PUF disks (sorbent-impregnated PUF or SIP disks). SIP disks have a higher retention capacity compared to PUF disks and are well suited for capturing the more polar and volatile priority chemicals. The modified samplers also are useful for screening and identifying new compounds of interest that may be present in the atmosphere.

Circumpolar Flaw Lead (CFL)

The circumpolar flaw lead (CFL), which forms in the Arctic sea ice each year, is a path of thinner ice and interconnected polynyas (areas of open water). It separates the mobile pack ice from coastal ice attached to the land. As a warming climate causes the sea ice to decline, the structure of the CFL changes. The main goal of the CFL study was to better understand changes in the flaw lead, and their repercussions for other physical and biological processes in the Arctic. The research priorities of IPY-CFL were performed in 10 thematic subgroups: (1) Physical oceanography, (2) Sea ice, (3) Primary production, (4) Pelagic and benthic food web, (5) Marine mammals, (6) Gas fluxes, (7) Carbon fluxes, (8) Contaminants, (9) Modeling, and (10) Arctic people. In addition, several public outreach initiatives were also part of IPY-CFL.

However, atmospheric POPs research was an integrated part in this large interdisciplinary scientific program. More details on the science program and the outcome of the CFL initiative can be found in a comprehensive review recently published (Bengtson Nash et al. 2008).

Organic Atmospheric Pollutants: Research and Monitoring in Antarctica

The establishment of atmospheric environmental pollutant research and monitoring in Antarctica is a demanding task, both from an economic and from a logistical point of view. In addition, as the major POP source regions have traditionally been located in the Northern hemisphere, pollutant distribution and environmental effects in the comparatively pristine Antarctic environments have to date been considered as of minor relevance for comprehensive environmental risk assessment. Therefore, pollutant research in Antarctica is documented only in relatively few scientific reports and studies. However, during IPY 2007–2009, several national Antarctic research programs established pollutant monitoring and screening in various environmental media including the atmosphere. Also the GAPS atmospheric monitoring program is maintaining several atmospheric units in Antarctic stations. These monitoring- and research-based scientific data will without doubt contribute to a better understanding of the global distribution processes underlying the ubiquitous distribution profiles identified for many legacy POPs. Here, few examples are described in order to illustrate the importance of these types of investigations and long-term commitment for the scientific understanding of fate and distribution of POPs in pristine environments.

Atmospheric POPs Monitoring at the Antarctic Troll Station (Trollhaugen)

Initiated and motivated by IPY supported priority funding, the Norwegian Antarctic Research Expedition (NARE) program extended the already-established Norwegian Summer Research station “Troll” in Dronning Maud Land (Queen Maud Land, Antarctica) into a year-around research station in 2005. In 2007, several IPY funded activities were conducted at Troll including the atmospheric pollutant monitoring. Currently, research campaigns and monitoring in a variety of research field are carried out at Troll.

The scientific monitoring and research program at the Troll Atmospheric Observatory is managed by the Norwegian Institute of Air Research (NILU). In 2014, the atmospheric measurements were moved from the Troll Atmospheric Observatory to the less perturbed site Trollhaugen situated one km east of research station Troll, 235 km from the Antarctic coast. Since 2007, NILU is performing continuous year-around measurements of Hg, surface ozone, CO, aerosols (chemical, physical, and optical properties), UV radiation and total ozone, selected POPs, and an extended number of light hydrocarbons and chlorofluorocarbons (CFCs).

The year-around atmospheric monitoring of the above-listed wide variety of pollutants at the Troll/Trollhaugen atmospheric monitoring station aims at improving today’s scientific knowledge on distribution mechanisms for atmospheric transport of pollutants into Antarctica. The overall strategies for the monitoring activities are comprehensively summarized in a scientific review publication (Bengtson Nash et al. 2013). The monitoring is also coordinated with other ongoing campaign-based screening activities at other stations in Antarctica, such as Germany’s Neumayer Station, the Australian Casey observatory, and the UK’s Halley Research Station.

POPs are measured at Troll/Trollhaugen using high-volume active air samplers. Samples are collected on a weekly basis (i.e., 52 samples per year). The

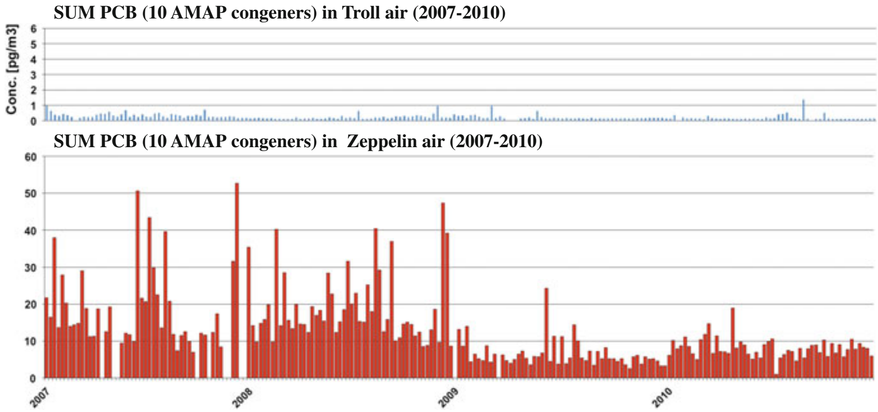

Fig. 2.3 Comparison of air concentrations (pg/m^3) of sum 10 priority PCBs (integrated weekly samples, PCBs: 28, 52, 99, 101, 118, 138, 149, 153, 170, 180) from Antarctica (Troll station) and Arctic (Zeppelin Atmospheric observatory, Ny-Ålesund, Svalbard) for the period 2007–2009 (Hung et al. 2010; Hansen et al. 2009; Kallenborn et al. 2013)

here-established year-around monitoring of POPs is currently considered as the longest continuous monitoring program in Antarctic air covering the time period 2007–2015. Details on the POPs atmospheric monitoring at the Troll station including discussion on POPs monitoring data collected in the weekly samples during the period 2007 until 2010 are summarized in a recent publication (Hansen et al. 2009; Kallenborn et al. 2013).

Targeted POPs are 32 PCB congeners, α- and γ-HCH, trans- and cis-chlordane, *trans*- and *cis*-nonachlor, *p,p'*- and *o,p*-DDT, DDD, and DDT as well as HCB. In general, the monitored concentrations of POPs at Troll in 2007–2010 are one order of magnitude lower than those measured in the Norwegian Arctic atmospheric background site; Zeppelin in Ny-Ålesund, Svalbard (Fig. 2.3). Similarly as observed in Arctic air samples, HCB is the predominant POP compound at the air samples from Troll with levels of around 20 pg/m^3 throughout the years (22.9 pg/m^3 average concentration in 2010). In general, the following concentration distribution has been found in air samples from Troll: HCB > Sum HCH > Sum PCB > Sum DDT > Sum chlordanes.

Even when considering the much lower POP concentration levels in Antarctic air compared to POP levels in Arctic air, atmospheric transport episodes were identified in the already-available data provided from the Troll/Trollhaugen observatory (Hansen et al. 2009; Kallenborn et al. 2013). The long-term monitoring data from Troll also confirm that atmospheric long-range transport is a major contamination source for POPs in atmospheric environments above the Troll station observatory. Several long-range transport events were identified characterized by unusual elevated levels of pesticides and/or industrial originated compounds. Back-trajectory calculations and complex transport modeling (FLEXPART) were applied for source apportionment and confirmation of transport pathways. In all cases, contaminated air masses arriving from potential South American and South African sources were identified.

The air concentrations of POPs at Troll station are comparable with earlier measuring campaigns reported from other Antarctic research station from the past 18 years. For all compounds, higher levels were found in samples collected during the early 1990s except for HCB for which in all samples regardless time period similar concentration distribution was observed. These concentration reductions for most of the target substances are obviously a direct consequence of international regulations restricting the usage of POP-like chemicals on a worldwide scale (Hansen et al. 2009; Kallenborn et al. 2013).

Atmospheric POPs Monitoring at the Antarctic Casey Station

In 2009, the Southern Ocean POPs Program of Griffith University was tasked by the Australian federal Department for Environment to undertake continuous atmospheric monitoring for POPs in the Australian Antarctic Territory to serve Australia's obligations under the GMP. The first year's worth of data, reported in the 2011 project report (Wild et al. 2014), provided the first information regarding the chemical composition of air masses of the Australian Antarctic Territory (constituting 42 % of the Antarctic continent) in over two decades.

Atmospheric sampling via a high flow through passive sampler has been continuously measuring POPs at the selected sampling site, the abandoned Wilkes station, near Australia's all-year Casey station (66°16'54"S and 110°31'27"E), since sampling was initiated in 2009. The routine repertoire of chemicals monitored at this location includes OCPs, PCBs, and polybrominated diphenyl ethers (PBDEs). The first results from the program found HCB and Endosulfan-I to be the compounds detected in the highest quantities. Findings of HCB are in accordance with previous reports which have shown HCB to be a dominant compound accumulating in Southern Ocean food webs. Sampling found strong indications of the active all-year Casey station acting as an emitter of PBDEs to the local environment, a finding that has since been further investigated and published by the SOPOP Program.

Analysis of the Casey Station air shed was ineffectual at linking contaminants to potential lower latitude source regions. The study hereby highlighted the limited application of back-trajectory-based analysis in Antarctica due to the circumpolar vortex and effective mixing of air masses that occurs once wind masses reach south polar latitudes.

Quantification of the chemical composition of Antarctic air masses provides information regarding system input from hemispheric sources via this pathway. Continuous long-term measurements are still needed to facilitate evaluation of temporal contamination trends and hereby the effectiveness of the aims of the Stockholm Convention in reducing or eliminating these chemicals from the environment. To this effect, sampling at Casey station remains ongoing with further reports and articles in preparation.

Achievements and Perspectives

The national IPY supporting programs officially funded only two project initiatives; from ca. 30 officially IPY-IPO endorsed initiatives, where the in-depth research on atmospheric distribution of POPs was an important scientific goal. The INCATPA and the (CFL) initiative were both funded by the Canadian IPY funding program. However, the establishment of extended atmospheric POP monitoring activities, where already-operative national programs were substantially extended with new emerging target substances, recently identified as relevant long-range transported pollutants, is considered a significant spin-off of IPY and the related research on polar environmental pollutants. In many circum-Arctic countries (i.e., Canada, USA, Norway, and Russia), already-operative POP monitoring programs were extended and upgraded. These updated long-term polar atmospheric POPs monitoring programs are today considered important data providers for a variety of interdisciplinary research activities in environmental sciences including fate modeling and environmental impact assessments. In Antarctica, two new POP atmospheric monitoring programs were established: one at the Norwegian "Troll/Trollhaugen" station and one at the Australian research station "Casey." Both monitoring programs follow today similar sampling and quality control protocols established according to the quality control guidelines of the AMAP atmospheric monitoring network. These new stations, thus, complement the already-established (Arctic) monitoring network with an Antarctic module, where data and results are easily comparable. Therefore, this established multinational network has the potential to develop into a global network of active sampling and monitoring for POPs in background polar environments.

The GAPS Network has been established before IPY, but was significantly advanced by establishing new stations during IPY. Today, GAPS is utilized actively and constantly referred to by the UNEP's global regulation for POPs (Stockholm Convention) as important surveillance tool for the evaluation of spatial and temporal trends of priority POPs on a global scale.

During the INCATPA initiative, a strong focus was laid upon identification and characterization of potential source regions for the atmospheric POP transport across the Pacific Ocean. In this context, several new atmospheric monitoring locations were established (i.e., in Vietnam, China) and are still operative, maintained by the hosting national institutions.

The CFL initiative followed a strongly interdisciplinary approach, where a research platform (in this case the RV Amundsen) was located in the Beaufort Sea during one year, allowing scientists coordinated research programs and fieldwork under Arctic environmental conditions. The here-obtained results could be discussed and interpreted already aboard across the disciplinary borders among the participating scientific groups. The CFL initiative has been highly productive and produced a large number of highly cited scientific publications (http://umanitoba.ca/ceos/research/cfl.html). The already-demonstrated efficiency of this strategy motivated other national program to embark on similar field studies. Recently, the field campaign of a Norwegian National Arctic research program has been finalized where the Norwegian Research Vessel Lance was located for six months in the marginal ice Zone northwest of Svalbard as research platform for interdisciplinary research on Arctic climate change (N-ICE; see www.npolar.no).

Although only two projects were officially supported by IPY national funding, a variety of ongoing research projects linked into IPY research activities gained considerable scientific profit from the national and international funding priorities and collocation with other funded IPY projects. New infrastructures for POPs research and monitoring were established (ATMOPOL, INCATPA, and GAPS). New future directed research strategies were developed and applied in the field (CFL). A significant legacy component for atmospheric monitoring of POPs in Polar Regions is associated with the establishment of the new POPs atmospheric monitoring locations in Antarctica, where the Troll/Trollhaugen and the Casey site are adding complementary value to the already-established circum-Arctic atmospheric POPs monitoring, officially established during the early 1990. In this way, the long-term circum-Arctic atmospheric POPs monitoring was extended into a clearly Polar network and hopefully many other stations will join this undertaken in the near future.

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Chapter 3
Atmospheric Pollution Research on Greenland

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Introduction

Danish studies in the Arctic atmosphere were almost all carried out on Greenland. There were not any specific programmes within IPY but activities were performed in the framework of Arctic Monitoring and Assessment Programme (AMAP) Financed from Denmark by means from DANCEA. A long series of studies in other spheres than the atmosphere have also been carried out but they will not be described here as they are beyond the scope of this chapter.

Most activities were carried out at Station Nord now the Villum Research Station (VRS) but from 2003 to 2006 the atmospheric measurements were carried out at Lille Malene Mountain outside Nuuk and to less extent in Nuuk. VRS and Lille Malene (Nuuk) stations are shown in Fig. 3.1.

Back in the 1950s, pilots reported about frequently observed periods with reduced visibility. This phenomenon was later called Arctic Haze. Around 1980, the first studies from Greenland investigated this phenomenon (Flyger et al. 1980; Heidam 1981). In the following years, it has been proven that the Arctic haze was due to long-range transport of air pollution from mid-latitudes into the Arctic region. When we first started our measurement activities in Greenland, the aim was to describe the mechanisms of long-range transport of pollution from emission regions at lower latitudes to the Arctic causing Arctic haze. Thus, comparison between measured and modelled parameters has focused on species relevant for

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Fig. 3.1 Location of Villum Research Station, Station Nord (VRS) (24 m above sea level, 81° 36′N, 16° 40′W) and Nuuk (345 m above sea level at 64.1°N and 51.4°W). The stations are indicated with *red ovals*

Arctic haze. The observed concentrations are strongly dependent on the position of the polar front and on the North Atlantic Oscillation Index and Arctic Oscillation Index. In winter, the front moves southwards and opens for large anthropogenic emissions at mid-latitudes can be transported into the Arctic. In summer, the front moves northwards and cuts off the transport from these source regions. This dependence is very well described by the Danish Eulerian Hemispheric Model (DEHM) (Christensen 1997; Heidam et al. 1999, 2004; Skov et al. 2006b). Later focus moved towards a general description of pollution in the Arctic and the deposition of xenobiotic to the Arctic environment. Persistent organic pollutants (POPs) and mercury were included. Lately, climate-related research is carried out while Arctic is experiencing a heating twice as fast as the globe on average (IPCC 2013). In particular, the processes that control CO_2, CH_4, tropospheric ozone, black carbon and particles are now of interest.

In the following sections, we will present and discuss the results that have been accomplished within the above-mentioned subjects so far.

Site Description, Experimental and Model Section

VRS

Monitoring of atmospheric air pollution in the Greenlandic part of Arctic was initiated in 1990 at Station Nord (now: Villum Research Station) in North Greenland (81° 36′N, 16° 40′W), as an AMAP monitoring site providing data on SO_2, SO_4^{2-}, NH_3, NH_4^+, HNO_3, NO_3^- and several elements (Heidam et al. 2004, 1999). Monitoring activities were paused from 2002 to 2007 and resumed in 2008, where PSAP measurements of black carbon (BC) and NO_X were established (Nguyen et al. 2012; Massling et al. 2015). VRS is a remote Arctic background site. Its location makes it ideal to follow the effect in the Arctic of the massive emissions in the Eastern part of Europe and Russia (Heidam et al. 2004). The POP measurements at VRS started in the end of 2007. Ozone and gaseous elemental mercury (GEM) were started in 1996 and in 1999, respectively, at VRS. Today, GEM data are a contribution to the European project within the 7th Framework Programme; Global Mercury Observation System (GMOS).

Lille Malene Station, Nuuk

More than 2000 km south of VRS is located Nuuk (64° 10′N, 51° 44′W, see Fig. 3.1). Measurements of SO_2, SO_4^{2-}, NH_3, NH_4^+, HNO_3, NO_3^- and several elements were conducted from 2002 to 2004 (Skov et al. 2006b) at Lille Marlene Mountain outside the Capital. Atmospheric monitoring of POPs in Greenland was done for the first time. The sampling station for POPs was located in the city of Nuuk because of lack of power at Lille Marlene Mountain. The activity was a part of AMAP.

Danish Eulerian Hemispheric Model (DEHM)

In order to understand the connection between emission far from the receptor point and the resulting concentration in High Arctic, comparison between measured values and model results was done. The DEHM was developed and applied. DEHM uses a spatial resolution of 150 km times 150 km south of 60N and a nested grid of 50 km times 50 km north of 60N with 29 layers up to 15 km using a chemical scheme of about 67 different species (Christensen 1997).

Chemical Composition and Sources to Atmospheric Aerosols

Local Sources Versus Long-Range Transport

In a recent study (Fenger et al. 2013), it was found that in late winter/early spring just after polar sunrise fine particles, smaller than 1 μm containing SO_4^{2-}, Cl^- and NO_3^-, most probably originate from long-range transport, which appears to be the most important source of major inorganic anions at VRS. SO_4^{2-} is by far dominating, accounting for 50–85 % of the analysed mass. However, the study suggests that Cl^- and NO_3^- in coarser particles (>1.5 μm) originate from local/regional sources, and Fenger et al. hypothesize that frost flowers on the sea-ice are a potential source to Cl^- and NO_3^- particles. This is supported in another study (Shaw et al. 2010), where particles in the Arctic in winter clearly had a sea spray profile despite the frozen polar ocean. Large NO_3^- particles can also be caused by blowing snow, while the snowpack contains NO_3^- originating from precipitation (Kuhnel et al. 2013) or dry deposition (Bjorkman et al. 2013). However, the uncertainties of the NO_3^- sources are still substantial and further studies on nitrogen processes in the high Arctic are needed (Fenger et al. 2013; Krnavek et al. 2012).

A Statistical Approach

Air quality measurements show clear differences among the Station Nord and Nuuk (Table 3.1); for example, the crustal elements Al, Si, Ti and Fe are measured in higher concentrations in Nuuk, while the opposite is reported for the anthropogenic species S, As and Pb. Br is measured in almost three times higher concentrations at VRS, which is closely linked to particulate sulphuric acid. While V and Cr have been observed in similar concentrations, the anthropogenic species Cu, Ni and in

Table 3.1 Average concentration of monitored elements at Nuuk (2002–2004) and VRS (unit; ng m^{-3}) as specified for the entire monitoring period (1990–2002 and 2008–2009) and recent data (2008–2009)

	Al	Si	SO_X^a	K	Ca	Ti	V	Cr	Mn	Fe
Nuuk (2002–2004)	44.4	143	119	42.9	51.7	3.1	0.17	0.11	0.60	33.4
VRS (1990–2009)	30.5	91.5	252	20.5	28.2	2.2	0.15	0.10	0.44	23.9
VRS (2008–2009)	38.4	76.2	256	23.4	30.1	1.9	0.12	0.11	0.48	19.8

	Ni	Cu	Zn	As	Se	Br	Sr	Zr	Pb
Nuuk (2002–2004)	0.17	0.36	2.6	0.02	0.05	0.72	0.79	0.10	0.48
VRS (1990–2009)	0.11	0.21	0.93	0.10	0.03	2.2	0.30	0.08	0.67
VRS (2008–2009)	0.11	0.14	1.4	0.07	0.05	1.5	0.38	0.08	0.72

[a] $SO_X = S\text{–}SO_2 + S\text{–}SO_4^{2-}$ in ng S m^{-3}

particular Zn have been found in higher concentrations in Nuuk, evidencing a stronger influence of metal industry most probably located in northern Canada.

Investigations over the past decades have shown that a considerable part of the Greenlandic, Norwegian, Russian, Alaska and Canadian Arctic troposphere is significantly influenced by atmospheric pollution of distant latitude origin (Barrie et al. 1981; Bourgeois and Bey 2011; Fenger et al. 2013; Flyger et al. 1980; Heidam et al. 1999, 2004; Nguyen et al. 2012; Pacyna et al. 1984). The phenomenon known as Arctic haze is widespread throughout the Arctic during wintertime, where the lower troposphere is partially isolated from other layers of the atmosphere by the 'Arctic Front' barrier, resulting in low potential temperatures at the ground surface (Klonecki et al. 2003; Stohl 2006). Sources include the industrial and other anthropogenic activities in the Eurasian region, in particular northern Russia, central Europe and North America (Bourgeois and Bey 2011; Heidam et al. 1999; Shindell et al. 2008). Arctic haze aerosols contain a mixture of SO_4^{2-}, organic matter (OM), black carbon (BC) and dust, nitrogen compounds, heavy metals and other elements (e.g. Li and Barrie 1993; Quinn et al. 2002). A typical annual variation with elevated concentrations of such elements during Arctic winter and a low deposition at the same time limits the removal of Arctic haze (Barrie et al. 1981; Heidam et al. 1999, 2004; Quinn et al. 2007). The sources to Arctic air pollution in Greenland have been evaluated in more detail by the chemical transport model such as DEHM and the receptor models such as positive matrix factorization [PMF; (Paatero 1997)] and constrained physical receptor model (COPREM) (Wahlin 2003) as described elsewhere (Heidam et al. 2004; Nguyen et al. 2012; Skov et al. 2006b). Source apportionment of Arctic aerosols is typically confined to a limited number of sources due to the unique meteorology and transport mechanisms characteristic for these remote regions as described earlier. In a recent study (Nguyen et al. 2012) based on data from 2008 to 2009, five sources were identified at VRS, i.e. a **crustal** and **marine** source, a **Cu/Ni** metal source and a **Zn** source, in addition to a **combustion** source representing various distant anthropogenic activities (Nguyen et al. 2012), see Fig. 3.2.

Among sources of natural origin, the **crustal** source is dominated by (Al/Si/Ca/Ti/Mn/V/Fe) and peaks in the spring and late summer. Na and Cl are dominating species but also (K/Ca/Sr/V) and small amounts of SO_X and BC are present (Fig. 3.2). The worldwide increasing ship traffic probably explains some BC, SO_X and V. However, V is relatively abundant in open ocean waters (Wang and Sañudo Wilhelmy 2009). The source shows a clear annual trend with lower summer concentrations and increased contributions in the autumn and winter periods. The sea surrounding VRS is frozen during the winter, and Cl^- containing particles at VRS may originate from frost flowers torn away at high wind speeds (Domine and Rauzy 2004). Although Br origin is from sea salt and marine organisms (Gribble 2000), it is virtually only associated with anthropogenic sources. Three anthropogenic sources have been characterized by high abundances of Zn, Cu, Ni, Pb and As, which are interpreted as anthropogenic sources. Two of the sources influenced by Cu and Ni smelters on the Kola Peninsula (Nikel, Monchegorsk and Zapolyarnyy) and Norilsk in Northern Siberia (Christensen 1997;

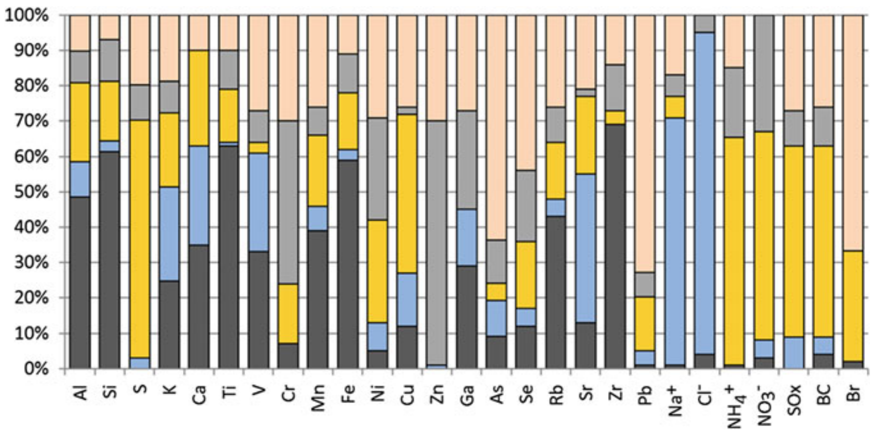

Fig. 3.2 Apportionment of sources to selected elements, bulk ions and black carbon (BC) using PMF (*black* soil; *blue* marine; *grey* Zn; *yellow* Cu/Ni; *pink* combustion)

Heidam et al. 1999, 2004) peak in spring/summer (**Cu/Ni** source) and winter (**Combustion** source). The Cu/Ni source is probably an agglomerate of sources from Eurasia, as evident from the contribution of these sources to the elements Cu (45 %) and Ni (29 %). Cu and Ni are present in both the Cu/Ni and Combustion sources but virtually absent in Zn and the natural sources. The impact of anthropogenic and natural sources on Cu points towards Eurasia and in particular the Siberian Cu/Ni industry, as a major source of pollutants in the higher Arctic. Primary and secondary combustion products are to a large extent attributed to this source, i.e. NO_3^- (59 %), SO_X (54 %) and BC (54 %), probably reflecting combustion processes fuelled by coal combustion, as indicated by the marker Se (19 %). Most likely oil combustion is not a source as evidenced by the low abundance of V (3 %) a typical oil combustion tracer. Alternatively, the combustion products may originate from power generation and other combustion processes connected to metal industry near the industrial sites. BC is mainly apportioned to anthropogenic sources (80 %), and predominantly found to have Siberian origin. The high contribution from Siberian metal industries to BC at VRS is inevitable, which agrees with other recent findings (Hirdman et al. 2010; Wang et al. 2011). The *Combustion* source is also influenced by Cu/Ni in addition to Pb (72 %), As (63 %) and typical anthropogenic pollutants, which are long-range transported to VRS (Heidam et al. 1999, 2004). V (27 %) and Se (44 %) indicate oil- and coal-fuelled industry and/or other anthropogenic combustion processes. Primary (BC) and secondary combustion products (SO_4^{2-}, NO_3^-) are also abundant in the Combustion source. As and Se have previously been apportioned to sources from the Kola Peninsula (Maenhaut and Cornille 1989). A recent source apportionment study (Nguyen et al. 2012) identified a **Zn** source, which likely evidence new activities related to Zn or unique meteorological conditions in the years 2008 to 2009. The source appears to have a different origin than *Cu/Ni* and *Combustion* source, since it is only minor influenced by Cu, which is the characteristic of these sources. Br is furthermore absent in the Zn source, whereas SO_X (10 %) and S (a proxy for sulphate; 10 %) are low in comparison with the Siberian sources imprinted with abundant Br and S. Air mass back trajectory analysis of the Zn source furthermore points towards Westerly origins of VRS (Canadian Arctic, North America and West Greenland), especially from Canadian Arctic. Br is apportioned to the *Combustion* source (66 %) and the *Cu/Ni* source (31 %) and plays a key role in ozone depletion (ODE) during Arctic sunrise (Skov et al. 2004), though the activation of bromine is not fully understood (Sumner and Shepson 1999). Sea-salt bromine may accumulate in the snowpack during the long polar night and evolve as Br_2 into the atmosphere at polar sunrise (Mcconnell et al. 1992). BrO is formed in the reaction with O_3 and Br atoms from photolysis of Br_2, or, e.g. HOBr from the reaction of, e.g. BrO with the HO_2 radical (Impey et al. 1999). Br may also react with organic compounds to form HBr and organic bromine compounds, which are suspected to be scavenged by Arctic aerosols (Impey et al. 1997, 1999; Mcconnell et al. 1992). Br_2 can also recycle with or without O_3 (Impey et al. 1999; Simpson et al. 2007) and photolyses to Br to further deplete O_3. The timely variation of Br resembles the anthropogenic elements Pb and S, which can probably be explained by S acting as a "transport container".

Modelled Black Carbon and Sulphate

Since 2008, the mass concentration of black carbon is monitored at St. Nord, Northern Greenland. This is done using a custom-built Particle Soot Absorption Photometer (PSAP), which measures the transmission of light through a filter that gets continuously loaded with the ambient aerosol. Using a reference filter without aerosol load, the change in transmission is measured by comparing the two signals and the black carbon mass is calculated by applying a specific absorption coefficient which is a site-dependent parameter as one has to account for the ageing of the observed aerosol. The measurements show a typical seasonal pattern as observed for the sulphate mass concentrations but with much lower concentration values. In general, the pattern depicts the occurrence of Arctic haze episodes. Mass concentrations of sulphate typically reach values about 0.4 $\mu g/m^3$ during late winter and early spring, see Fig. 3.3. Sulphate as well as black carbon mass concentrations was simulated using DEHM. DEHM was able to catch the seasonal cycle of both pollutants matching the concentrations within for most of the time periods.

As one interesting finding, a high correlation was observed between sulphate and black carbon mass concentrations, see Figs. 3.3 and 3.4 where black carbon shows the same seasonal pattern as sulphate. The two pollutants have only partly similar sources as there are, for example, anthropogenic emissions from ships or from road traffic and industrial activities in Eurasian countries. Other sources such as biomass combustion emit high concentrations of black carbon particles but on the other hand only precursors for sulphate aerosols to a very minor extent. Black carbon is emitted

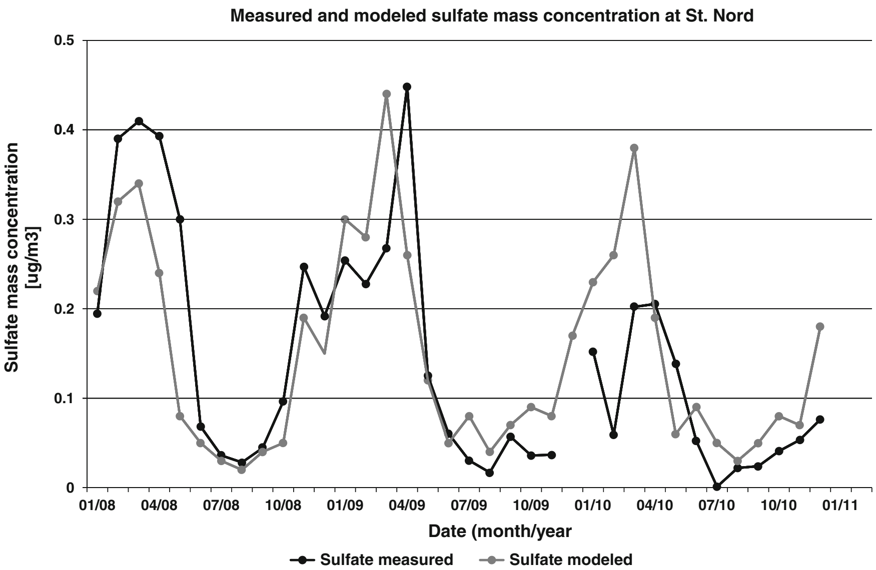

Fig. 3.3 Measured and modelled sulphate mass concentration (unit; μg S/m^3) at VRS

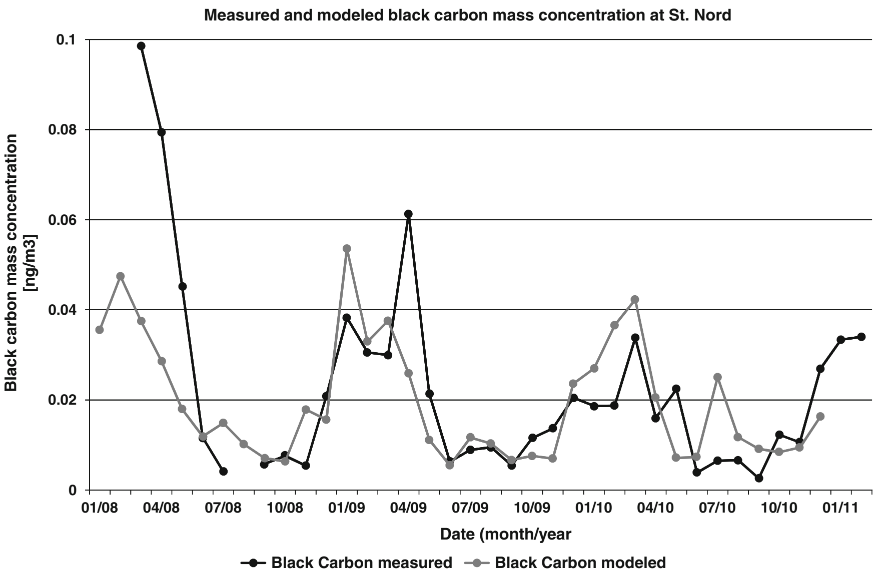

Fig. 3.4 Measured (with a PSAP) and modelled black carbon mass concentration (unit; ng black carbon/m^3) at VRS. Note that measured black carbon is connected with increased uncertainty due to high uncertainty on the determination of the flow in the PSAP

as hydrophobic particulate matter but within an hour it is converted to hydrophilic particles (Huang et al. 2010; Quinn et al. 2011). Thus, the black carbon particles are expected to be associated with sulphate aerosol and we hypothesize that sulphate acts as a kind of transport container for black carbon favouring the conditions for the long-range transport of black carbon to the Arctic.

Ozone and Mercury

In the early 1980s, Barrie et al. (1988) discovered that ozone was depleted every year at Alert and later Schroeder et al. (1998) showed that the ODE occurred simultaneously with atmospheric mercury depletion events (AMDE). Ozone and GEM were started in 1996 and in 1999, respectively, at VRS, see Fig. 3.5.

The challenges that we have worked with thereafter have been to determine the mechanisms behind this phenomenon and to determine the amount of mercury transported from the atmosphere to the Arctic environment (Ariya et al. 2008; Brooks et al. 2006; Christensen et al. 2004; Douglas et al. 2012; Ferrari et al. 2004; Goodsite et al. 2004, 2012; Moller et al. 2011; Skov et al. 2004, 2006a; Steffen et al. 2008).

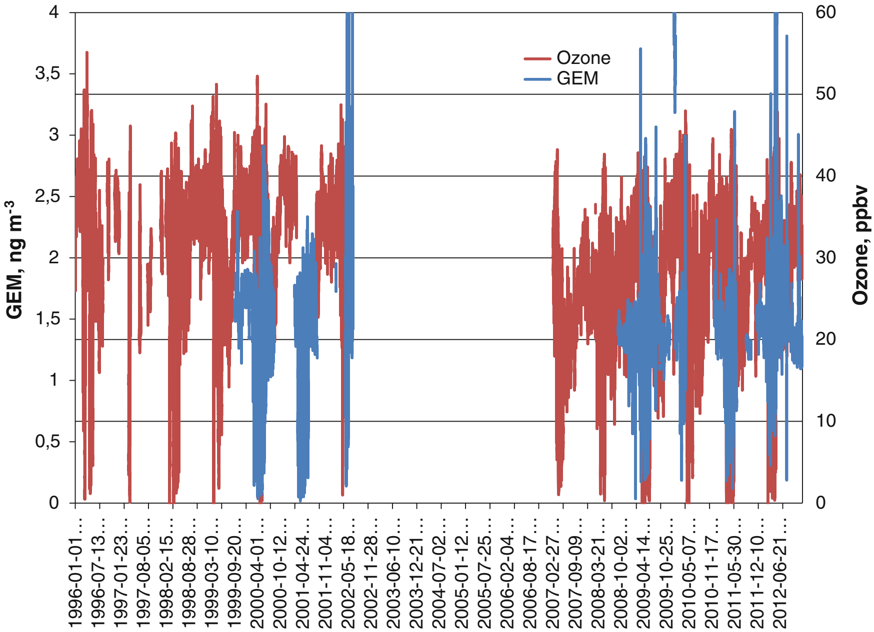

Fig. 3.5 GEM concentration (in; ng^{-3}) and ozone mixing ratio (in ppbv) at VRS

In the paper of Schroeder et al. (1998), it was shown that ozone and GEM concentrations were linearly correlated. Both ODE and AMDE were found to be correlated and to occur ubiquitous in the Arctic and, for example, also at VRS, north-east Greenland, see Fig. 3.5. In the paper of Skov et al. (2004), it was found strong evidence for that this correlation could be explained by competing reactions between a compound X with GEM or with ozone. The analysis showed that the removal of ozone and GEM agreed with Br chemistry. This result has been confirmed by theoretical studies (Goodsite et al. 2004, 2012). Thus, GEM reacts with Br in the Arctic

$$Hg + Br \rightarrow HgBr \quad (1)$$

followed by the addition of another Br atom

$$HgBr \rightarrow HgBr_2 \quad (2)$$

The results of the kinetic studies in the field, laboratories and from theoretical studies are summarized in a review (Ariya et al. 2008) and latest a new study (Dibble et al. 2012) and update of Goodsite et al. (2004, 2012) confirming the results though the reaction rates appear to be slightly slower than in the original paper.

However, Dibble et al. argued that the initially formed HgBr adduct will react further with NO_2 and other more abundant species than Br in the general troposphere. Thus, the second step in the oxidation of GEM to form Hg^{II} products is the first scientific priority in the road to fully understand the fate of atmospheric mercury.

The concentration of mercury species is far below the limit value where it can represent a health risk. The importance of the atmosphere is as transport media. The lifetime of mercury in the general atmosphere is long enough that it can be transported globally (Steffen et al. 2008) though we have found evidence from measurements in the marine boundary layer that the lifetime most probably is as short as about a month (Soerensen et al. 2010a, b, c). The relaxation time for GEM is most probably longer.

The precipitation in the high Arctic is very limited and, for example, at VRS it is about 200 mm $year^{-1}$. During AMDE, GEM is seen to be quickly removed followed by a build-up of gaseous oxidized mercury (GOM) which is also disappearing fast. Therefore, GOM must have a very fast deposition velocity. In a study of the fluxes of GOM and GEM at Barrow, Alaska, GOM was observed indeed to have a very fast deposition velocity leading to a surface concentration of mercury in snow as high as 600 ng L^{-1}. For comparison, it is typical around 1 ng L^{-1} in Denmark (unpublished results). The flux of GOM was measured by relaxed eddy accumulation and GOM was sampled on quarts denuders (Skov et al. 2006a). The flux was found to be very fast but with interference from reactions that sometimes gave fake emission signals. v_d was found to be between 1 and 10 cm s^{-1} and as a first approximation, the surface resistance r_c was calculated to be 0 for GOM. GEM was found to be re-emitted and thus for a 2 week period 1.7 µg Hg m^{-2} was deposited, 1.0 µg Hg m^{-2} re-emitted leaving a gain of 0.7 µg Hg m^{-2}.

The monitoring of GEM is continuing at VRS. A new set-up for measuring mercury fluxes has been designed that will make it possible to measure the fluxes of mercury species with higher time resolution and time coverage. Furthermore, it is planned to start measurements of BrO concentrations using maxDOAS instruments to get understanding of the chemical conversion and of the fluxes of mercury species from the atmosphere. Finally, wet deposition of mercury is measured within the framework of European financed project GMOS.

POP Measured in Nuuk and at VRS, Greenland

POPs Measurements in Nuuk

The POPs measured in Nuuk included a series of chlorinated pesticides (OCs), trifluralin, polychlorinated dibenzo dioxins and furans (PCDD/F), coplanar PCBs, polychloronaphthalenes (PCNs) and polybrominated diphenyl ethers (PBDEs). The results are summarized in Table 3.2.

Table 3.2 Annual mean and median concentrations of chlorinated pesticides, trifluralin, PCNs and PBDEs (pg m^{-3}), as well as PCDDs, PCDFs, co-PCBs (fg m^{-3} WHO-TEQ) in Nuuk together with minimum and maximum values. The d/n column indicates a number of detected concentrations above detection limit

Compound	d/n	Mean	Median	Min	Max
alpha-Chlordane	13/13	0.4	0.4	0.2	0.9
gamma-Chlordane	13/13	0.5	0.5	0.3	0.7
4,4′-DDT	1/13	0.2	0.2	n.d.	0.2
2,4-DDT	0/13	n.d.	n.d.	n.d.	n.d.
p,p′-DDE	13/13	0.4	0.3	0.2	0.8
o,p′-DDE	11/13	1.1	0.2	0.0	5.0
Dieldrin	7/13	2.5	2.6	0.9	3.6
Endosulfan I + II	13/13	6.3	5.9	3.7	10.3
Heptachlor epoxide	13/13	0.8	0.7	0.3	1.2
Hexachlorobenzene	13/13	128.1	*	*	*
α-HCH	13/13	18.3	*	*	*
γ-HCH (Lindane)	13/13	5.0	4.6	2.2	11.1
Trifluralin	4/13	0.2	0.2	0.0	0.6
ΣPCNs	13/13	0.2	0.2	0.06	0.3
ΣPBDEs	13/13	1.2	0.9	0.1	3.4
PCDDs	3/6	6.7	4.2	0.3	20.2
PCDFs	4/6	12.4	10.5	0.5	31.5
co-PCBs	3/4	0.6	0.6	0.2	1.0

*Not calculated due to missing results in the summer period (poor collection efficiency)

Generally, the average annual concentrations of OCs in the atmosphere of Nuuk were comparable to those measured in the Eastern Canadian Arctic, indicating a degree of uniformity in the contamination of Arctic atmosphere. The concentrations of OCs and trifluralin were clearly influenced by air mass transport from the North American continent. Model calculations using the DEHM were performed in order to correlate anthropogenic CO to atmospheric POP concentrations. Furthermore, POPs were correlated with temperature to see whether they followed Clausius–Clapeyron equation. The results showed a clear correlation with CO for trifluralin and Σ-BDE 47, 99 and 100 (penta-BDE), indicating an anthropogenic origin of these compounds from current use. Compounds such as dieldrin, heptachlor epoxide, γ-HCH and Σ-BDE 47, 99 and 100 had positive correlation with the temperature, which suggests that there evaporation energy is a determining factor for the observed concentration and thus re-emission from previously contaminated surfaces (soil or snow) is an important process for transport of these compounds to west Greenland. The annual variation profile of PCDD/F displayed a summer maximum that was suggested to come from local garbage burning. Very low levels of c-PCB concentrations were observed. They followed the annual profile of PCDD/Fs and were therefore likely to originate from the same source (Bossi et al. 2008).

POPs Measurements at VRS

POPs measurements started at the end of 2007 at VRS and the activity has been carried out since then with small interruptions due to technical problems (Bossi et al. 2013). Weekly atmospheric samples (about 5000 m^3) are collected with a high-volume sampler. Samples are collected on filters and PUF/XAD cartridges. The analytical programme has been extended to perfluorinated compounds (fluorotelomer alcohols, perfluorinated sulphonamides and sulphonamidoethanols). PCNs and PCDD/F are not included in the present programme. One sample is analysed each month for OCs and PBDEs, and one sample is analysed for perfluorinated compounds. All the other collected samples are stored at −18 °C for, for example, the analysis of new contaminants or other special analyses.

The data for organochlorine pesticides have been recently published (Bossi et al. 2013).

There are only few very minor sources of POPs in Greenland, and long-range atmospheric transport represents the largest and quickest source to the Greenlandic environment. One way to study atmospheric transport is to use atmospheric transport models as, e.g. the DEHM, presented earlier. The model covers the entire Northern Hemisphere, and in the model domain, all important sources for the Greenland/Arctic are included. This model has been developed further to include four chemical groups: a group related to ozone chemistry, a group related to primary particulates, a group with mercury species/chemistry and finally a group with POPs. The model has a spatially detailed 3D atmosphere up to 15 km over the

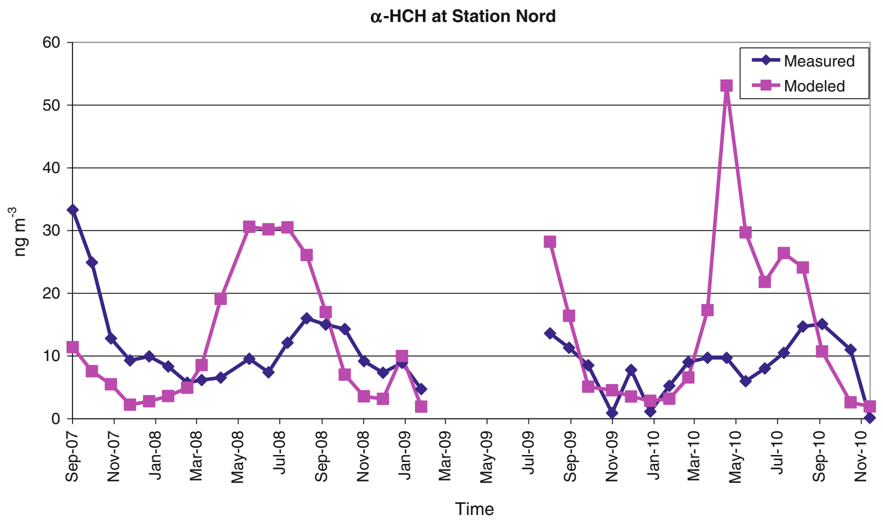

Fig. 3.6 Comparison between the measured and the modelled concentrations of α-HCH at VRS, North Greenland

surface. In addition, it has four surface compartments: a 75-m-thick ocean layer, a 15-cm-thick soil layer, and dynamic evolving vegetation cover and seasonal snow pack layer.

Model calculations have been made for a few POPs (HCHs and selected PCBs). These compounds have been selected because emission inventories are available, which is mandatory, and there are only emission inventories for limited number of compounds as these selected PCBs and HCHs. The yearly mean concentrations of α-HCH for the lowest atmospheric model layer are shown in Fig. 3.6. The level of α-HCH is captured well by the model but not the seasonal variation. This is most probably due to an overestimation of α-HCH concentrations in sea water. Work is currently under way to improve the parameterization of atmospheric α-HCH.

In 1990, the emissions of α-HCH were high in many countries in the Northern Hemisphere but in 2011 only a few countries in the Northern Hemisphere are emitting α-HCH. A large fraction of the atmospheric concentrations of α-HCH are due to re-emission from oceans, which was confirmed by the fact that concentrations follow Clausius–Clapeyron equation and are inversely proportional to the ice cover of the Arctic Ocean (Bossi et al. 2013). The origins of α-HCH in the oceans are only due to depositions in the past characterized by high anthropogenic emissions.

The results for Greenland show that the concentrations of α-HCH in 2011 compared to those in 1990 are factor of 3 lower, while concentrations of PCB-52 are a factor 7 lower (Figs. 3.7 and 3.8). This observation is again in agreement with the reduced emissions of these two POP’s. The emissions of α-HCH are reduced

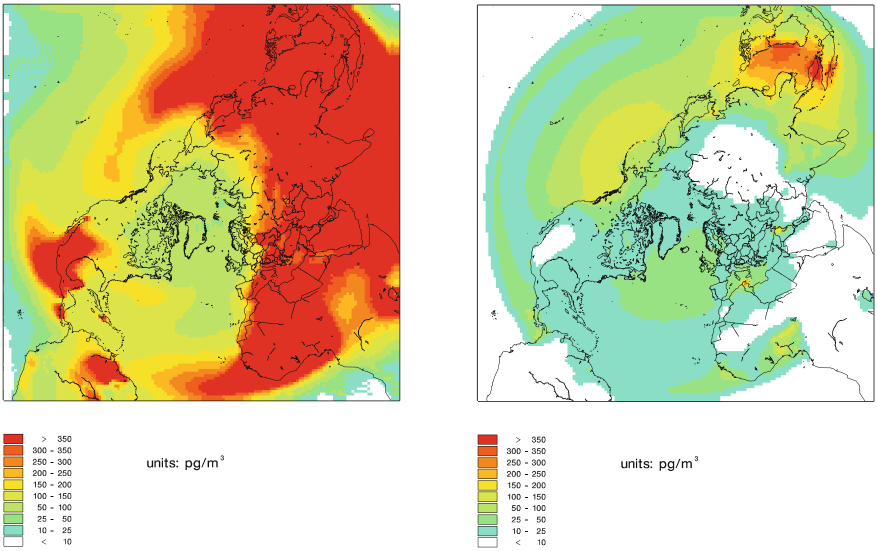

Fig. 3.7 Mean concentrations of α-HCH for the lowest part of the atmosphere for 1990 (*left*) and 2011 (*right*). Unit is pg/m^3

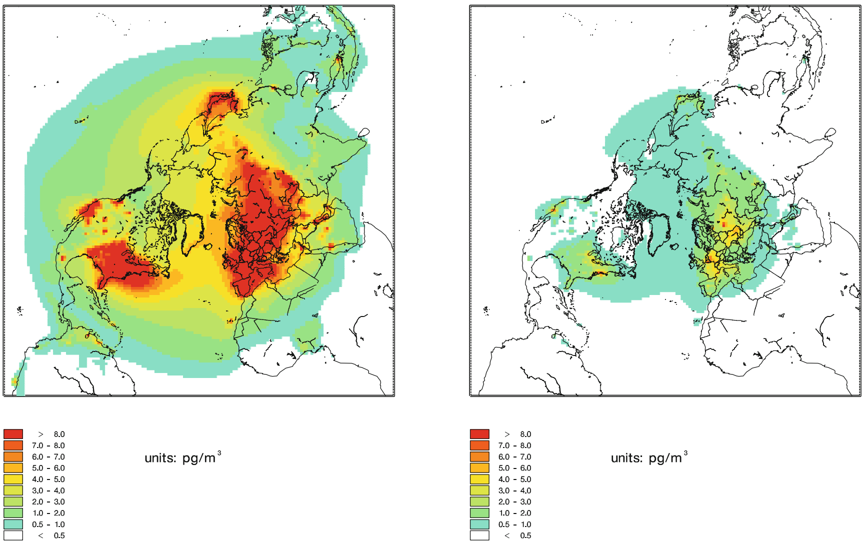

Fig. 3.8 Mean concentrations of PCB52 for the lowest part of the atmosphere for 1990 (*left*) and 2011 (*right*). Unit is pg/m^3

with factor more than 300 between 1990 and 2011, while the emissions of PCB52 is "only" reduced by factor 6, again indicating the importance of reemissions, especially for α-HCH.

Conclusion and Outlook

Time series of atmospheric pollutants at VRS are presented. Compounds and components that have either toxic effects on the sensitive Arctic environment or climate-forcing properties are prioritized. From measurements, DEHM model calculations and receptor model results we now know that pollution is transported from Eurasia into the Arctic during the winter half-year leading to a strong seasonal pattern. Specific sources have been identified for the individual compound or parameter. During spring and summer, evidence of locally produced particles is presented, which is hypothesized to originate from compounds released from frost flowers (in spring) by strong wind effects or from release from open leads or melting ponds (both spring and summer).

We have a good qualitative understanding of the processes during polar spring that remove ozone and convert GEM to GOM where GOM is removed fast by deposition and where Br appears to play a central role in the Arctic atmospheric chemistry.

The first years of data for a long series of POPs are now available. For legacy POPs with relatively high vapour pressure, a clear climate signal was identified with higher concentrations at high temperatures and at minimum ice cover. The seasonal pattern for these POPs could not be captured by DEHM most probably due to wrong concentrations in seawater. Model calculations show that, for example, α-HCH and PCB-52 have decreased with factors of 3 and 7, respectively, comparing 2011 with 1990. The main reason for this decrease is the decreasing emission due to international abatement strategies for these compounds. In the case of α-HCH, it is notable that the emission has been reduced with a factor 300.

Currently, we are measuring the following climate forcers at VRS: O_3, CH_4, CO_2, black carbon and particle size distribution. Work is undergoing to publish these data, and we here show the first data on black carbon.

One of the great challenges for global society over the coming century is impending climate change driven by anthropogenic greenhouse gas emissions. In the Arctic, the warming climate has already had strong impacts on ice sheets, sea-ice cover, ecosystems and biodiversity, with additional consequences for local communities, industries and international politics. These effects are likely to increase dramatically over the next 100 years with strong feedback effects for the rest of the world, and the next large environmental battle will most probably be in the Arctic where the needs for energy and other resources will be counteracted by the need to protect the sensitive Arctic environment and the climate of the Earth. The primary factors limiting research in the high Arctic are (i) lack of access and (ii) lack of well-equipped research infrastructures. It is thus of crucial strategic importance to establish new and well-suited research facilities in the high Arctic. Therefore, we have built a new research infrastructure: the VRS. It is equipped with state-of-the-art laboratory facilities and equipment. The station is a base for interdisciplinary work of the effect of climate change on the high Arctic physical, chemical and biological environment and how the environment may have impact back on the climate. Another important point is to assess the effect of new activities in the Arctic as oil drilling and mining activities. Though there has been gained a significant new knowledge in the Arctic, the high Arctic is still a region where our knowledge is most limited. For the atmospheric part, we will work on the following research questions:

- What is the origin, chemistry and physics, composition and effect of particles in the Arctic atmosphere?
- What is the vertical distribution of particles and other greenhouse forcers and how do they fit with model calculations?
- How important are the exchange processes between the atmosphere and the land, sea-ice and open sea for determining the load of black carbon as a climate-forcing agent, and which role does the transport and fate of xenobiotic compounds play?
- How are the chemical, physical and biological processes in snow and ice affecting the exchange processes between the atmosphere and the land, sea-ice and open sea as described above?
- How will new anthropogenic sources effect the Arctic environment?

Besides these points, there is a long series of related scientific questions focusing on other matrices such as sea, terrestrial and cryosphere but they are beyond the scope of this chapter as written in the introduction.

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Chapter 4
Photochemistry of Organic Pollutants in/on Snow and Ice

Amanda M. Grannas

Abstract Organic pollutant cycling and fate is impacted by the presence of snow and ice, which can serve as a repository for deposited species and also as a chemical reaction medium. Photochemistry (light-induced chemistry) occurring in/on snow and ice at Earth's surface is now known to play an important role in a variety of environmentally relevant processes including the production and release of atmospherically relevant species such as halogens, nitrogen oxides, and volatile organic compounds. Less is known about the role of snow and ice photochemistry in organic pollutant fate, but increasing recent evidence points to the potential importance of photochemical alteration of organic pollutants in sunlit snowpacks. This chapter describes recent work during and since International Polar Year (2007) aimed at investigating the potential importance of photochemistry to organic pollutant processing in snow and ice.

Introduction

A wide variety of organic pollutants including pesticides, herbicides, industrial chemicals, and various byproducts are used globally and emitted to atmospheric and aquatic systems. Through predominantly atmospheric and aquatic transport pathways, many of these compounds can be transferred to polar regions, far from the original emission site. Volatile and semi-volatile organic contaminants are volatilized in a region of higher temperature and undergo long-range atmospheric transport to a region of lower temperature, and then condense from the atmosphere on the surface. This results in relatively high concentrations of pollutants far from their source regions, a process known as the cold condensation effect, or global distillation. Figure 4.1 illustrates this effect for the legacy pollutant α-hexachlorocyclohexane (Arctic Monitoring and Assessment Programme 1998). A similar cold condensation

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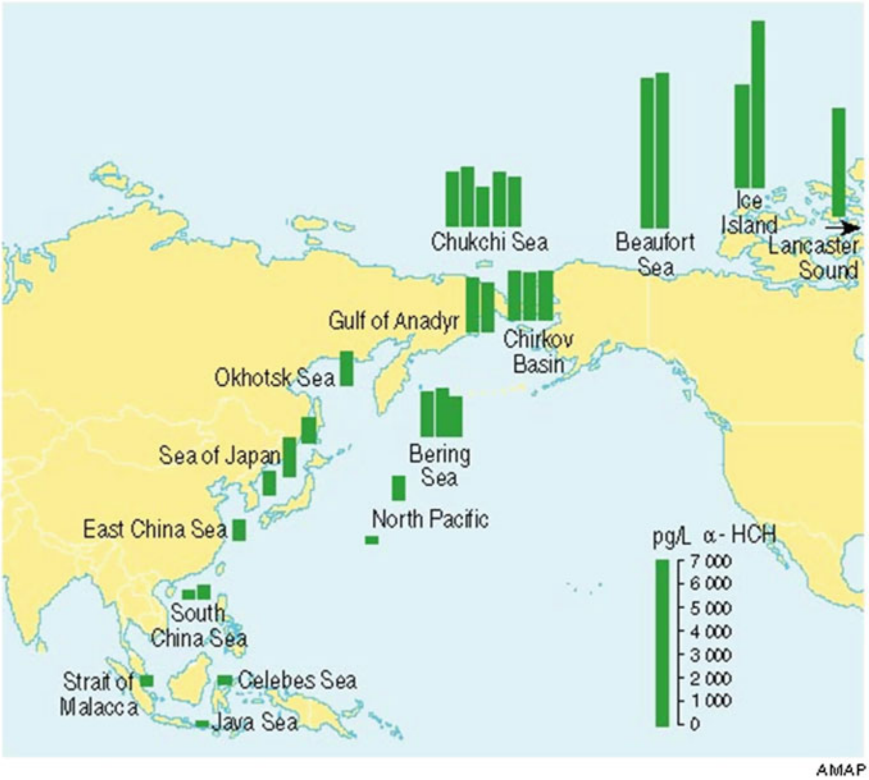

Fig. 4.1 Concentrations of α-hexachlorocyclohexane measured in seawater increase from lower latitude areas of emission toward the Arctic, an illustration of the cold condensation effect. AMAP Assessment Report: Arctic Pollution Issues, Arctic Monitoring, and Assessment Programme, 1998

effect has also been observed in high-altitude mountain regions (Daly and Wania 2005).

Organic pollutants are now known to be widespread in polar regions (e.g., Arctic Monitoring and Assessment Programme 1998, 2009), and a recent review has discussed the role of the global cryosphere in organic contaminant fate from both the standpoint of physical and chemical processing (Grannas et al. 2013). The negative health impacts of persistent organic pollutants (POPs) have long been reported in the literature. Such impacts include changes in eggshell thickness of birds of prey (Pearce et al. 1979; Ratcliffe 1967, 1970), altered gene expression in rats due to in utero exposure to POPs (Adeeko et al. 2003; Bondy et al. 2003), increased incidence of breast cancer in Greenland Inuit (Nielsen 2000), alterations to reproductive potential of fish-eating birds of the Great Lakes (Giesy et al. 1994) and Europe (Bosveld and Van den Berg 1994), reproductive impairment of seals in the Baltic Sea (Bergman and Olsson 1985), reproductive impairment of whales in the St. Lawrence seaway (Beland et al. 1993), endocrine disruption in humans and wildlife (Kavlock et al. 1996; Harrison et al. 1995), and immune system damage of wildlife (Ross et al. 1995; Safe 1994). Many POPs are listed as carcinogens by the US Environmental Protection Agency and European government agencies. Undoubtedly, the concerns over these myriad negative effects provide an impetus for the study of the environmental fate of organic contaminants and the potential for transformation of these pollutants into more, or less, toxic compounds. This is particularly important in Arctic regions where higher trophic level food sources are an important component of subsistence diets.

Photochemistry in/on Snow and Ice

Snow precipitation can be an effective scavenger of organic pollutants from the atmosphere, and the role of snowfall in contaminant delivery to Earth's surface has been summarized in a recent review by Grannas et al. (2013). The amount of scavenging that will occur depends on both the rate of snowfall and also the effectiveness of the snow crystal as a scavenger. The latter is dependent on the physicochemical properties of the pollutant, temperature, and snow physical properties. A review of air–ice chemical and physical interactions was recently produced by Bartels-Rausch et al. (2012), and a more detailed discussion of organic pollutant interactions with falling snow and fresh and aged snowpack is provided in a review by Grannas et al. (2013).

Snow and ice have historically been viewed as a relatively inert sink for atmospheric species. Recent studies, however, illustrate the previously overlooked importance of snow and ice as reactive chemical media in the environment. It has been shown that the Arctic snowpack plays an important role in processing atmospheric species, particularly via photochemical reaction. Photochemistry (referring here specifically to sunlight-driven chemistry at wavelengths >300 nm) in snow and ice is recognized as an important driver of gas fluxes from sunlit snowpacks and leads to significant impacts on the overlying atmosphere (Grannas et al. 2007a; Simpson et al. 2007). Arctic tropospheric ozone and mercury depletion events are inextricably tied to snow photochemistry processes (Simpson et al. 2007; Steffen et al. 2008; Pratt et al. 2013). Much work over the past decade has focused on better characterizing and quantifying snow and ice physical and photochemical processes (summarized in reviews such as Grannas et al. 2007a, 2013; Dominé et al. 2008; Simpson et al. 2007; Steffen et al. 2008; McNeill et al. 2012; Bartels-Rausch et al. 2012).

Any photochemical process begins with the absorption of light by a chromophore (light-absorbing constituent) imparting excess energy to the absorbing material and resulting in an excited state species. Once in the excited state, the chromophore may lose this excess energy by any number of processes. The various processes occurring upon absorption of radiation can be summarized by a Jablonski diagram, shown in Fig. 4.2, including chemical reaction (indicated here by R if unimolecular, B if bimolecular with a partner present at a concentration of [B]),

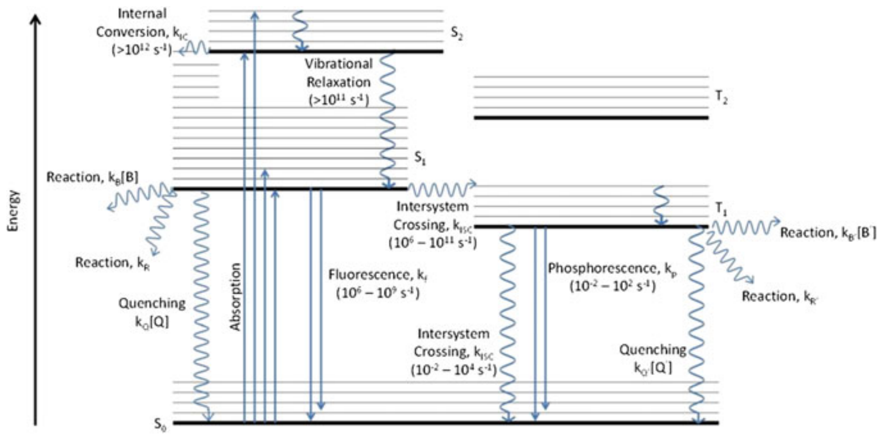

Fig. 4.2 Jablonski diagram, including processes that commonly contribute to a photochemical mechanism, with associated rate constants

quenching (Q) with a quencher molecule present at a concentration of [Q], internal conversion (IC, transition from a vibrational state of an electronically excited state to a vibrational state of a lower electronic state), intersystem crossing (ISC, transition to a state with a different spin multiplicity), or luminescence (emission of light at longer wavelengths than absorbed) including fluorescence (F, from singlet state) or phosphorescence (P, from triplet state). Processes involving absorption or emission of radiation are shown by straight arrows, while radiationless processes are indicated by wavy arrows. Corresponding rate constants are k_R, k_B, k_Q, k_{IC}, k_{ISC}, k_f, and k_p.

A chromophore may undergo any combination of these processes, and the efficiency of a given process is known as the quantum yield (Φ) for that particular process. For example, nitrate ion can absorb radiation and undergo any of three reactions (R1–R3), and the quantum yield of NO_2 production via R1 (relevant for snow photochemical production of NO_x) can be calculated by Eq. 4.1.

$$NO_3^- + h\nu \rightarrow NO_2 + O^- \qquad (R1)$$

$$NO_3^- + h\nu \rightarrow NO_2^- + O(^3P) \qquad (R2)$$

$$NO_3^- + h\nu \rightarrow {}^-OONO \qquad (R3)$$

$$\text{Quantum yield of } NO_2 \text{ production} = \Phi_{NO_2} = \frac{\text{number of } NO_2 \text{ molecules produced per unit time}}{\text{number of photons absorbed per unit time}}. \qquad (4.1)$$

or

$$= \frac{\text{moles of } NO_2 \text{ produced per unit time}}{\text{moles of photons absorbed per unit time (Einstein)}}.$$

The observed rate of a photochemical reaction will be a function of the intensity and energy of light available, the rate of photon absorption (quantified by the molar absorption coefficient, ε, of the molecule), and the quantum yield of the process of interest (Φ_x). Actinometry methods allow for the quantification of photon flux for a system of specific geometry and under defined spectral irradiance, and several reviews discuss chemical actinometers that are useful for various regions of the spectrum (e.g., Scaniano 1989; Kuhn et al. 2004). Several actinometry methods have also been established that are useful in frozen systems, including Galbavy et al. (2010) and Rowland and Grannas (2011).

In an irradiated snowpack, light penetration into the snow is an important variable that will impact the photochemical processes that may occur. Light penetration into an environmental snowpack is impacted by several variables including the physical characteristics of the snowpack, seasonality, and snowpack constituents (e.g., materials that contribute to absorption or scattering). For wavelengths of light relevant to most snowpack photochemical processes (UV and visible), absorption of light by snow is weak and scattering is dominant, and multiple scattering events enhance the effective pathlength of light penetrating an environmental snowpack (King and Simpson 2001). As such, even trace levels of absorbing materials can greatly impact the depth to which photochemically relevant wavelengths of light penetrate into snowpack. The light penetration depth is characterized by the e-folding depth, the depth at which light decreases to 1/e (37 %) of the incident light, and can vary greatly depending on snowpack properties. King and Simpson (2001) reported that 85 % of photochemical processes occur within two e-folding depths of the snow/ice surface. Propagation of light within snowpack and ice depends on snow density, grain size, and impurity content (Warren 1982). As such, layering in a snowpack will greatly influence light propagation, with e-folding depths varying from about 10 cm in cold, dry, and polar tundra snowpacks to 15–60 cm in warmer, wetter, and midlatitude (maritime) snow. King et al. (2005) reported an e-folding depth of about 50 cm for first-year sea ice in Antarctica.

Photochemical processes in snow and ice are uniquely impacted by the nature of the sample (compared to strictly liquid systems). It has been shown that a liquid-like (quasi-liquid or disordered) layer exists on the surface of pure ice (Faraday 1859; Wei et al. 2000) and that a disordered interface can also exist at the surface/grain boundaries/interstitial pores, etc., of ambient snow crystals and ice at temperatures relevant to polar conditions (Cho et al. 2002; Domine et al.2008). As ice forms from freezing water, pure water crystallizes first, and solutes are excluded from the bulk and concentrated at the surface (Koop et al. 2000). This freeze concentration effect will lead to a chemically "enriched" liquid-like layer at the surface, grain boundaries, and interstitial pores of snow and ice, which can affect the types of chemistry that can occur (e.g., enhanced potential to form coupling products due to a relatively concentrated medium at the surface). For example, recent work by Lignell and Gudipati (2015) showed that polycyclic aromatic hydrocarbons exist as aggregates rather than monomers trapped in crystalline water ice when cycled through temperatures that convert amorphous ice to crystalline ice. As such, organics are expected to be trapped not in the ice crystal lattice, but in the ice grain boundaries, which would be the active site of chemical processes. Potential reactants are highly concentrated (depending in part on their aqueous solubility), and the pH at the surface could be significantly different from what would be measured in a bulk sample. Many of these factors can influence photochemical processes by altering the relative effectiveness of excited state quenching versus photochemical product formation (Bower and Anastasio 2013); altering the chromophoric nature of the absorbing species (Heger et al. 2005; Kahan and Donaldson 2010); or potentially changing the light flux within the sample due to scattering of light from solid ice surfaces (or scattering of light as a function of depth at the snowpack scale) (Fisher et al. 2005).

Much of snow and ice photochemistry research to date has focused on the behavior of inorganic and small organic molecules. For example, photochemical production of formaldehyde, acetaldehyde, acetone, molecular halogens, alkyl halides, HONO, and NO_x have been reported to occur in snow/ice in polar regions, and mechanisms for many of these processes have been proposed/investigated (Dominé and Shepson 2002; Grannas et al. 2007a; Simpson et al. 2007; Hamer et al. 2014). A body of astrophysical literature also exists related to photochemistry of organics on interstellar ices (e.g., Ashbourn et al. 2007; Evans et al. 2012; Nuevo et al. 2012; Menor-Salyan and Marin-Yaseli 2013); however, these processes involve radiation wavelengths and temperatures typically not relevant to Earth's surface. The photochemical transformations of anthropogenic organic pollutants in ice also have been observed, although published information regarding these processes has historically been limited.

Initial Evidence of the Environmental Importance of Photochemistry in Snow and Ice

A complex picture of active photochemistry in snow and ice has emerged over the past ~15 years of research, and this chemistry is now recognized as a potentially critical component of biogeochemical cycles, particularly in polar regions. A variety of large-scale field campaigns were initiated during International Polar Year (IPY 2007–2009) to study environmental interactions in polar regions, and these have shed light on the complex biotic and abiotic processes occurring in all phases of the polar environment, including ocean, atmosphere, sea ice, and snowpack (termed OASIS interactions, Shepson et al. 2012). Figure 4.3 illustrates the myriad interactions that may occur and fluxes that result in part from photochemical reactions in snow/ice. Missing from this illustration, however, is the potential cycling and fate of organic contaminants in these environments, which certainly could have an impact on the biological species underlying sea ice, or could be re-emitted to the atmosphere under a changing climate. Figure 4.4 illustrates similar interactions that may occur within the cryosphere, but focuses specifically on the fate of organic contaminants in these regions.

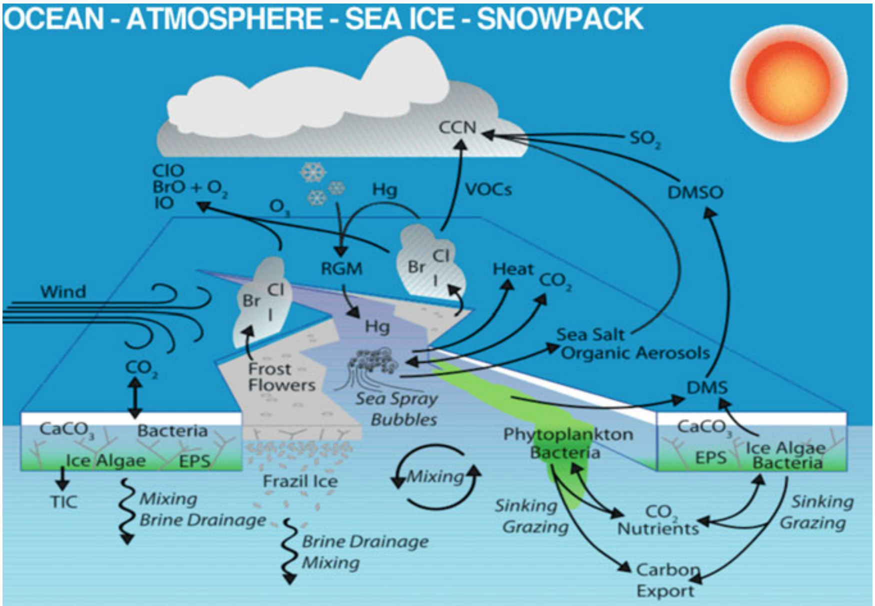

Fig. 4.3 Schematic of some ocean–atmosphere–sea ice–snowpack interactions among the chemical, physical, and biological processes in polar regions. These include but are not limited to feedbacks involving such chemical species as volatile organic compounds (*VOCs*), cloud condensation nuclei (*CCN*), total inorganic carbon (*TIC*), dimethyl sulfoxide (*DMSO*), reactive gaseous mercury (*RGM*), dimethyl sulfide (*DMS*), exopolymeric substances (*EPS*), and other molecular exchanges, as shown

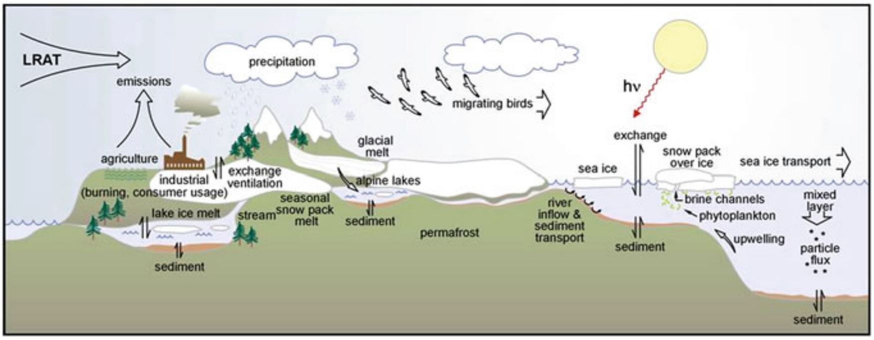

Fig. 4.4 Schematic illustrating the various elements of and interactions occurring in the cryosphere that impact contaminant cycling and fate. Long-range atmospheric transport (LRAT) is primarily responsible for the global distribution of contaminants, but the various exchanges represented here play important roles in cycling and redistribution of contaminants. Reproduced from "The Role of the Global Cryosphere in the Fate of Organic Contaminants." Grannas et al. (2013), Creative Commons Attribution 3.0 License

Studies reported since 2000 point to photochemistry as a potential transformation mechanism of organic contaminants in snow and ice. In Klán and Holoubek (2002) provided an initial review of the relatively scant (to that point in time) available published information on organic contaminant photochemistry in snow/ice. These initial studies indicated that active contaminant photochemistry in snow/ice was possible and the rates and mechanisms of degradation could be quite different in frozen systems compared to liquid water (Holoubek et al. 2000; Klán et al. 2000a, b, 2001). These early papers reported unusual behavior of halobenzene photochemistry in ice, where the products observed in ice were different from those observed in liquid water, as illustrated in Fig. 4.5. Due to the freeze concentration effect, coupling products (bi- and triphenyls) were observed in ice, rather than products resulting from photosolvolysis from the water solvent (e.g., phenols).

The observed production of triphenylene was unique to ice and provided evidence of the importance of the exclusion of solutes to liquid-like layers in ice upon freezing and how that process might impact reaction mechanisms in snow/ice compared to liquid water. The authors also proposed this chemistry as a potential secondary source of pollution in polar regions. Around this same time, a study was reported by Dubowski and Hoffmann (2000) indicating that the photochemical reaction of 4-nitrophenol in ice pellets resulted in the same photoproducts as occurs in liquid water (hydroquinone, benzoquinone, 4-nitrosophenol, etc.), and they suggested that a similar mechanism was responsible for both liquid and ice photochemical degradation. It was clear from the initial literature that photochemistry in snow/ice might be an important component of pollutant fate, but that the

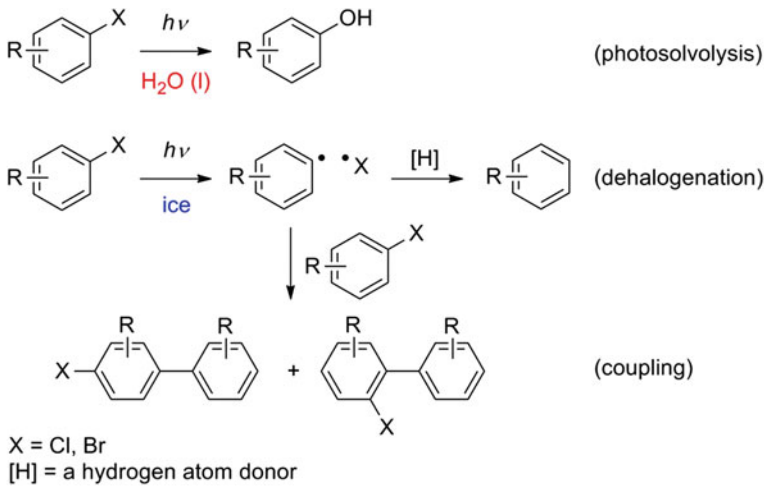

Fig. 4.5 Comparison of photochemistry of haloarenes in water and frozen aqueous solutions, illustrating the different mechanisms and products that occur in liquid water and ice. Reproduced from "The Role of the Global Cryosphere in the Fate of Organic Contaminants." Grannas et al. (2013), Creative Commons Attribution 3.0 License

mechanisms and reactivity of these systems could not necessarily be extrapolated from liquid conditions for all analytes. This initial work provided the impetus to continue the study of snow/ice contaminant photochemistry, and much progress has since been made in this arena.

Progress in Understanding Organic Pollutant Photochemistry in Snow and Ice

Numerous investigations since 2000 have unambiguously shown that organic compounds, including anthropogenic contaminants, can undergo photochemical changes in snow and ice. These degradation pathways can be brought about by *direct* or *indirect* photochemical reactions. *Direct* photochemistry results when a chromophoric contaminant absorbs radiation directly and undergoes some chemical transformation as a result. For species that are not chromophoric, photochemical degradation is still possible via *indirect* pathways. In this case, a chromophoric species present in the snow or ice (known as a photoinitiator) can absorb radiation and produce a reactive intermediate (such as OH radical or singlet oxygen). This intermediate subsequently reacts with the non-chromophoric contaminant. Hydrogen peroxide, nitrate, nitrite, and organic materials have been shown to be efficient photoinitiators in sunlit snow and ice (see, e.g., Jacobi et al. 2006; Ram and Anastasio 2009; Beine et al. 2012; Grannas et al. 2014).

The direct photochemical degradation of contaminants can be different in snow/ice compared to liquid water. This can be due to several factors including the freeze concentration effect (as discussed in Section "Initial Evidence of the Environmental Importance of Photochemistry in Snow and Ice"), changes in pH upon freezing [known as the freezing potential (Workman and Reynolds 1950)], and changes in the chromophoric nature of the contaminant upon freezing. The freeze concentration effect was shown to increase the rate of bimolecular reactions of organics in ice (Grannas et al. 2007b) and increased the probability of coupling reactions over photosolvolysis reactions for chlorophenols (Klánova et al. 2003a, b). Ruzicka et al. (2005) investigated cage effects at the grain boundaries of ice and found that the freeze concentration effect caused efficient molecular segregation of their probe molecules (dibenzyl ketone and 4-methyldibenzyl ketone) in frozen aqueous solutions over a temperature range of 273–193 °K (Ruzicka et al. 2005). Heger et al. (2005) used the well-characterized dye methylene blue to study the dynamics of the freeze concentration effect. Characteristic spectral shifts upon freezing, combined with quantitative spectroscopy, were used to determine that the nature of the freezing process could impact the freeze concentration process, e.g., fast freezing at 77 °K resulted in an approximately threefold increase in apparent concentration at ice grain boundaries, whereas slower freezing at 243 °K resulted in a sixfold increase in methylene blue concentration at grain boundaries in the ice. Thus, the formation process of snow and ice could impact the distribution of species within the sample (and thus the chemistry that might happen in turn).

The absorption characteristics of organic contaminants may change when incorporated into snow/ice, which could also impact their photochemical fate. For example, Matykiewiczová et al. (2007) and Kahan and Donaldson (2010) showed that (when frozen) the spectra of phenol derivatives and benzene exhibit bathochromic shifts into the region of the electromagnetic spectrum that overlaps with available solar radiation at Earth's surface. This phenomenon increases the likelihood of photochemical degradation under environmentally relevant conditions in snow/ice matrices.

When solutes are excluded to liquid-like layers during freezing, some species are more readily segregated than others. For example, when freezing a solution containing sodium chloride, the sodium is preferentially excluded to liquid layers more so than chloride. Negatively charged hydroxide ions present in water are drawn to the liquid-like layers to offset the charge imbalance, which produces a more basic environment in the liquid-like layers than pure water. This preferential exclusion of some solutes to liquid-like regions in a frozen sample is called the "freezing potential" (Workman and Reynolds 1950). The existence of the freezing potential phenomenon results in highly variable pH of liquid-like layers, depending on the solutes present in the sample. If the chromophoric behavior of a molecule is pH-dependent, the freezing potential will impact photochemical degradation due to shifts in the absorption spectrum. This has been observed by Heger et al. (2006) who found varying degrees of cresol red protonation depending on the sample phase, as well as due to the presence of other solutes (e.g., NaCl or NH_4Cl which could induce different freezing potential effects).

Several techniques have been used to generate snow and ice samples for examination in the laboratory setting. The majority of work published to date has used ice samples prepared by freezing the aqueous solution containing the solutes/analytes of interest. The ice could subsequently be crushed to smaller crystal sizes (e.g., to change total sample surface area). Although this does not represent the formation mechanism of snow crystals in the environment, arguments have been made that the microscopic domains where most solutes will reside are still consistent with those in environmental snows/ices (Guzman et al. 2007) and represent an adequate mimic for laboratory-based experimentation. Other sample preparation methods include "snow" produced by a shock-freezing method that could subsequently be exposed to gas-phase organic compounds, or containing contaminants from the aqueous solution used to make the snow. These methods have been used to investigate air/ice surface chemical exchange processes, as well as photochemistry of hydrophobic organic compounds (Matykiewiczova et al. 2007; Heger et al. 2011; Kurkova et al. 2011; Ray et al. 2011). The surface coverage of hydrophobic organics on artificially generated snow grains produced by the shock-freezing method was estimated using a photofragmentation process (Heger et al. 2011; Kurkova et al. 2011) or ozonation reaction (Ray et al. 2011). An interesting result of this work was the observation of greatly enhanced ozonation reaction rates for 1,1-diphenylethylene at the ice–air interface (increasing reaction rates with decreasing sample temperature). This was attributed to an increase in the number of surface reactive sites on the ice and possibly a higher ozone uptake coefficient to ice (Ray et al. 2013). Surface-specific reaction rate enhancements have been observed when organics self-associate on the ice/snow surface. This has been shown to impact the rates of photochemical reaction of anthracene (Kahan et al. 2010), benzene (Kahan and Donaldson 2010), and methyldibenzyl ketone (Kurkova et al. 2011) and ozonation of phenanthrene (Kahan and Donaldson 2008).

A number of recent studies have focused on the photochemical degradation of environmentally relevant organic contaminants and POPs. A motivating research question is how photochemical processing could impact the fate of contaminants in polar regions and whether these processes could produce environmentally hazardous photoproducts. Studies have been reported for species such as halogenated aromatic compounds (e.g., chlorobenzenes, chlorophenols, PCBs, aldrin, and dieldrin) (Klán et al. 2000a, b, 2001; Klánova et al. 2003b; Literak et al. 2003; Dolinova et al. 2006; Matykiewiczová et al. 2007; Rowland et al. 2011), nitroaromatic compounds (Dubowski and Hoffmann 2000), and organophosphorus compounds (Weber et al. 2009). Of interest is not only what photochemical reactions might occur, but also the physical fate of the contaminants (or byproducts) upon reaction. For example, production of higher vapor pressure products could lead to increased evaporative losses of contaminant from snowpack, while production of lower vapor pressure (or more hydrophilic) species could result in more efficient transfer of photoproducts with subsequent snowpack melt waters. Matykiewiczová et al. (2007) used a laboratory approach to investigate the photochemistry of PCBs in artificial snow at environmentally relevant concentrations and simultaneously monitored the photochemical changes and volatilization fluxes of PCBs from the solid ice matrix. They found that the major degradation pathway was reductive dehalogenation, but a desorption process competed with photochemistry for the observed contaminant loss from snow. The authors also investigated the role of photooxidation (by OH radicals) of PCBs and found that in cases where sufficient amounts of OH precursors were present in the snow, photoinduced oxidation processes could surpass both photoreduction and volatilization losses of PCBs as the major sink for this species in a sunlit snowpack at environmentally relevant conditions.

The majority of available literature has focused on investigation of photochemistry in controlled laboratory settings. However, several studies conducted in the field also point to the potential importance of photochemical degradation of contaminants in/on snow and ice surfaces. Klán et al. (2003) showed that several types of organic compounds (aromatic carbonyl, chloro, nitro, and hydroxyl species) were susceptible to photochemical reaction using ice samples prepared from clean water, but irradiated in natural sunlight in Svalbard, Arctic Norway. Alarmingly, they found that the observed photoproducts could pose a high toxicological risk to biota if they entered the environment. Other recent work has focused on the potential role of snowpack photoinitiators on contaminant degradation. As part of an IPY activity, Rowland et al. (2011) studied the photochemical loss of several organochlorine POPs, via both direct and indirect photochemical reaction during a field campaign in Barrow, Alaska. Samples containing POPs were made using snow collected from the Barrow area, and a number of control experiments were conducted to investigate the role of OH radical chemistry on POP degradation. They found that samples made using Barrow snow (thus containing the various photoinitiators that would be present in a natural snowpack) were the most highly reactive, even when compared to a control sample containing 500 μM H_2O_2. (Typical Barrow snow contains on the order of 0.2 μM H_2O_2). This rather surprising level of photochemical reactivity was hypothesized to result from natural organic matter present in the snowpack, which could play an important role in indirect photochemistry, producing species such as OH, singlet oxygen, peroxy radicals, and triplet state organic matter. A follow-up study by Grannas et al. (2014) investigated this potential role of natural dissolved organic matter in ice photochemistry, using commercially available humic and fulvic acids of both terrestrial and microbial origin, as well as an authentic Arctic organic material sample. They found that the dissolved organic matter played an important role in indirect photolysis processes in ice through production of reactive oxygen species such as OH and singlet oxygen, which could then subsequently react with organic contaminants present in the ice. IPY work of Beine et al. (2012) indicates that chromophoric organic matter is a dominant absorber in snow/ice/frost flowers in the Barrow region, and they calculated that it could be the dominant source of reactive OH radicals in these matrices. These studies point to the importance of natural organic material as an important component of snow/ice photochemical processes and generation of reactive intermediates that may impact organic contaminant fate.

In addition to natural organic materials that may impact contaminant fate through indirect photochemical processes, the presence of inorganic species (e.g., transition metals) may also influence reactivity. Kim and Choi (2011) showed that the reduction of Cr(VI) by various organic acids was negligible in aqueous solution but was significantly accelerated in ice, ascribed to the freeze concentration of both electron donors and protons in the ice crystal grain boundaries.

In cases where active photochemical degradation of organic contaminants can occur in snow/ice, it is imperative to assess the nature of the products that are formed. The toxicity or bioaccumulation potentials of photochemical products could be very different from the original contaminant. For example, Rowland et al. (2011) found a major product of aldrin photochemical degradation in snow/ice was dieldrin—a species that is much more toxic than the original pollutant. Blaha et al. (2004) evaluated the toxic effects of products formed upon photolysis of 2- and 4-chlorophenol in ice using a bacterial luminescence test. They found that irradiated ice samples elicited significant inductions of dioxin-like effects and were far more toxic than chlorophenols irradiated in liquid water. In this case, the products formed in ice were primarily chlorobiphenyldiols, while dihydroxybenzenes were produced in liquid water. The results from these types of studies indicate that photochemical degradation of contaminants could have unexpected impacts in snow-/ice-covered regions and may play an important, but poorly understood, role in ecosystem health.

Conclusion and Future Directions

Although much progress has been made in the field of snow and ice photochemistry since 2000, this research is still in its infancy. There are many challenges associated with both laboratory- and field-based approaches, particularly for the study of organic contaminant degradation. One main limitation is due to the low concentrations of contaminants that are typically present in natural snow and ice samples, requiring large volumes of sample (many liters of meltwater) to quantify contaminants at their native concentrations. For researchers attempting to mimic environmental photochemistry in the laboratory, generating samples of this size is cumbersome if not impossible. As such, laboratory studies often rely on the use of samples with elevated contaminant concentrations.

Another limitation to laboratory-based investigations of snow/ice chemistry is the method of sample generation. Most natural snow forms from water condensing from the gas phase onto an ice-forming nucleus; however, few studies have made snow/ice in this manner. It is certainly easier (and likely more reproducible) to generate ice of a given composition by freezing the solute-containing water; however, Loewe et al. (2011) have proposed a method to make snow by condensing water vapor. Arguments have been made both for and against the applicability of laboratory-generated snow/ice samples to mimic environmentally relevant samples (Guzman et al. 2007; Domine et al. 2013). An additional difficulty of making laboratory snow/ice mimics is the appropriate way to incorporate particulate matter, which is invariably present in natural snow and ice samples. No one to date has investigated the role of solute/particulate interactions in the liquid-like interfaces of snow and ice. These particulates could be relevant photochemically, particularly species containing organics or metals. Particulate species could serve as effective photosensitizers, could impact gas/surface partitioning, or could impact photochemistry due to indirect light-screening effects. As discussed in Grannas et al. (2007a), the research community still lacks a comprehensive understanding of both the macroscopic and microscopic incorporation of impurities (both dissolved and particulate) into snow/ice and how this might impact contaminant photochemical fate.

The role of halogen chemistry, related to the widespread observed ozone depletion events that occur in polar regions (Simpson et al. 2007), is an additional unknown that could impact organic contaminant fate. The release of significant amounts of photochemically reactive halogen species could result in an additional contaminant oxidation pathway above sunlit snowpacks or within the interstitial air of snowpacks. Of particular interest is the significant amount of Cl_2 released (which readily photolyzes to form Cl radicals), as measured for example in Barrow, Alaska (Liao et al. 2014). Although gas-phase OH radical reaction rate constants have been determined for many organic contaminants, similar values are not readily available for Cl atom reaction. For many organics, the Cl atom reaction can compete with the OH radical reaction pathway (or becomes dominant) under high Cl atom concentration conditions. It will be important to assess the potential for halogen atom reactions (particularly Cl) with contaminants in the gas-phase as a mechanism that could impact contaminant fate in polar regions.

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Chapter 5
Aeolian and Ice Transport of Matter (Including Pollutants) in the Arctic

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Introduction

Numerous studies have shown that aerosols are of importance for atmospheric chemistry and climate of the Arctic (Pacyna 1991; Barrie 1996; Leck et al. 1996; Kutsenogii 2006; Shevchenko 2006, 2010) and other parts of the World Ocean (Duce et al. 1991; Lisitzin 1996, 2011). The rapid movement of air makes it an important pathway for delivering heavy metals, black carbon, and other contaminant to the Arctic. The atmosphere is the fastest and most direct route from the sources of pollution: Transport from the sources to the Arctic occurs in a matter of days or weeks (Pfirman et al. 1995; Shevchenko et al. 2003; Kallenborn et al. 2007; Vinogradova et al. 2008a; Hung et al. 2010).

Black carbon (BC) or soot is the part of carbon-containing aerosol particles which absorbs the sunlight and a lot of pollutants (heavy metals, PAH, PCB, etc.) (Quinn et al. 2008, 2011; Doherty et al. 2010). BC is a product of incomplete combustion of various fuels (especially, coal and diesel oil), biomass (forest, grass, and agricultural waste), and of biofuels. Being adsorbed on sooty particles, sulfate radicals, heavy metals, and dangerous for human health persistent organic pollutants (POPs) are transported by air masses for hundreds and thousands kilometers,

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thus polluting the Arctic environment. As the lifetime of aerosol particles in the atmosphere is not longer than a month, the main part of them is precipitated on the surfaces of coastal seas and the Arctic Ocean, polluting the snow cover and accumulating in it.

We have limited data on BC concentrations in air and snow for the Russian Arctic, while in other arctic regions experimental investigations have been carried out during the last 30 years. That is why studying of BC and soot in lower atmospheric layers (near land and water) is of special importance for the Russian Arctic.

There are several more or less localized industrial regions in Russia (the Ural, the Norilsk region, the Kola Peninsula, and others) which produce intense atmospheric emissions being of importance on a global scale. For the Arctic environment, the influence of large industrial complexes located inside the Arctic (on the Kola Peninsula and in the Norilsk region) cannot be neglected. These sources emit to the atmosphere anthropogenic copper and nickel, as well as lead, black carbon, and other pollution. Real monitoring of environmental state throughout the large territory of the Russian Arctic is limited by economic and technical problems. Thus, modeling atmospheric transport and estimates of anthropogenic pollution is useful approach for partial solution of this task.

Materials and Methods

Black carbon concentrations in the marine boundary layer over the White, Barents, and Kara seas were studied in August—September 2007 in 53rd and 54th cruises of the RV "Akademik Mstislav Keldysh" using aethalometer (Hansen et al. 1984, 1997; Pol'kin et al. 2004, 2008, 2011; Watson et al. 2005). The studied area is shown in Fig. 5.1.

In April 2008, snow, ice, and under-ice water were studied in the vicinity of the North Pole at the area from N 88°19.89′ to N 88°30.28′ and from E 014°16.95′ to E 017°22.42′ (Fig. 5.1) during the Pan-Arctic Ice Camp Expedition (PAICEX). Samples were collected at the distance more than 4 km from the Barneo ice camp. Snow from upper 7 cm was collected in clean plastic buckets. At the same places, ice cores were taken by titanium Cherepanov's ring ice-corer, inner diameter 14 cm (Russia). Parts of ice cores were collected in the clean plastic buckets also. Under-ice water samples were taken by Niskin bottles (General Oceanic Inc., USA). Melted snow and ice and under-ice water were filtered through pre-weighed nuclear pore filters (diameter 47 mm, pores 0.45 μm, Dubna, Russia). Filters after drying at 50 °C were weighed and particulate matter amounts were calculated gravimetrically.

Scanning electron microscopy of particulate matter from snow, ice, and under-ice water was carried out on pieces of filters at JSM-U3 microscope (Jeol, Japan) in P.P. Shirshov Institute of Oceanology, Moscow.

For modeling, we used two approaches—"forward" and "backward" trajectory estimations. The first approach investigates forward transport of air masses and pollution from potential anthropogenic source and spatial pollution distributions

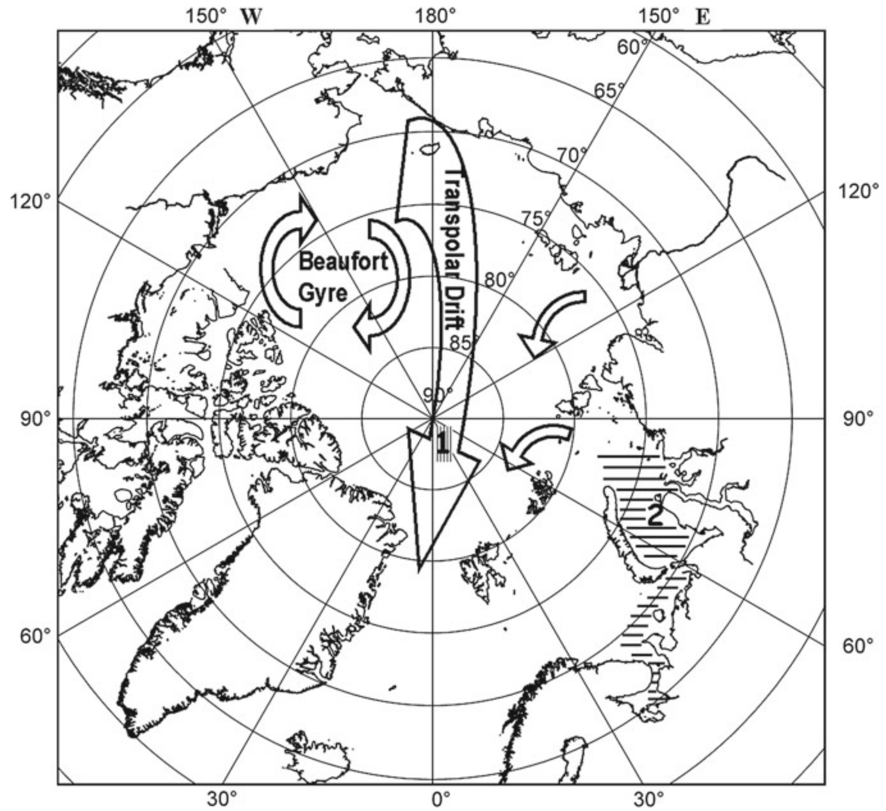

Fig. 5.1 Major routes of ice drift (Lisitzin 2002) and position of areas of author studies in 2007–2008: *1* vicinity of the North Pole; *2* the White, Barents, and Kara seas

around it. The second model investigates the backward transport of air and pollution to selected target locations from potential anthropogenic sources.

In both cases, the 5-day air mass trajectories (forward or backward) for selected locations were computed using Hybrid Single Particle Lagrangian Integrated Trajectory Model (Draxler and Rolph 2003) and NCEP/NCAR Reanalysis Data Files for simulations. Calculations were done for 925 and 850 hPa levels, all trajectories started at 00 UTC (with calculating interval of 1 h). In total, trajectories were computed for every day in January, April, July, and October (assuming that these months are representative for corresponding seasons) during the last three decades (for 1980s, 1990s, and 2000s) for every location. The decadal averaged spatial distributions of trajectories and pollution characteristics (air concentration and flux onto underlying surface) were analyzed. Assuming equal seasons, annual values and totals were calculated, and long-term variations were assessed in seasonal and annual values by comparing results for selected three decades.

Atmospheric transport of heavy metals (HM) from the sources was evaluated using approach by Vinogradova (2000), Galperin et al. (1995), and Rovinskii et al. (1994).

For simplicity reasons, it has been assumed that about 20 % of the receiving emissions were deposited onto the surface near the expected source locations (within a range of 30–50 km), and the rest 80 % have been subject to further long-range atmospheric transport. Pollutants are assumed to be vertically homogeneously distributed within the topospheric mixing layer H of until 1 km altitude. In cold seasons, this altitude is determined by the well-defined surface air temperature inversion. The longer the time pollution moves along a calculated trajectory, the lesser amount will remain in the atmosphere. It is assumed that the remaining amount is nearly proportional to exp ($-Kt/H$), where K is the deposition velocity, and t is the real time of air transport from the source. The deposition velocities onto the surface (sum of dry and wet components: $K = K_D + K_W$) are assumed to be the same for all pollutants under investigation because anthropogenic heavy metals are transported for a long distance mainly associated with submicron aerosol particles (Galperin et al. 1995), but the seasonal variations of K (based on literature data generalization) are taken into account. Thus, our results may be used for environmental pollutants with assumed deposition velocities as follows: 0.05–0.07 (W), 0.1–0.2 (Sp), 0.9–1.2 (Su), and 0.4–0.8 (F) [in cm/s] in winter, spring, summer, and fall, respectively. Our estimates are mainly for

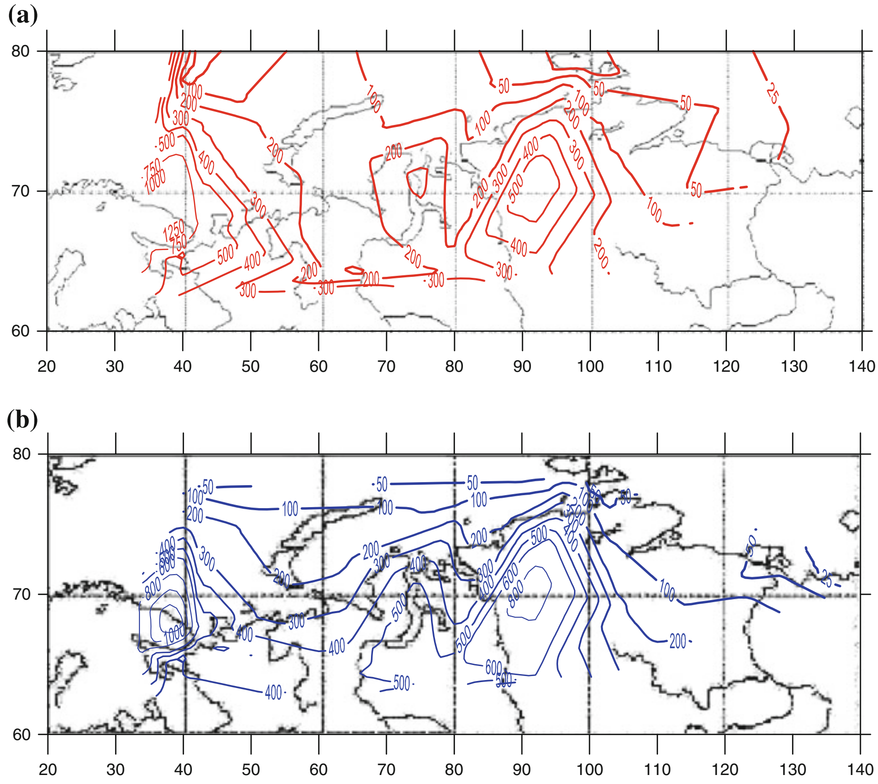

Fig. 5.2 Spatial average flux distribution (for the 2000s) onto the surface of anthropogenic Ni (**a**) and Cu (**b**), coming through the atmosphere from Kola Peninsula and Norilsk region, g/km^2/year

anthropogenic heavy metals (HM). However previous results for anthropogenic black carbon (BC) are also available (Vinogradova and Veremeichik 2013a, b).

The forward trajectory approach was applied for two major anthropogenic source regions for the central Arctic, namely the Kola Peninsula and the Norilsk region—situated in the Russian sub-Arctic/boreal region (Vinogradova et al. 2008a, b, c; Baklanov et al. 2013). The decadal averaged spatial distributions of forward trajectories and pollution characteristics from each source for selected months were calculated in each cell of spatial grid (1° × 1°) and mapped (Fig. 5.2).

The backward trajectory approach was applied for three receptor locations at Russian Arctic Islands (Vinogradova 2000; Vinogradova and Ponomareva 2001, 2012)—Franz Josef Land (FJL), Severnaya Zemlya (SZ), and Wrangel Island (WI). The annual source emission characteristics during the last three decades were estimated from available the data published by Roshydromet (Russia) from 1990 to till 2009 (Annual emission of pollutants in atmospheric air of cities and regions of the Russian Federation in 2007, 2008; Pacyna et al. 1984, 1985).

Results and Discussion

Black Carbon

The average BC concentrations in the marine boundary layer over the White Sea in August in different years varied from 0.12 to 0.55 μg m^{-3} (Table 5.1) depending mainly on the local meteorological conditions; the lowest for the White Sea BC

Table 5.1 Black carbon concentrations in the atmosphere over the Russian Arctic seas and adjacent land (μg m^{-3})

Place	Time	BC concentration (μg m^{-3})	Source
Tiksi	February 1995	0.31	Fukasawa et al. (1997)
Tiksi	May 1995	0.081	Fukasawa et al. (1997)
Tiksi	July 1995	0.064	Fukasawa et al. (1997)
Severnaya Zemlya	March–May 1990	0.014–0.514	Polissar (1993)
N Barents Sea near Franz Josef Land	September 1998	0.07	Kopeikin et al. (2010)
White Sea	August 2003	0.55 ± 0.6	Pol'kin et al. (2004)
White Sea	August 2006	0.12 ± 0.13	Kozlov et al. (2009)
White Sea	August 2007	0.31 ± 0.23	Pol'kin et al. (2008, 2011)
Kara Sea	September 2007	0.09 ± 0.21	Pol'kin et al. (2011)

concentrations were registered in its central part, and highest values were in the vicinity of industrial center Arkhangelsk (Pol'kin et al. 2008).

The BC concentrations in the marine boundary layer over the Kara Sea in September 2007 were lower than found for the White Sea; in average, they were 0.09 μg m^{-3} (Pol'kin et al. 2008). The lowest values (0.004–0.005 μg m^{-3}) were measured in the atmosphere of the Kara Sea. These estimates are based on measurements in the northern and central parts of the Kara Sea and in the Ob Estuary.

The role of different natural and anthropogenic sources for atmospheric black carbon and its pathways into and within the Arctic (especially in the Russian Arctic) are still poorly studied but during the IPY 2007/2008, scientific focus was laid upon this priority research area (Quinn et al. 2011).

Particulate Matter in the Snow, Ice, and Under-Ice Water Near the North Pole

Concentrations of particulate matter in snow in the vicinity of North Pole in April 2008 varied from 0.21 to 0.37 mg/l (0.29 mg/l), that is at the background level for the Central Arctic (Shevchenko 2006) and close to values registered near the North Pole (Shevchenko et al. 2004, 2010). According to synchronous studies at 4 sites in the vicinity of North Pole, concentrations of particulate matter in ice varied from 0.12 to 0.36 mg/l (0.23 mg/l in average). Such low values of PM concentration are typical for ice conditions when ice algae are not active due to low amount of sunlight (Melnikov 1997). These studies also confirmed that multiyear sea ice has been formed in Central Arctic waters characterized with low levels of suspended

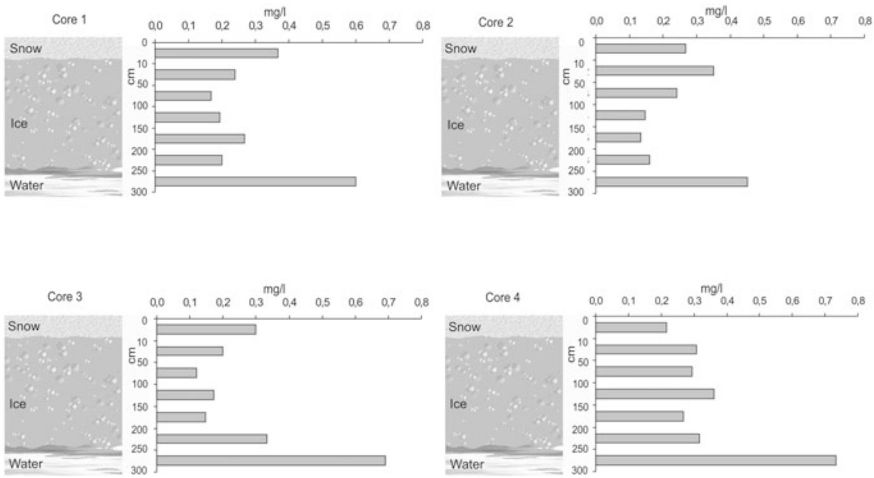

Fig. 5.3 Concentration of particulate matter in snow, ice, and under-ice water near the North Pole at the area from N 88°19.89′ to N 88°30.28′ and from E 014°16.95′ to E 017°22.42′ in April 2008

particulate matter (PM). The ice formed in the shelf seas has higher PM concentrations because the particulate matter is incorporated there in the new-forming ice as a result of numerous interacting formation processes (Nürnberg et al. 1994; Eicken et al. 1997; Lisitzin 2002, 2010; Dethleff and Kuhlmann 2010).

For vertical distribution of PM in ice cores, concentrations are non-significantly higher in the upper 10-cm surface layer (usually ice of snow origin) and at the interface in contact with surface seawater (Fig. 5.3).

Concentrations of SPM in under-ice water were higher than in ice and snow; they varied from 0.45 to 0.73 mg/l (0.62 mg/l in average). In this layer, at the end of winter more active development of phyto- and zooplankton begins (Melnikov 1997) during the annual spring bloom events in the Marginal Ice Zones (MIZ) of the Arctic.

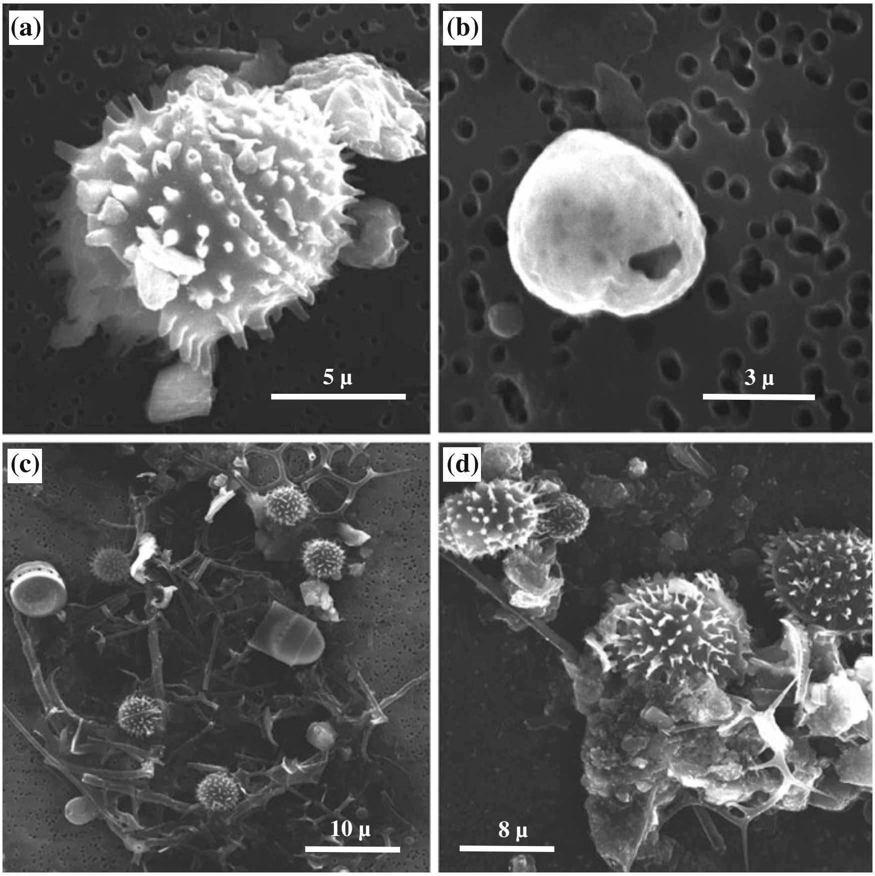

Fig. 5.4 Scanning microscope photographs of typical particles from snow **a**, **b** and ice **c**, **d** in the vicinity of the North Pole: **a** diatom spore and mineral grains; **b** ash particles; **c** diatom spores and valves, black carbon aggregates, silicoflagelate, mineral grains; **d** spores, black carbon aggregates, silicoflagelate, and mineral grains

Results of scanning electron microscopy (Fig. 5.4) show that content of biogenic particles in particulate matter varies from 45 to 80 %. Here, mainly spores of diatoms, diatoms, and plant fibers are present. Mineral and anthropogenic (fly ash and sooty aggregates) are less abundant in snow. Aerosol studies carried out at the same time at Zeppelin Mountain Atmospheric Research Station at Svalbard (Weinbruch et al. 2012) demonstrated that concentration of ash particles was relatively low.

In the upper sections of ice cores, mineral particles (up to 30 %) were the major constituents as a result of delivery from snow and accumulation in the upper layers of drifting ice (Nürnberg et al. 1994; Pfirman et al. 1995). Biogenic particles in middle and bottom sections are presented by diatoms; they are abundant in the middle parts of ice cores where they make up to 40 % of particles. In the middle parts of ice cores, mainly spores and remnants of protozoa are presented.

In suspended particulate matter from under-ice water, biogenic particles were the major constituents (up to 70 %).

In April 2008, near the North Pole average content of Si in particulate matter from snow was 3.9 %, from ice 5.3 %, and from under-ice water 1.8 %; average content of Al in particulate matter from snow was 1.7 %, from ice 1.8 %, and in suspended particulate matter from the under-ice water it was 1.2 %. Using Al as indicator of lithogenic material and taking its content in the upper part of continental crust as 8.15 (Rudnick and Gao 2003), we estimated content of lithogenic component of particulate matter in near North Pole area as 20 % for PM from snow, 25 % for PM from ice, that corresponds to the results of scanning electron microscopy. It corresponds to the data on C_{org} (for snow to $C_{org} + C_{BC}$).

Anthropogenic Load on the Russian Arctic Environment Due to Anthropogenic Atmospheric Emissions

Regarding the results from the backward approach, we revealed that in the 2000s (similar as shown for the 1980s) marine air masses were characteristic for the atmosphere above the northern Eurasian islands (Vinogradova 2011; Vinogradova and Ponomareva 2012). Therefore, the surface air composition resembled the marine atmosphere. This feature was confirmed in particular for the Central and Pacific sectors of the Russian Arctic. This effect is more important in summer and early fall when surrounding land and ocean surfaces are free from snow and ice, and continental air strongly differs (in its composition) from marine one. Such shifts possibly should initiate changes in compositions of soils, ocean, river, lake waters, etc., influencing Arctic flora and fauna.

During the past decades, Arctic atmospheric pollution variations (through atmosphere) were mainly due to changes of atmospheric emission from different anthropogenic source regions in combination with strong secondary diffusive sources (AMAP 2009, reference required, check report on HM and SLCF) in particular for HM and BC. The estimated average levels (from 2000 to recent) of air and surface indices of anthropogenic impacts of HM and BC at FJL and SZ are

Table 5.2 Annual average atmospheric levels and fluxes of selected heavy metals at FJL and SZ (Vinogradova and Ponomareva 2012)

Pollutant	Annually averaged air concentration, ng/m^3		Annual fluxes onto the surface, μg/m^2/year (g/km^2/year)	
	FJL	SZ	FJL	SZ
Pb	0.081	0.061	6.8	4.3
Cd	0.0043	0.0041	0.35	0.27
As	0.011	0.019	0.88	1.2
Zn	0.26	0.22	21	15
Ni	0.11	0.13	9.9	9.5
Cr	0.030	0.063	2.4	3.9
Cu	0.16	0.20	14	15
BC	0.53	1.2	39	82

presented in Table 2. Site WI is under prevailing influence from the sources situated at Alaska and in Canada with so far unknown (unpublished) characterized emission sources (Vinogradova 2011).

In general, atmospheric circulation over the Northern Eurasia has significant spatial and seasonal variability. On an annual scale, the long-term differences in atmospheric circulation are shown to result in remobilization on anthropogenic pollution (in particular for HM and BC) and remobilization which even can compensate for documented emission reduction (Vinogradova et al. 2008a, b). According to our study, the atmospheric pollution in our target locations stems mainly from two potential primary emission sources, namely the Kola Peninsula and Norilsk region. These atmospheric pollution contributions to the Russian Arctic, however, decreased during the past three decades by 45–60 % at the different study sites (Fig. 5.5). Combined anthropogenic loading from these two sources shows considerable decreasing trends from the west to the east of the Russian Arctic being reduced by a factor of 10 around the area of the Laptev Sea with values comparable with the White Sea area.

Annual atmospheric HM fluxes originating from the two major anthropogenic sources identified (Kola and Norilsk) and deposited into the central waters of the White, the Barents, the Kara, and the Laptev seas are obviously contributing with comparable amounts to the overall pollutant load as shown for the inflowing river waters (Gordeev 2002) from the coastal zones (Fig. 5.6). Thus, atmospheric transport of heavy metals is an important contamination source not to be neglected in the validation of modeled and comparisons of empirically derived data both for source elucidation in the atmosphere and for the surface ecosystems in the Arctic (Vinogradova et al. 2008c).

Seasonal variations in anthropogenic pollution are mainly governed by deposition processes (Vinogradova and Ponomareva 2012): The maximal HM contents in air are reported for the cold Arctic seasons, whereas the maximal HM depositions onto the surface occur during spring and summer. Our estimates are in reasonable

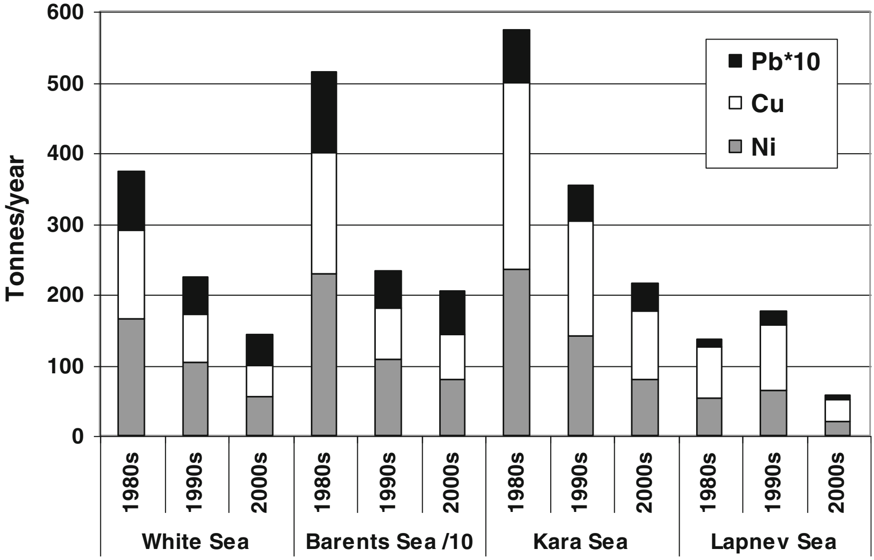

Fig. 5.5 Long-term variations (through three decades) in total annual fluxes of anthropogenic Pb, Cu, and Ni to the surfaces of Russian Arctic seas (special scales are for the Barents Sea and for lead fluxes)

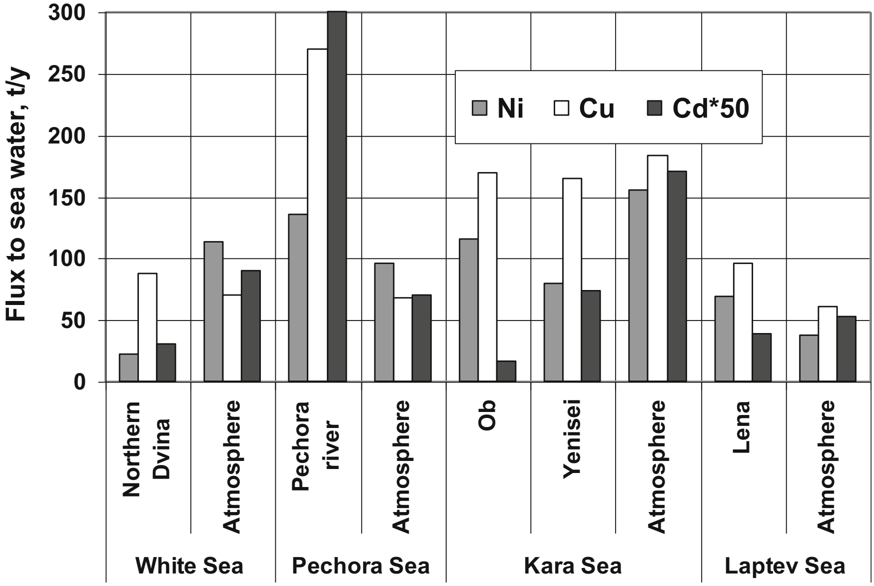

Fig. 5.6 Atmospheric and rivers' fluxes of HMs to Russian Arctic seas (special scale is for Cd)

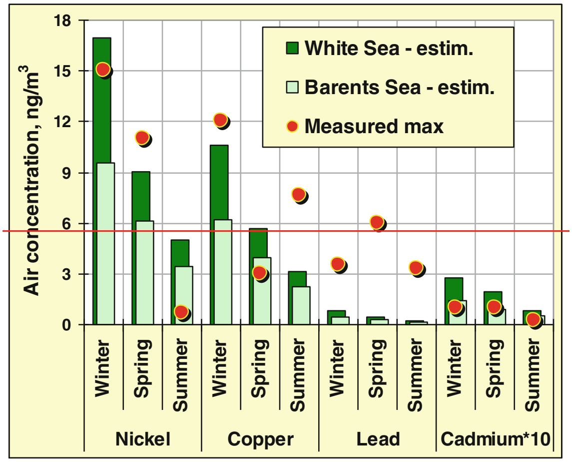

Fig. 5.7 Comparison of computed Ni, Cu, Pb, and Cd air concentrations with measurements (AMAP 2004) in the regions of the White and the Barents seas (special scale is for Cd)

accordance with previously reported experimental data (Fig. 5.7). At high latitudes, higher fluxes onto the surface may be observed during spring and fall as a result of higher precipitation frequency (mainly in the liquid form) in combination with still (or already) considerable concentrations of heavy metals in the air. For the forecasts, it is important that the cleaner air at the place will not guarantee the safer surface environment there (Baklanov et al. 2013).

Although HM concentrations in air, water, and soil within the Arctic region do not exceed maximum permissible concentrations (MPC), the detected Pb and Cd amounts in muscles, livers, and kidneys of Arctic reindeers, predatory animals, and birds often exceed their MPC as a result of HM deposition in natural food chains (AMAP 1998, 2015; Soinienen et al. 2005; Van Oostdam et al. 2005).

Ice is an ideal medium for aerosol accumulation (for subsequent dry and wet precipitation). All the major drift systems merge together in the Fram Strait (between Greenland and the Svalbard archipelago) at the junction between the warm North-Atlantic waters and cold Arctic currents. This area is considered the major (Greenland) cryodepocenter of the Arctic where most ice-rafted sediment discharge takes place (Lisitzin 2010).

Conclusion

The atmospheric pollution (HM and BC) contribution from two main primary source regions (Kola Peninsula and Norilsk region) to the Russian Arctic environment decreased considerably during these past three decades by 45–60 %. However, the decreasing trends were site specific (numbers required). The subsequent investigation of the combined anthropogenic loading from these two source locations confirmed again spatial level differences with decreasing ice/snow concentrations from the west to the east of the Russian Arctic being lower by a factor of 10 in the area of the Laptev Sea compared to snow/ice levels in the White Sea area.

The estimated atmospheric HM contribution from the identified two major anthropogenic sources (Kola and Norilsk) to the White, the Barents, the Kara and the Laptev seas is considered comparable with the river inflows in the respective coastal regions.

A large amount of ice-rafted sedimentary material (with pollutant originated from atmospheric deposition) is transported continuously to marginal regions. During the annual ice melting in these Marginal Ice Zones (MIZ) located in the Fram Strait and the Central and Northern Barents Sea, sedimentary materials (including transported pollution) are directly released into the marine ecosystems with obvious consequences for environment and ecosystems.

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Chapter 6
Schools Taking Part in a Research Project Investigating Dioxins in Fish

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Abstract The world is facing large environmental threats such as climate changes and spread of contaminants. Environmental awareness has become a part of our daily lives. In order to engage youth in environmental issues and ignite their interest for natural sciences, a scientifically based project was run where pupils participated in environmental research on dioxins in fish. Dioxins and dioxin-like PCBs were screened by the use of *BDS DR CALUX*® bioassay in 203 fish samples from 13 countries: Australia, Croatia, the Czech Republic, Denmark, Estonia, Finland, France, Iceland, Latvia, Liechtenstein, Norway, Poland and Sweden. The project was part of the Norwegian education and outreach initiative of the International Polar Year (IPY), and schools were invited to participate. Pupils from 54 schools followed a scientific protocol for fish sampling; they recorded and published important field and fish data through a Web interface and labelled, packed and shipped the samples for CALUX analysis. We conclude that collaboration between schools and research institutions was beneficial for both partners. The results showed that the majority of the fish samples had dioxin levels below the maximum limit set by the EU commission. These results would be difficult to obtain without the effort of the involved schools.

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Introduction

The world is facing major challenges with climatic changes and spread of environmental contaminants. The combined threats of climate changes and environmental pollution are affecting vulnerable ecosystems in the polar regions and might also pose a threat to human consumers by tainting important food resources. Most pollutants can be transported with winds and ocean currents to remote areas such as the Arctic and Antarctic. Pesticides and industrial chemicals contribute to the high levels of pollutants that are found in polar bears and other species in the Arctic as well as in wildlife elsewhere. Persistent organic pollutants (POPs) readily enter food chains, especially in the marine environment, and accumulate in fat deposits, a process known as bioaccumulation (Borgå et al. 2005). They thus become more concentrated in predatory organisms that are found higher in the food chain. POPs can even be traced in common grocery goods, and consumers can unintentionally be exposed for instance by consuming farmed fish (Hites et al. 2004). Dioxins are a group of POPs that are by-products of industrial processes. They are considered to be the most toxic man-made contaminants known today, and they may have major impacts on human health. The permitted levels of dioxins in food are strictly regulated in most countries, and a so-called toxic equivalent factor (TEF) system is established, which enables authorities to set guidelines for intake levels that do not exceed harmful levels (Van den Berg et al. 1998). Fish are especially prone to accumulate dioxins, and most fish on the commercial market, both farmed and wild caught, are monitored for their dioxin levels. However, almost nothing is known about dioxin levels in locally caught fish from recreational fishing, which is often brought home for consumption. This can be fish caught in nearby lakes or streams, or fish with marine origin that are caught close to industrial areas or harbours that are not regularly monitored for their dioxin levels.

In the recent history, there has been an increasing awareness of education for sustainable development, and the United Nations appointed the decade 2005–2014 to be the Decade for Education for Sustainable Development. This has increased the focus on education and outreach in national and international scientific researches, making it easier to get funding for projects and activities that incorporate education for sustainable development. Education and outreach were integral to the International Polar Year (IPY) 2007–2008, and teachers and pupils were here given the opportunity to take part in scientific activities in various scientific disciplines.[1,2]

It is not very common to involve pupils in scientific projects. There may be several reasons for this reluctance; researchers may be concerned that this will

[1]http://www.iasc.info/files/Education%20&%20Outreach/ICSU_Report_Digital.pdf.

[2]http://ipy.arcticportal.org/hidden/item/2297-promotional-material-for-the-polar-resource-book.

detract the focus from their own scientific careers and publications, and they may be worried that they will not be taken seriously by their peers within their own scientific community. There is also reason to believe that many scientists hesitate to rely on the pupils' capability to do trustworthy scientific work.

In order to deal with the current environmental challenges, it is imperative to increase knowledge and consciousness about environmental topics among people. One important challenge is to inspire youngsters to take an interest in environmental and natural sciences. Practical involvement, excursions, laboratory work experiences and reflective thinking might be central to achieving this. Involvement of young people in authentic research projects where they are working as scientists may be a first important step to stimulate the motivation of pupils to learn, and coupling such activities with sound reflections on the subject matter has been found to promote learning (Schwartz et al. 2004; Knain and Kolstø 2011).

Research Design and Methodology

In the project Global POP, pupils followed a scientific protocol for preparing tissue samples of fish caught in their own geographical region. The project was divided into two parts: practical hands-on activities and a more theoretical Web-based activity. Invitation of schools on a global scale was facilitated by Norwegian Centre for Science Education and the GLOBE Network (http://globe.gov).

Most of the communication between the researchers and the schools was done via e-mail and supported through the website (http://sustain.no/projects/globalpop). NILU was responsible for the content of the Web together with the Centre of Schools' Science Education. Written guidelines for fish sampling and a video showing sample preparation were provided on the website along with the descriptions of the analysis of dioxins in fish and guidelines to how to write a report of the results. The practical part of the project consisted of fishing and logging of field data. Schools then processed the fish samples adhering to scientific standards and uploaded the data on their samples to the website (GPS coordinates, description of field sampling site, species, size, sex, sampling method, maturation stage, etc.). Then, the pupils packed and marked the samples to scientific standards and shipped the fish to the research institute NILU for analysis, i.e. the use of a high-throughput screening method for dioxins (Murk et al. 1996).

When the results had been analysed, NILU uploaded the results to the website using a form that coupled the analysed samples to the information provided by the schools. On the website, structures for viewing results were programmed that allowed participants to compare dioxin levels by species and nationality. There were also map views of the participant schools and listings of all data entered by the schools. A calculator was also added to the website allowing schools to enter portion size, number of meals per week and body mass to see how the levels of dioxins in the fish they had caught compared to maximum intake levels set by the European Commission. The participants were encouraged to upload photographs of their project work available for all to view on the website. There was also a section where the participants could upload movies, presentations and project reports. From this, pupils gained insight into contaminants in fish, hands-on scientific field sampling, logging of data, fish biology and digital competence through the Web-based reporting.

This website also provided useful links to relevant scientific themes, presentations given by the researchers involved in the project, references to dioxin analysis and maximum levels of dioxins given by the European Commission.

Workshops for teachers were arranged at Svalbard (Longyearbyen) in Norway, Warsaw in Poland by the GLOBE Network and Riga in Latvia. At these workshops, scientist gave presentations of the various parts in the project and demonstrated fish sampling and trained the participant teachers in fish dissection.

Researchers must be able to come up with their own questions, to employ appropriate methods to provide answers to the questions and to evaluate the answers. By giving pupils the chance to take part in the scientific process, they acquire competence on scientific inquiry. Evaluations of similar projects have shown that inviting pupils to take part in research activities is a good way to inspire and engage them to take on further tasks within the area of natural sciences and environmental care issues. Taking part in an important research project and reaching the realization that the society actually finds use for their work, tends to instil a feeling of ownership, pride and responsibility in pupils (Andresen et al. 2015, and references therein) (Fig. 6.1).

Fig. 6.1 Images of pupils at work with research activities

As far as we know, this project is one of very few projects where schools have actively participated in a research project. Our experience with involving pupils in environmental research shows that students are very attentive and meticulous and that they follow scientific instructions very carefully (Heimstad et al. 2003; Heimstad and Herzke 2004). The pupils' behaviour is, of course, also strongly dependent on how dedicated and engaged their teachers are. There are studies indicating that pupils get inspired by taking part in real research projects where the outcome of the project depends on the quality of their contribution and where their data are used for scientific work and in publications by researchers (Nali and Lorenzini 2007; Creilson et al. 2008). Researchers in an educational project on biological monitoring of air quality concluded that participation in the project generated a widespread curiosity, involvement and a sense of responsibility among the pupils. The pupils also developed a long-lasting interest in environmental sciences (Nali and Lorenzini 2007).

The Web platform that hosted the Global POP website, http://sustain.no, is the international interface of the Norwegian learning platform miljolare.no, and it is a tool for education for sustainable development funded by the Norwegian government. This was the education sector's response to the report Our Common Future (WCED 1987) and local Agenda 21. The miljolare.no website provides methodology and guides to help schools to investigate their local environment and to publish their findings on the miljolare.no website. It aims to be a meeting place between schools, scientists, management authorities, NGOs and decision-makers, and it strives to facilitate dialogue, collaboration and partnerships between schools and other stakeholders in the society. This approach has been found to be fruitful (Andresen et al. 2015 and references therein).

Motivation

The IPY initiated by the Norwegian Research Council has merited research with the focus on the climate changes in the polar regions. IPY also challenged the scientific environment in Norway to include different educational and outreach aspects in their research. However, few projects were launched where school students were actually involved in the scientific work of the research projects. In this Global POP project, researchers in environmental chemistry benefited from schools contributing to fish samples, data logging and their local knowledge about the region and habits for recreational fishing for food consumption. Aside from the direct findings of the project, schools benefited from getting first-hand knowledge of how scientific research projects are conducted. The help from diligent and engaged pupils in the analysis of fish from various parts of the world provides the scientific community with a unique data set valuable to national and international food authorities, pollutant conventions and regulations. This data set enabled us to investigate trends and patterns in dioxin levels within and across fish species as well as across geographical regions. At the time when the project was finalized in 2010, no CALUX data had been published from Norway and only a very few international publications on this existed.

Challenges and Possibilities

A major challenge in such a collaboration project is to get schools aware of the project and to engage their interest. In Global POP, we took the advantage of an already existing website, http://sustain.no, which was, and still is, maintained and developed by the Norwegian Centre for Science Education. This website is a resource for promoting training in sustainable development at all levels, from elementary to secondary school. Further, we invited schools to participate through the GLOBE network (http://www.globe.gov), which is a worldwide hands-on, primary and secondary school-based science and education programme with 110 participating countries. Through the GLOBE programme and selected educational conferences and workshops, we have also invited teachers to participate in demonstrations, both nationally and internationally.

Our experience is that it can be logistically challenging for schools to allocate time to such projects within the constraints of the mandatory curriculum plan. The involvement of the schools is also very dependent on the teachers' engagement. We think the best solution is to have a flexible sampling period so that schools may have some flexibility when planning the fieldwork and sampling.

Some schools produced reports where they evaluated the scientific results and the outcome of the projects, and these reports can be found on the website.[3] Ideally, we would have liked to carry out a more comprehensive evaluation of the learning outcomes of the project, but this was beyond the resource scope of the project. However, evaluations of similarly structured Web-based school projects exist, and they conclude that projects where pupils collect their own data provide a great starting point for problem-based learning and education for sustainable development as outlined in the UN Decade for Sustainable development (Andresen et al. 2015 and references therein).

The sustain.no website is in English, and pupils in secondary schools and high schools were therefore the natural target group. The majority of communication was done via e-mail and the website. Because electronic communication alone may not be ideal for establishing a close interaction between schools and researchers, or among participating schools, we also hosted several workshops where participants could get together and discuss the project and other issues. This was very important for network building and for facilitating collaboration between schools. From previous experiences, we knew that such interactions are essential for the success of such projects, and when planning such projects, we would strongly recommend

[3]http://sustain.no/projects/globalpop/uploads.php.

yearly workshops where pupils, teachers and scientists meet. Such workshops can also be combined with other ongoing hands-on projects, alternative scientific themes and environmental threats such as climate changes etc.

Results

In total, 54 schools from 13 countries participated in the project during the years 2007–2010. The majority of the schools came from Norway (36 schools), followed by Estonia (4 schools), France (3 schools) and 1 or 2 schools from Australia, Croatia, the Czech Republic, Denmark, Finland, Iceland, Latvia, Liechtenstein, Poland and Sweden.

Field and fish data from many of the participating schools were published on the website.[4] Within the finalization of the project in 2010, 41 schools had reported data on the website. Schools documented their participation and sampling by photographs (138 images were uploaded) and videos and uploaded project reports on climate change issues and contaminants. Two hundred and three fish samples were received by NILU and analysed for dioxins and dioxin-like compounds by the use of an in vitro bioassay (*BDS DR CALUX*®; BioDetection System, the Netherlands) for Ah receptor active compounds, such as dioxins and coplanar PCBs (Murk et al. 1996). The method is a rapid, sensitive and cost-effective tool for screening food and feed ingredients for dioxin and dioxin-like compounds.

In total, 29 fish species were sampled, among which some species had not previously been analysed for their dioxin level (Table 6.1). The majority of the fish samples came from Norway with a total of 139 samples followed by France (18), Estonia (13), Denmark (6) and the Czech Republic (5). The number of samples from the other countries varied between 3 and 1. The fish species with the highest number of samples was Atlantic cod (*Gadus morhua*), of which all samples came from Norway. Brown trout and pollock were the second and third most abundant fish species in the submitted samples, and most of these also came from Norway. Three samples of brown trout came from Liechtenstein.

Results revealed that almost all the fish samples had concentrations of dioxins that were below safety limits set by WHO for dioxins as a single group (4 pg TEQ/g ww) and dioxins and dioxin-like PCBs (8 pg TEQ/g ww).

Two of the common dab samples from Norway had relatively high levels of contaminants, and one of these had a concentration close to the maximum level set by EU for dioxin and dioxin-like PCBs in fish (8 pg TEQ/ww). Congener-specific analysis of the two samples revealed that the concentration of PCB126 dominated and contributed to over 90 % of the measured TEQ concentration. In general, the majority of the samples from the participating countries had low concentrations of

[4]http://sustain.no/projects/globalpop.

Table 6.1 Sampled fish species sorted by the number of samples (No.), with average pg TEQ/g ww ($\bar{x}$) and standard deviations (St. dev)

Fish species	Latin name	No.	($\bar{x}$)	St. dev
Atlantic cod	*Gadus morhua*	49	0.26	0.16
Brown trout	*Salmo trutta*	31	0.35	0.18
Pollock	*Pollachius pollachius* *Pollachius virens*	26	0.21	0.16
European perch	*Perca fluviatilis*	15	0.38	0.57
Arctic char	*Salvelinus alpinus*	10	0.49	0.29
Flounder	*Platichtys flesus*	6	0.48	0.33
Common whitefish	*Coregonus lavaretus*	6	1.25	0.78
Pike	*Esox lucius*	6	0.45	0.29
Atlantic mackerel	*Scomber scombrus*	5	0.86	0.55
Common carp	*Cyprinus carpio*	5	0.28	0.13
Haddock	*Melanogrammus aeglefinus*	5	0.38	0.42
Fat chub	*Leuciscus cephalus*	4	0.61	0.56
Mullet	*Mugil cephalus*	4	0.43	0.15
Herring	*Clupea harengus*	4	1.94	1.10
Burbot	*Lota lota*	3	0.23	0.06
Common dab	*Limanda limanda*	3	3.94	3.44
Yellowfin bream	*Acanthopagrus australis*	3	0.52	0.16
Narrowhead grey mullet	*Mugil capurrii*	3	1.00	0.35
Greater weever	*Trachinus draco*	3	0.17	0.03
Gilthead bream	*Sparus aurata*	3	0.78	0.54
Rose fish	*Sebastes marinus*	2	0.88	0.17
Common roach	*Rutilus rutilus*	2	0.20	0.13
European plaice	*Pleuronectes platessa*	1	0.14	–
Common ling	*Molva molva*	1	0.54	–
Silver bream	*Blicca bjoerkna*	1	1.3	–
European plaice	*Pleuronectes platessa*	1	0.14	–
Eel	*Anguilla anguilla*	1	0.22	–

dioxins and dioxin-like compounds as revealed by the use of BDS DR CALUX® bioassay (Fig. 6.2).

When separating the concentration data in intervals of 0.1 units below 0.7 pg TEQ/g ww and with larger intervals above 0.7, the highest number of samples revealed low levels between 0.1 and 0.2 pg TEQ/g ww. Only 18 samples had concentrations above 1 TEQ pg/g ww (Fig. 6.3).

Compared to other studies (Baeyens et al. 2007; van Leeuwen et al. 2007; Schoeters et al. 2004) using the CALUX bioassay method, the levels of dioxins for the most dominating and comparable fish species are lower in the present study. Baeyens et al. (2007) measured the dioxin levels in fish on the Belgian market (2004–2006). The average value for cod and brown trout was 0.85 and 1.04 pg

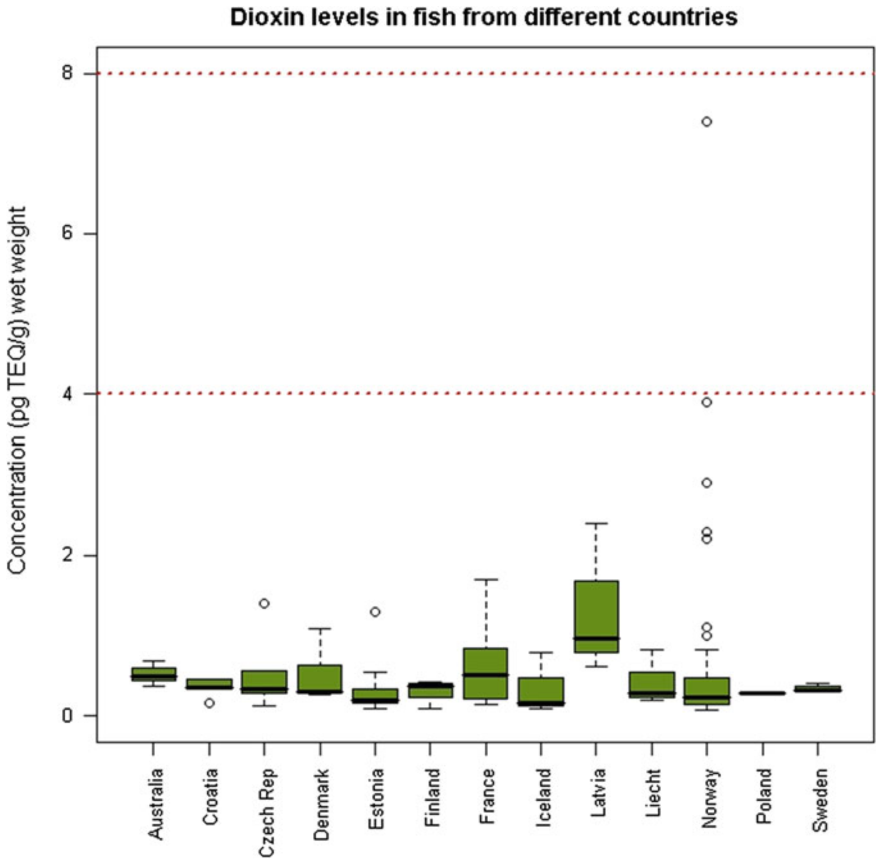

Fig. 6.2 Dioxin levels (pg TEQ/g wet weight) in fish from the various countries. The *black horizontal line* shows the median values for the analysed fish species by country. Bottom 25th percentile, top 75th percentile and ranges are also indicated. The *red dotted lines* are the maximum levels set for dioxins (4 pg TEQ/g ww) and for dioxin and dioxin-like PCBs (8 pg TEQ/g ww) in fish muscle

CALUX-TEQ/g wet weight, respectively, compared to 0.26 and 0.35 pg CALUX-TEQ/g wet weight, respectively, in the Global POP study. In a study by Schoeters et al. (2004), it was reported an average concentration of 2.53 pg CALUX-TEQ/g wet weight in herring, whereas in our study we found a concentration of 1.9 pg CALUX-TEQ/g wet weight.

Atlantic cod and species from the Gadidae family (cod, pollock and haddock) were the dominating fish species in the project. After log-transforming lipid weight concentrations to normalize the sampling distribution, we compared the toxin levels in northern and southern Atlantic cod. The results revealed a significantly higher concentration in the southern samples than in the northern samples (Fig. 6.4, left figure). Larger fish commonly represents older fish which again often have higher

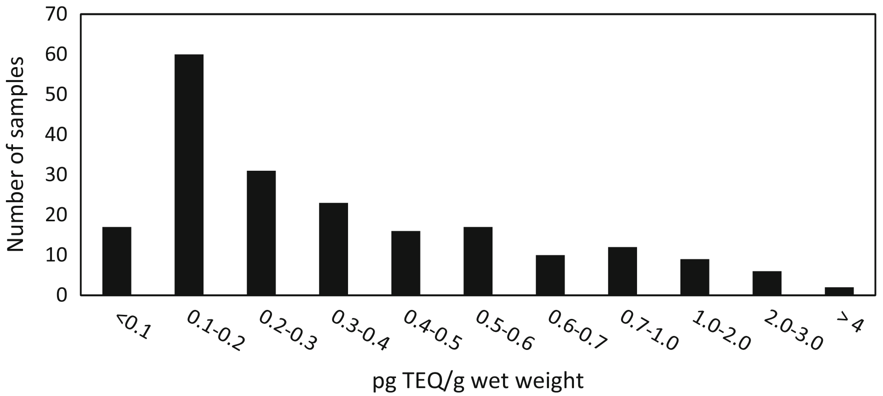

Fig. 6.3 Number of samples by concentration intervals from below 0.1 pg TEQ/g ww to above 4 pg TEQ/g ww

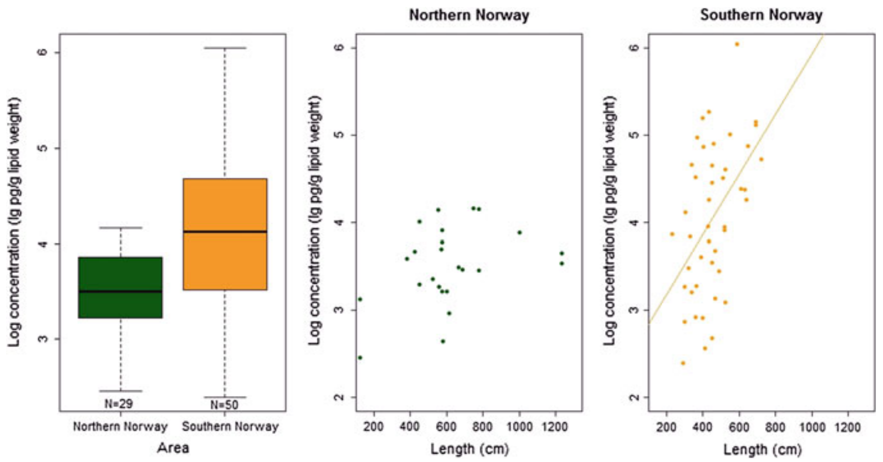

Fig. 6.4 Differences in TEQ concentrations between Atlantic cod from north and south Norway were evaluated using ANOVA (analysis of variance). Log-transformed lipid weight (lw) levels were used in the analysis (figure to the *left*). There was a significant difference ($p = 0.005$) in TEQ concentrations between the north and south with the south having higher concentrations than the north. The *black horizontal line* shows the median, bottom 25th percentile and top 75th percentile of the data based on all analysed fish species in each country. Cods from south Norway reveal statistically significant correlation of fish length with dioxin levels ($R^2 = 0.23$, $p < 0.001$)

loads of contaminants due to bioaccumulation with time. This was, however, not the case in our data set, as the northern fish in general were larger than the fish from the south. From the site-specific data published from schools, the fish sampling site from south Norway was fjord areas closer to cities and more densely populated areas than the sampling sites in the north. This may explain the higher dioxin concentrations of the fish from south Norway compared to north Norway. Fish from north Norway were fished in regions near Nordkapp, Sjøvegan, Vannareid, Bodø and Ørsnes. Fish from the southern part of the country were fished in areas near Steinkjer, Molde, Sunnylven, Førde (Sande), Risør, Stavanger, Klepp, Karmøy, Egersund and Oslo. Data from some of these sites are available on the results' site of the website.[5]

Conclusion

In order to meet the environmental challenges we are faced with, it is imperative to increase the knowledge and consciousness about environmental topics among people. From a research point of view, we are very happy to be able to present this large and important data set to national and international scientific communities and authorities. It would have been difficult to acquire this data set without the efforts and skills of the involved schools, and we encourage the scientific communities to consider a more extensive collaboration with schools in order to establish similar joint ventures. Results revealed that the majority of the fish samples had concentrations of dioxins that were lower than the safety limits set by European Commission.

Although we did not carry out a comprehensive evaluation of how the schools perceived the project, we have received positive feedback from a number of schools conferring that being part of a real research project engaged the pupils. We consider this project a good example of how pupils can be engaged and learn from authentic experiences with scientific inquiry, and we do hope that more collaborative projects between schools and scientists will emerge in the future, as this most likely would benefit both researchers and pupils.

Acknowledgments We would like to express our gratitude to all the enthusiastic teachers who have arranged fishing field trips and inspired their pupils to do scientific work, to pose their own questions and to formulate their own answers and conclusions. Finally, we will say to the pupils: "We are impressed over your enthusiasm and skills as real scientific researchers, and hope to work with you in the future!" We are grateful for the financial support from the Research Council of Norway, project 18218: "A global network of schools investigating environmental pollutants in fish from the Arctic and worldwide". We are very grateful for the help by Therese H. Nøst with the graphics. Thanks also to an anonymous referee for constructive input.

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[5]sustain.no/projects/globalpop/results/.

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Chapter 7
Arctic Monitoring and Assessment Programme (AMAP)—IPY Meeting in Iqaluit, Nunavut, Canada (June 2009), and AMAP Human Health Assessment 2009

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Executive Summary

The Arctic Monitoring and Assessment Programme (AMAP) Human Health Assessment Group (HHAG) undertook an assessment and meeting to summarize arctic contaminant and human health research undertaken during the International Polar Years (IPYs). This meeting took place in Iqaluit, Nunavut, Canada, on June

In Memoriam for Jens C. Hansen Jens C. Hansen passed away in 2012 only a few years after his retirement from Aarhus University, in Denmark. He devoted his academic life to research, which focused on understanding the exposure pathways for and the effects of contaminants on Arctic residents. Jens was a founding member and chair of the Human Health Assessment Group of the Arctic Monitoring and Assessment Programme (AMAP). He provided leadership and encouragement on all aspects of the work of the group and AMAP for over 17 years. He also created and became the first head of the Centre for Arctic Environmental Medicine at Aarhus University (now Centre of Arctic Health). His kindness and sensitivity toward all he met, worked with, and trained were inspirational. We very fondly dedicate this work to his memory: Jens C. Hansen, an excellent scientist, mentor, and friend to many.

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10–12, 2009. This report summarizes the much more voluminous document *AMAP Assessment 2009*: *Human Health in the Arctic* (AMAP 2009a) that was released and discussed at the Iqaluit meeting.

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Population Health and Effects of Contaminants

In light of current studies, many indigenous populations in the Arctic region have poorer health than national averages. While socioeconomic conditions and lifestyle choices are major determinants of health, contaminants may also have a contributing effect. Toxicological studies show that contaminants, at the levels found in some parts of the Arctic, have the potential for adverse health effects in people. Epidemiological studies, looking at Arctic residents directly, provide evidence for subtle immunological, cardiovascular, and reproductive effects due to contaminants in some Arctic populations. These results indicate that POPs, mercury, and lead can affect health of people and especially children at lower levels of exposure than previously thought. Genetic characteristics of the various Arctic populations also affect their response to contaminants and susceptibility to certain diseases.

A major dietary shift from traditional to store-bought food is underway in most of the Arctic, with important health implications. In addition to environmental concentrations of the contaminants in traditional foods, lifestyle factors and social and cultural practices play a large role in determining human exposure to contaminants in Arctic areas. Despite changes in lifestyle and diet that are resulting in increasing consumption of store-bought foods, traditional foods remain important to Arctic indigenous peoples for social, cultural, nutritional, economic, and spiritual reasons. Store-bought foods are increasingly the main source of dietary energy, but traditional foods provide many nutrients and are still a major contributor to healthy diets in many communities. Some traditional foods can also carry potential risks from contaminants. The combination of high prices for store-bought foods and the work, risks, and costs associated with obtaining traditional foods has made food security a large concern for many Arctic residents.

Recent studies have found a number of mechanisms by which contaminants can affect metabolism. Obesity is associated with an increased risk of cardiovascular disease and of developing diabetes; as in other parts of the world, obesity is increasing in Arctic communities. POPs, even at low concentrations, also increase the risk of diabetes. These new findings emphasize the need to consider the interactions between contaminants and other health conditions.

Trends in Exposure and Contaminant Levels

Human exposure to most legacy POPs and mercury is decreasing in several Arctic populations. This reflects changes in diet, changing levels of environmental contamination, and health advice to critical groups in some areas concerning consumption of certain foods; however, exposure remains high in some populations. The proportion of women of childbearing age who exceed blood-level guidelines for PCBs, mercury, and lead is decreasing. For PCBs and lead, in particular, there is evidence that this reflects the declines in environmental levels of these contaminants.

Marine mammals remain a major dietary source of POPs and mercury, so that people who eat large quantities of marine mammals have higher POPs and mercury levels than those who do not.

Emerging compounds such as brominated flame retardants and fluorinated compounds are a concern for three reasons: They are present in Arctic people and biota, levels globally have increased over the last 15 years, and their toxic effects have not been studied in detail. There is little information on the routes of exposure or trends of these contaminants in Arctic populations.

Reliable interpretation of information on trends and inter-regional differences is critically dependent on an ability to compare data from different studies and different laboratories. Laboratory performance-testing procedures initiated by AMAP and others, including the AMAP interlaboratory comparison program for analysis of contaminants in human tissue, have markedly improved analytical cooperation, data comparability, data reliability, and data accuracy in studies using the participating laboratories and have led to more reliable data on contaminant levels in human tissues. Further improvements can be achieved through continued efforts in this respect.

Increased industrial activity in parts of the Arctic is likely to lead to an increase in local sources of contaminants. Anticipated changes in global and Arctic climate may also result in changes in contaminant transport to the Arctic. Such changes may affect exposure patterns to some contaminants.

Communication

Communicating the results of studies concerning contaminants and people is important in helping Arctic residents make informed food choices. Health advisories issued in response to findings reported in past AMAP assessments have succeeded in reducing exposure to contaminants in some Arctic population groups.

Risk communication must be carried out with great care and respect for culture at a community level. The involvement of community members and organizations, regional health officials, and indigenous organizations is the key to developing and disseminating messages that are appropriate and relevant.

Introduction

The Arctic Monitoring and Assessment Programme (AMAP) Human Health Assessment Group (HHAG) in 2005 envisioned undertaking an assessment and meeting to summarize arctic contaminant and human health research undertaken during the International Polar Years (IPYs). The proposed meeting was accepted by the IPY steering committee as an “educational event.” This meeting took place in Iqaluit, Nunavut, Canada, on June 10–12, 2009. This report summarizes the much more voluminous document *AMAP Assessment 2009*: *Human Health in the Arctic* (AMAP 2009a) that was released and discussed at the Iqaluit meeting. Readers are encouraged to consult AMAP (2009a, b) for a more in-depth evaluation of all the topics summarized here.

Factors Influencing Human Exposure to Environmental Contaminants and Population Vulnerability

While most Arctic contamination by POPs and metals is from long-range transport, there are several potential sources of contaminants from within the Arctic that could add to loadings of metals and POPs. Human population exposure comes primarily from country food consumption, and local sources of contamination have the potential to contribute marginally to food contamination.

Climate change is almost certain to affect the biogeochemical fluxes of contaminants within the environment. These fluxes will cause increases and decreases in current human exposures. Current climate trends and future model predictions illustrate the incomplete understanding of the interactions between contaminants and climate. Not all Arctic regions will be affected to the same extent or in the same way. Climate change is also likely to change exposures to pathogens such as viruses and bacteria, and to insect vectors. Climate change can also make human populations more vulnerable. Climate-related "stress" associated with changes in diet, lack of access to traditional prey species and foods, loss of traditional hunting, changes in community practices, and changes in occupation can affect psychosocial health. The combination of changing exposures and vulnerability has the potential to affect health and well-being significantly.

The only practical approach for evaluating the implications of climate change on contaminant fluxes is continued monitoring of humans and key wildlife species to directly assess tissue levels of contaminants. Knowledge of personal contamination levels and awareness of contaminant levels in traditional food species will enable Arctic residents to select, where necessary, alternative and culturally appropriate foods which preserve cultural traditions and the benefits of the traditional diet. Research into climate and contaminant interactions should continue to assist policy development of regionally appropriate adaptation strategies.

Knowledge of how climate and local weather affect contaminant distribution and redistribution and the rates at which these occur are in the early stages of development.

Detailed knowledge of climate-mediated transport pathways is also a relatively new field of investigation. Continued research in this area, coordination between researchers, and careful development of joint research plans are critical components for future progress in the *climate–contaminant transport–human exposure* research effort. Findings from coordinated research of this type will have a direct bearing on how exposure and health will affect Arctic residents in the future.

Arctic countries need to continue to support the implementation of international and regional agreements to control chemical and metal releases to the environment. The USA and Russia have yet to ratify some key environmental agreements that are important for the protection of the Arctic and its people.

Monitoring programs to assess the success of national, regional, and global control initiatives currently in place are essential. Without information on where and how levels are changing and which populations are most at risk, it is impossible to revise control actions to make them more effective.

Mineral exploration and extraction could lead to contamination of the local environment with mine tailings. Depending on the amount, type, and location of the contamination, these activities could increase human exposures to some metals of significance through the intake of local foods, air, water, and wildlife. The in-migration of workers to oil, gas, and mining operations can lead to the introduction and spread of disease if health monitoring and treatment are not adequate. General health is a significant determinant in evaluating how an individual may respond to contaminant loads. This assessment has not undertaken a thorough review of mining activity in the circumpolar Arctic to assess how mining activity may co-contribute to current human exposures to mercury, cadmium, and lead.

Oil and gas exploration and extraction activities are likely to increase in the Arctic, but have the potential to contribute only very small amounts of POPs and metals to the environment. Most on-site wastes are contained and managed to similar high standards. While a large oil spill on land could contaminate a large area, it would not add appreciably to the entry of POPs or metals into the environment. A large spill could lead to significant social disruption, which can influence mental health and affect individual responses to current exposures to POPs and metals received primarily through diet A detailed review of the health implications of oil and gas exploration and extraction in the Arctic has been undertaken by AMAP (2009b). It is worth noting that spills from tankers, where oil can spread among floating ice or under the surface covered by ice, are often very difficult to manage (AMAP 2009b). Also, a well rupture under water in the Arctic (like the BP underwater rupture in the Gulf of Mexico) would be difficult to arrest during the ice season and would likely continue for many months until it was safe for underwater crews and submersibles to operate in and around summer ice. The environmental implications for marine life could be enormous and could affect the health of those living in the Arctic who depend on marine species for food and for part of their social systems.

The security and integrity of domestic and chemical waste sites in the Arctic are important, especially under the conditions of a changing climate. Many military sites in the Arctic, which had caused contamination in local areas, have now been cleaned up or are in the process of being cleaned up. These and other waste sites need to be monitored to ensure that they do not affect community water and food supplies as the permafrost thaws. Leaking waste sites have the potential to add to local population exposure to chemicals, metals, and pathogens that are likely to impact negatively on health.

Social factors can affect health outcomes and should be considered carefully in risk assessments related to contaminants. Excessive smoking, alcohol, and drug use, and crowding and underemployment can affect health. How individuals interact within their communities and families can be affected by their knowledge of the extent of contamination of traditional food, breast milk, and the local environment.

Food, Diet, Nutrition, and Contaminants

This chapter presents recent research findings concerning food sources and diet in Arctic populations and explores how food choices and availability influence human nutritional status and exposure to contaminants. Other factors influencing human exposures to contaminants were addressed in Section "Factors Influencing Human Exposure to Environmental Contaminants and Population Vulnerability." The term traditional *food* is used to represent foods that originate in the populations' habitat (generally foods such as seal, whale, caribou, and fish from the land); other foods are called *imported food.*

In the Arctic, traditional food is beneficial to indigenous populations for cultural, social, psychological, and economic well-being; however, foods from mammals, birds, and fish have been documented as the main source of human exposure to contaminants. Changes in food sources and food choice can influence the nutritional quality, density, and security of the diets of indigenous populations. In addition, the incidence of food-borne botulism and trichinosis is associated with the preparation and consumption of local meat. As a result, studies in Canada indicate that food insecurity is more prevalent among indigenous populations than non-indigenous populations. One of the observed consequences of the changes in diet and lifestyle is an increased prevalence of overweight and obesity.

In indigenous populations where most of the dietary energy was provided from imported food before the turn of the millennium, this pattern has remained the same or increased further. In Russia, on the other hand, the socioeconomic changes and deterioration of the farming and livestock system in the northernmost parts after the dissolution of the former Soviet Union seem to have led to increased use of local foods in some populations. In indigenous populations, the trend toward a higher proportion of imported foods in the diets of young people has continued and appears particularly strong in Alaska, Yukon, and among the Canadian Inuit. Sweet and fatty store-bought food is becoming the main source of energy for children in this region, which has also contributed to an inverse relationship between nutrient density in the diet and age group. In the future, the low consumption of traditional food in the young age groups may perpetuate itself as they grow older and contribute to additional decreases in the consumption of traditional food. The overall contribution of traditional foods to energy intake in indigenous populations of Arctic Canada ranges from 10 to 36 %, with an average of 22 %; this figure is lower among children. Within that proportion, there have also been reports of changes in food choice; for example, more fish and fewer marine mammals are being consumed. In southern

Greenland (Inuit) and northwestern Alaska, local food constituted a lower proportion of the diet than in Canada. In Greenland, studies indicate that the consumption of local food among the Inuit has been reduced by about 50 % over the last 30 years, and today, the community-level proportions among adults range from 11 to 22 %. Women in most indigenous populations in Alaska, Greenland, and Arctic Canada generally consume proportionally less traditional food than men. Consumption is positively associated with the proximity of settlements to a coast or river, but varies with season. Education above elementary school level tends to lead to the consumption of more imported food. Alcohol consumption tends to be underestimated or not accounted for when estimates of energy consumption are made. Thus, the true proportions of energy intake from traditional foods among adults, especially men, are likely to be lower than those reported.

In general, the decreasing proportion of traditional foods in the diet has had a negative impact on the intakes of most nutrients, but imported foods appear to have contributed positively to the intake of vitamin C, folate, and possibly calcium. According to measurements of blood and dietary intakes, nutritional deficiencies of vitamin A, iron, calcium, and magnesium are prevalent in some communities. However, the amount of calcium provided through consumption of local foods is insufficiently researched. In terms of vitamin A, the types of imported foods consumed do not seem to provide the recommended amounts. The transition to diets mainly based on imported foods in the indigenous populations is likely to have led to insufficient intakes of other nutrients as well, especially in the young age groups. Nutritionally, the problem is not the imported food itself, but rather the widespread replacement of traditional food by a diet that is high in sugar and other foods with low nutrient density. However, updated, accessible, and comparable information about nutritional intake is relatively sparse from circumpolar populations outside Alaska, Greenland, and Arctic Canada. Information concerning populations in Russia, where the majority of indigenous and non-indigenous people live, has become more available and accessible since the previous AMAP assessment of human health in the Arctic (AMAP 2003), but still relatively little is known.

Contaminant levels in the Arctic, including levels in dietary items of fish, birds, seals, and whales, are in most cases lower than in more densely populated and industrialized regions. This geographic difference is especially pronounced for the organochlorines. Nevertheless, based on measured levels in Arctic biota and food samples, and based on studies of total dietary intakes of contaminants, it is apparent that dietary exposure to persistent contaminants and metals in Arctic indigenous communities is higher than in neighboring non-indigenous communities. The main explanation is that indigenous populations consume tissue from marine top predators that are not normally eaten in other parts of the world. Within the Arctic, there is therefore a higher exposure to contaminants through the diet in coastal dwelling ethnic communities that eat traditional foods such as marine mammals and some bird species, than in inland dwelling communities that eat reindeer/caribou and freshwater fish. Recent data from Arctic Russia illustrate the markedly higher intake of marine mammals by coastal people and greater intake of terrestrial mammals by inland peoples (Fig. 7.1).

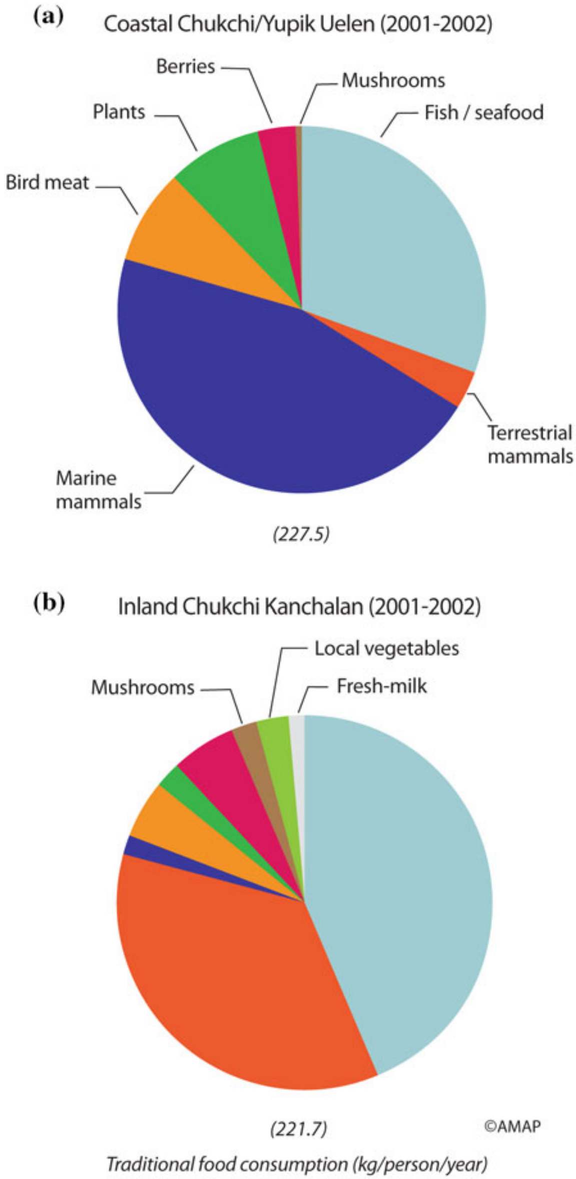

Fig. 7.1 Annual consumption of traditional food in **a** Uelen (coastal Chukchi and Inuit) and **b** Kanchalan (inland Chukchi) on Chukotka Peninsula, northeast Russia. Diagram provided by A. Dudarev (unpublished)

However, for the contaminants investigated, the levels in local food sources appear to be decreasing for some compounds in some areas, for example, polychlorinated biphenyls (PCBs) and DDT in Arctic Canada and Russia and PCBs in Greenland, whereas mercury (Hg) seems to be increasing in Arctic Canada and northwestern Greenland. The overall picture for chlordanes, toxaphenes, hexachlorocyclohexane (HCH), and mirex is uncertain; their levels appear to be decreasing in Arctic Canada, while increasing levels have been measured in Greenlandic food items and high levels of β-HCH were found in both animal biota and human plasma in the eastern part of Arctic Russia.

The research findings presented concerning exposure to contaminants through food consumption indicate that food items from marine mammals have the highest contaminant levels. But other marine foods, such as burbot (Lota *lota*), Greenland shark (Somniosus *microcephalus*), and liver from Greenland halibut (Reinhardtius *hippoglossoides*), and birds such as marine gulls and fulmars (including their eggs) also have relatively high levels of contaminants. Thus, further research and monitoring is needed for these species and for others with a similar role in the food chain. However, the levels of contaminants in the biota vary to a large extent within and between populations and within and between areas of the circumpolar Arctic. Exposure also very signifcantly depends on the amount of local foods consumed.

Analytical Quality Assurance and Quality Control

For more than two decades, researchers from AMAP countries have relied on the data produced by various laboratories to determine the exposure of Arctic populations to persistent organic pollutants (POPs) and heavy metals. In order to establish meaningful temporal trends and spatial distributions in exposure levels, the uncertainties resulting from variations in laboratory performance over time or among laboratories should be as low as possible. Given the analytical difficulties, variations of 20 % or less are considered acceptable. If there are significant variations in the accuracy of results, the biases should be estimated, so as to enable the application of correction factors, or such data should at least be flagged.

Laboratories should therefore participate in intercomparison exercises on a regular basis to determine their performance, relative to that of their peers, in an objective manner. Several international external quality assessment schemes (EQASs) exist for metals in human biological fluids, and laboratories should participate actively in one or more. This was not the case for POPs in human serum, however, where no ongoing intercomparison program existed when the AMAP Ring Test was initiated in 2001 (with the exception of the German EQAS based at the Institute and Out-Patient Clinic of Occupational, Social and Environmental Medicine of the University Erlangen—Nuremberg). Laboratories producing AMAP data were required to participate in the AMAP Ring Test. Results indicated that performance between 2001 and 2007 was generally acceptable. Some biases were identified, however, especially when new compounds were introduced. It was shown that lipid determinations were very method-dependent, with gravimetric methods yielding significantly underestimated results.

The difficulties inherent in comparing data from several laboratories, with different detection limits, were illustrated by reference to the new Russian data on persistent toxic substances. The use of appropriate statistical techniques was crucial in allowing meaningful comparisons.

Emerging persistent compounds create new challenges for laboratories, requiring new analytical technologies and lower detection limits. To ensure comparability of data produced by different laboratories, it is essential that EQAS be developed for these substances.

It is recommended that AMAP take a more proactive role in ensuring that all contributing laboratories produce good quality data. AMAP should determine, promulgate, and enforce formal performance requirements for the measurement of POPs, including emerging compounds, and in future should accept data only from laboratories conforming with these performance requirements. It is also recommended that serum lipids be measured using standard enzymatic methods rather than gravimetric methods.

Human Tissue Levels of Environmental Contaminants

Legacy Persistent Organic Pollutants (POPs) and Metals

This assessment is the third AMAP assessment (AMAP 1998, 2003, 2009a) of human health in Arctic regions but is the first to contain initial trend data for some organochlorines and metals. The assessment also incorporates the first comparison of all Russian Arctic regions as part of a circumpolar human health assessment.

Several important conclusions can be drawn from the information that has been presented in this section on the concentrations of organochlorine chemicals and certain metals in pregnant women and mothers, as well as in adults (both men and women) living in the circumpolar region.

Inuit populations of Greenland, Canada, and the USA continue to have higher concentrations of legacy contaminants, similar to what was seen in the previous assessments (AMAP 1998, 2003) as seen in Fig. 7.2, 7.3, 7.4. These concentrations are linked to the consumption of marine mammals. In addition, this assessment incorporates new regional sampling from Russia, and again, the Inuit or coastal populations that consume marine mammals have the highest concentrations of contaminants. Also, the patterns of DDT and DDE concentrations in some parts of Russia (with greater proportions of DDT) suggest that human exposures to pesticides are associated with local sources of contamination (e.g., soil contamination during food processing) and use of pesticides on the commercial food supply (see AMAP 2009a).

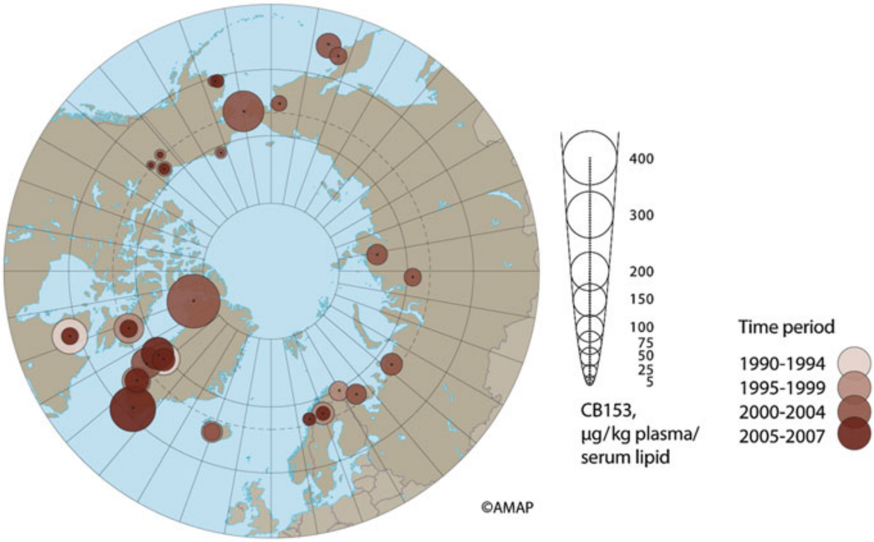

Fig. 7.2 PCB 153 concentrations in blood of mothers, pregnant women, and women of childbearing age in the circumpolar countries

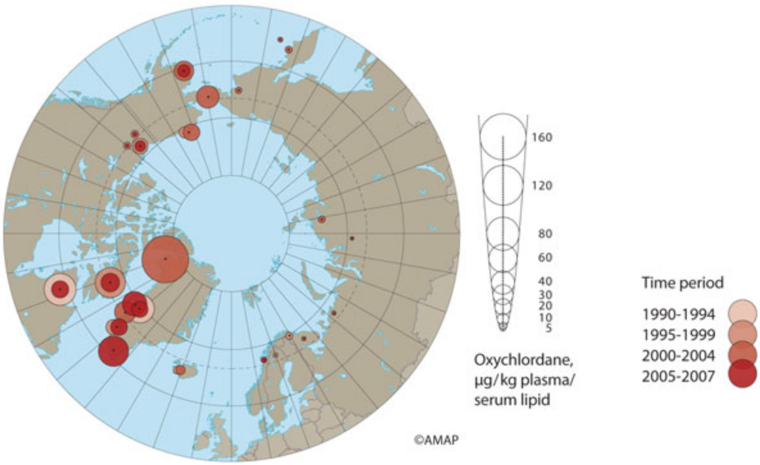

Fig. 7.3 Oxychlordane concentrations in blood of mothers, pregnant women, and women of childbearing age in the circumpolar countries

A series of biomonitoring studies have taken place in the Arctic which allow an initial assessment of changes in contaminant concentrations. The recent data presented in this assessment suggest that concentrations of contaminants such as DDE, PCBs, oxychlordane, Hg, and Pb are decreasing in many Arctic populations as

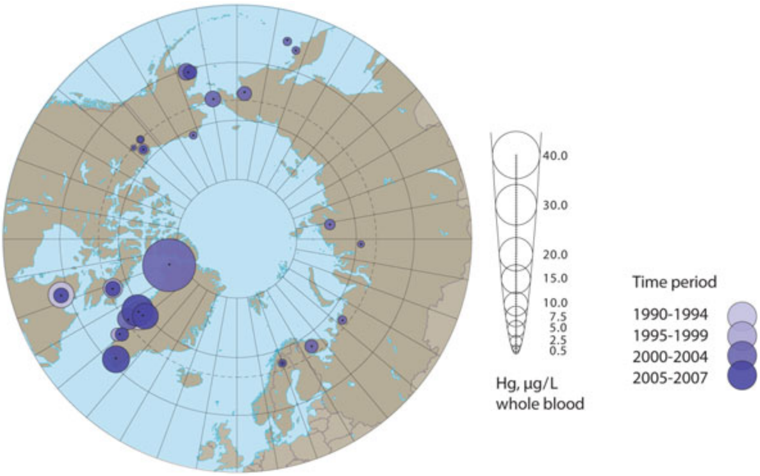

Fig. 7.4 Total mercury concentrations in blood of mothers, pregnant women, and women of childbearing age in the circumpolar countries

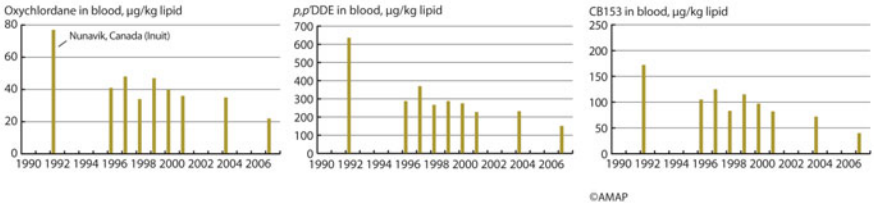

Fig. 7.5 Temporal trends of POPs and metals in maternal blood samples from Nunavik, Canada

shown in Figs. 7.2, 7.3, and 7.4 and in more detail for Nunavik, Canada, and Nuuk/Disko Bay, Greenland as shown in Figs. 7.5 and 7.6, respectively. The decrease may be related to lower concentrations of organochlorine chemicals in the environment as well as to dietary changes. Limited dietary data from Arctic Canada and Greenland suggest that changes are taking place in the traditional diet of Inuit mothers. Consumption of traditional food species that have higher concentrations of contaminants (e.g., marine mammals) is decreasing, while consumption of other species with lower concentrations of contaminants (e.g., fish and caribou) is increasing. This would result in an overall decrease in dietary exposure to contaminants. However, decreasing consumption of these traditional foods is not a positive indicator for human health as there are positive cardiovascular and neurobehavioral benefits to the consumption of these foods.

Guidelines are available for concentrations of PCBs, Hg, and lead (Pb) in blood. Inuit from the high Arctic that consume marine mammals continue to have higher

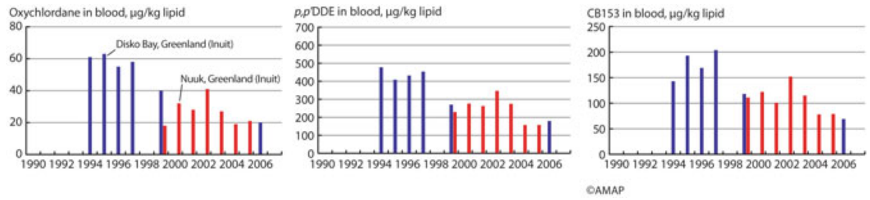

Fig. 7.6 Temporal trends of POPs and metals in maternal blood samples from Nuuk and Disko Bay, Greenland

proportions of the population that exceed these guideline values. In parallel with the decreases in the concentrations of PCBs and Hg in some populations of Arctic mothers, the proportion of mothers exceeding these guidelines is also decreasing.

Both blood and breast milk can be used for monitoring contaminant concentrations. The preference for blood monitoring in the Arctic is based on a need for information on both males and females, multiple age groups (children, adolescents, adults, senior citizens) and both POPs and metals. Several studies and data provided in previous AMAP assessments indicate that contaminant concentrations in blood and breast milk are well correlated and intercomparable.

The results presented in this chapter illustrate the need for continued monitoring of organochlorines and metal concentrations in human populations in the Arctic, especially for mothers or women of reproductive age and children. These data are important to allow a more complete assessment of trends in contaminant concentrations in a broader range of Arctic populations. The issue of decreasing contaminant exposure and decreasing traditional food consumption is very important in a number of circumpolar countries and is specifically addressed in Section "AMAP and Risk Communication."

Emerging Contaminants of Concern for Humans in Arctic Regions

In addition to the legacy POPs discussed in the previous section, a growing number of compounds are being detected in humans from the Arctic regions of the world. Some of these are fairly "new" compounds, whereas others have been around for decades but have only recently been detected due to advances in analytical techniques. The main compounds addressed in this section are the brominated flame retardants and the perfluorinated compounds, in particular the polybrominated diphenylethers (PBDEs), perfluorooctane sulfonate (PFOS), and perfluorooctanoic acid (PFOA). Until recently, there were few data available on human concentrations of these compounds.

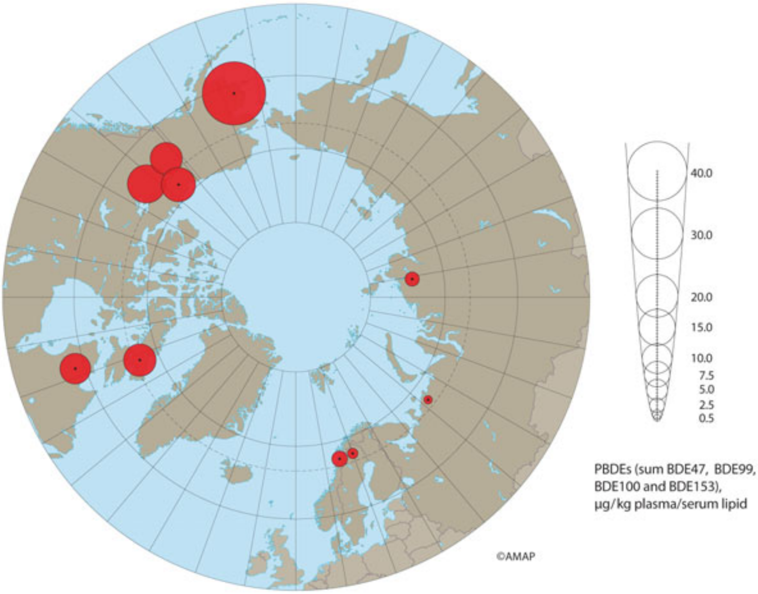

Fig. 7.7 PBDE (sum of BDE47, BDE 99, BDE 100, and BDE153) concentrations in blood of mothers, pregnant women, and women of childbearing age in the circumpolar countries

Concentrations of PBDEs in the Arctic are low in general, with the exception of Alaska where concentrations are much higher. Reported concentrations in Alaska are comparable to, or even higher than, those that have been reported in the US general population. Concentrations in the Canadian Arctic were considerably lower than in Alaska, but higher than in the European Arctic (Fig. 7.7). BDE47 was the predominant PBDE congener. Other brominated flame retardants investigated were tetrabromobisphenol A (TBBPA) and hexabromocyclododecane (HBCD), which showed low exposures based on limited data. Concentrations of PFOS were generally high throughout the Arctic and comparable to concentrations reported in several other heavily populated areas. There were minor intercountry differences, but comparisons were limited by small sample sizes. Concentrations of PFOA were much lower than the concentrations of PFOS but were elevated compared to the legacy POPs, and were clearly distributed throughout the Arctic. Limited data from Nunavik (Canada) showed an increase in ΣPBDE concentrations and a decrease in PFOS concentrations. There is, however, a clear need for ongoing monitoring of these compounds. Several other compounds of concern, such as short-chain chlorinated paraffins (SCCPs), parabens, and siloxanes plus metabolites of PCBs, should also be screened at regular intervals.

Genetics and Contaminants

This chapter describes the concepts of natural and selected gene diversity in relation to different populations, with emphasis on the Inuit and the possible link with and consequences of chemical exposures. Maternal inheritance of mitochondrial DNA (mtDNA) has been used to elucidate the ancestry of the Inuit and has demonstrated a close similarity in the circumpolar region with the hypothesis that the genetic impact of neo-Inuit (Thule groups) interbred with existing Dorset populations in Canada and Greenland.

Differences in the occurrence of diseases between Inuit and Caucasian populations have been reported, and the possible relation to gene polymorphism is speculated but needs further study. Serum lipids and apolipoproteins are risk factors for atherogenesis. Apolipoprotein E (APOE) differs between populations, and APOE polymorphisms were found to be associated with atherosclerosis in US white and black people that seems independent of serum lipids. However, the risk of coronary heart disease in Canadian Inuit was lower despite a higher incidence of disease-related *APOE* alleles. Nor was any association found between *APOE* genotypes and atherosclerotic lesions in Greenlandic Inuit. Studies have supported the hypothesis that the risk of coronary heart disease in Inuit is influenced by inherited genes as well as by diet and lifestyle.

In contrast to their Asian ancestors, the Inuit do not seem to be genetically protected against alcoholism. Co-exposure to alcohol and environmental chemicals might influence metabolism because some of the genes involved in metabolism of both types of exposure are identical. Ethnic differences in lactose tolerance are well known, and a low range of lactose tolerance is found in Inuit. Future studies might elucidate whether changes in diet and lactose tolerance have any impact on health risk.

The balance between xenobiotic absorption and elimination rates in metabolism is important factors in detoxification and prevention of chemical carcinogenesis as illustrated in Fig. 7.8. Approximately 80 % of all cancers have been estimated from epidemiological studies to be related to environmental factors, and cancer susceptibility can result from differences in genetic background for metabolism, DNA repair, and altered gene expressions of tumor-related genes. Thus, gene polymorphism in metabolizing enzymes, for example cytochrome P450, is suspected to influence susceptibility to environmental carcinogens. Studies have suggested a link between gene polymorphism in metabolizing genes, levels of POPs, and the risk of, for example, breast cancer in Caucasians. The incidence of breast cancer in Inuit is low, and the very few studies on polymorphism in metabolizing genes in Inuit await further studies with respect to the risk of breast cancer.

Epigenetics is a new paradigm in toxicology and teratology, a phenomenon that can be hereditary without any change in primary DNA sequence but which reflects a change in control of gene activity that arises from the interplay of DNA methylation, histone modification, and RNA-mediated pathways. Epigenetic regulation is a part of normal development and differentiation; however, by

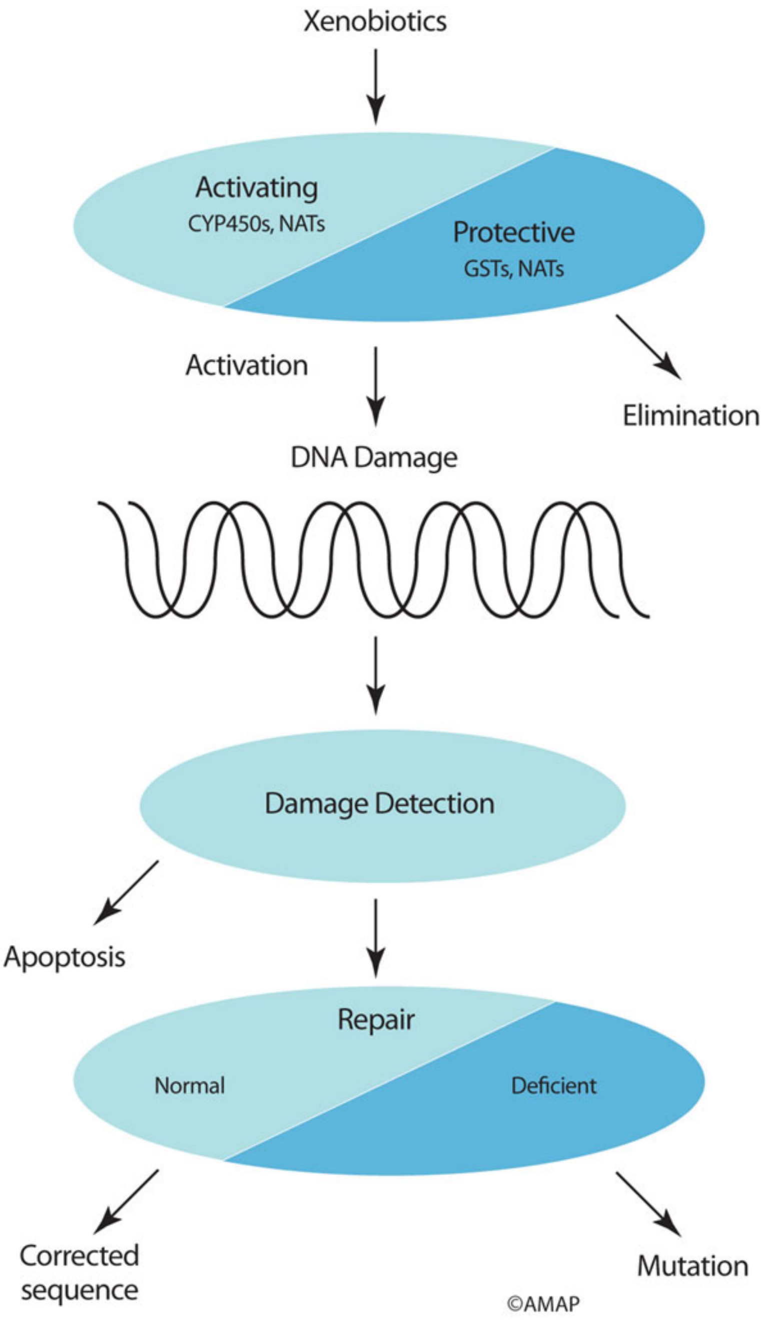

Fig. 7.8 Genetic variability in susceptibility to toxicants. *Source* cited in AMAP (2009a)

misdirection, it can cause diseases including cancer. Environmental exposure to PCBs and polybrominated biphenyls (PBBs), which are effective promoters in two-step-cancer models, implies the involvement of epigenetic mechanisms, and epigenetic hypermethylation of the tumor suppressor breast cancer *BRAC1* gene has been related to breast and ovarian tumors. In vitro studies showed that non-coplanar PCBs can decrease the *BRAC1* expression implying epigenetic mechanisms. A link between epigenetic mechanisms, exposure to gamma-hexachlorocyclohexane (HCH), and risk for breast and prostate cancer has also been suggested. Recent data for Greenlandic Inuit showed a link between the levels of serum POPs and the level of "global" DNA methylation, and further studies are needed to elucidate these cellular and biological epigenetic responses in relation to health.

Finally, the chapter describes ethnic gene polymorphism for the aryl hydrocarbon receptor (AHR), the androgen receptor (AR), and the estrogen receptor (ER). For the *AHR* gene, similar polymorphism was found for Caucasians and Inuit and was different to that for Japanese and African populations. Ethnic differences in the *ERα gene* have been reported and related to PCB exposure and breast cancer. However, no report on *ER* polymorphisms was found for Arctic Inuit people. In vitro *studies have shown complex patterns of responses* following exposure to natural as well as to synthetic estrogens (e.g., hydroxyl-PCBs) dependent on the cell context and ER form that must encourage researchers to elucidate the possible interaction between POP exposure, *ER* polymorphism, and health.

In summary, genetic background affects the impact and susceptibility to contaminant exposure and thus health risk. To date, little research on genetic polymorphism especially in relation to diseases has been undertaken on Arctic populations. Future studies should include genetics in parallel with lifestyle and contaminant exposure in order to provide a better insight into individual and population vulnerability to contaminants.

Interactions Between Contaminants and Nutrients

Over the last five decades, increases in the prevalence of obesity and subsequent health problems—metabolic syndrome, diabetes type II, and cardiovascular diseases—have been observed worldwide, including within populations living in the Arctic. These conditions have been regarded as lifestyle-related metabolic disturbances caused by hypercaloric and misbalanced diets in combination with a sedentary lifestyle. However, it has recently been suggested that exposure to contaminants might also play a role as illustrated in Fig. 7.9.

Maintenance of good metabolic health goes, however, beyond weight control. In this connection, the quality of the macronutrients plays a pivotal role. The dietary fat composition is especially important. Some of the saturated fatty acids, for example, palmitate, induce inflammation in adipocytes and will thus promote and exacerbate obesity and insulin resistance. Polyunsaturated fatty acids (PUFAs), both the n-6 and n-3 families, are essential. The two interact in the regulation of

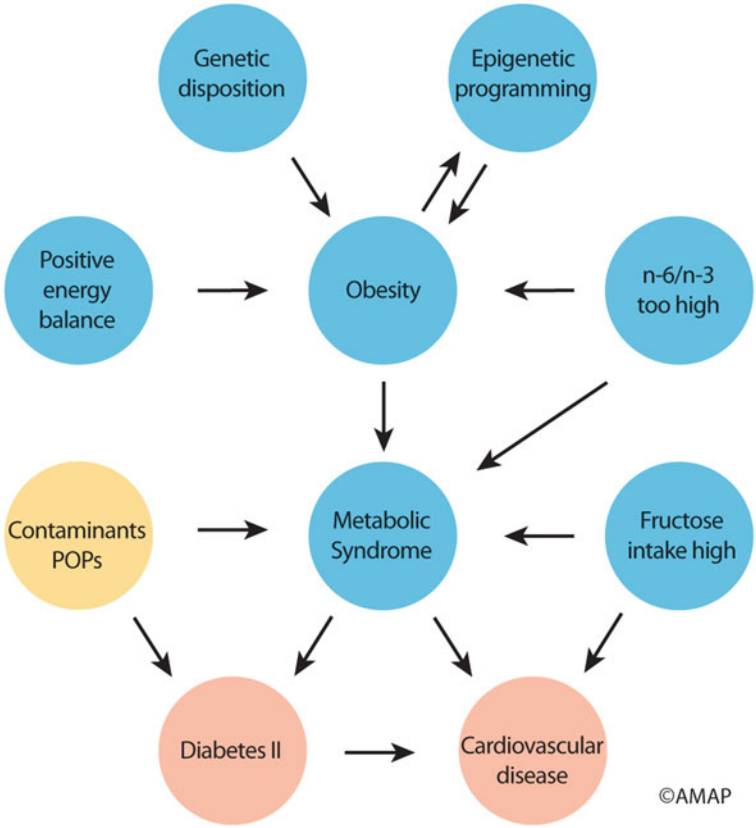

Fig. 7.9 Genetic and environmental factors increase in concert the susceptibility of an individual for public health problems such as type II diabetes and metabolic syndrome. All these conditions may have complications involving the cardiovascular system. *Source* cited in AMAP (2009a)

pro- and anti-inflammatory processes, and for this reason, a balanced dietary intake of n-6 and n-3 fatty acids is very important.

A high n-6:n-3 ratio will tend to be pro-inflammatory, while a very low ratio will induce immunosuppression. Furthermore, there is evidence that n-6, but not n-3, fatty acids act in a lipogenic manner. The optimal ratio is still under debate. The precolonial traditional Inuit diet provided a PUFA ratio of around 1, while a present-day Westernized fast-food diet provides a ratio of 10–20. In relation to cardiovascular health, a n-6:n-3 ratio of ~6:1 has been recommended.

Carbohydrate quality does not seem to play as important a role as does PUFAs, because carbohydrates do not interfere with gene expression of metabolic regulating enzymes. Carbohydrates serve as an energy source, and as long as the total energy supply is eucaloric, even the glycemic index value seems to be of minor importance. An exception is the high intake of fructose prevalent today as a result of the increasing industrialization of food production. Existing literature provides convincing evidence for this to be a major contributor to the increasing prevalence of obesity and metabolic syndrome on a global scale.

A common feature for dietary-induced obesity and metabolic syndrome is induction of oxidative stress at a cellular level and thus development of inflammation. This is also a characteristic effect of environmental xenobiotic exposure through the diet.

As a consequence, it is reasonable to speculate on an interaction between an unbalanced diet and concomitant exposure to xenobiotic compounds, where the contaminants may play a role as aggravating factors. Early findings from studies in the 1980s of a PCB-related and persistent increase in serum triglyceride concentrations clearly support a connection to xenobiotic exposure. Similarly, recent findings from Greenland were that the risk factor for cardiovascular diseases (TG/HDL ratio) increased with increasing body mass index, as expected, but that there was an additional increase in the TG/HDL ratio related to the POP exposure concentration. This indicates that in relation to obesity and metabolic syndrome, neither dietary imbalances nor exposure to contaminants should be evaluated separately, but should be addressed as a single entity.

Except for some Inuit populations, the general exposure level in the Arctic is below the guidelines for safe exposure to contaminants (AMAP 2003). The question is whether these relatively low exposures can have an influence on metabolic disorders. From the existing literature, it seems reasonable to speculate that in an organism already susceptible to developing a state of metabolic disorder, a concomitant exposure to dietary contaminants will, even at a relatively low level, be able to accelerate the processes, in an additive manner. At the moment, there is not much epidemiological evidence for this; however, experimental evidence indicates that this potential interaction is a public health issue that should be accounted for in future studies.

Divergent scientific and regulatory agency perspectives on contaminants in food have lead to contradictory advice and often to confusing public messages. It is, however, widely recognized that owing to the multifactorial character of the problems "cancer and non-cancer health impacts associated with environmental exposures generally cannot be directly isolated and measured," and for this reason, the discipline of risk assessment was established. A risk assessment approach using the formal tools established for this purpose is likely to be the most helpful approach for establishing risk and facilitating suitable risk management strategies (including dietary advice).

The concept that nutrition can modulate the toxicity of environmental pollutants, and vice versa, is a new way of thinking in the area of environmental health. Nutritional awareness in environmental toxicology is critical because of the opportunities to develop guidelines which specifically target exposed populations. In this way, nutrition may provide the most sensible means to develop primary prevention strategies for diseases associated with environmental toxicology.

To improve the understanding of the health effects associated with exposure to contaminants in the Arctic, it is recommended that circumpolar studies, including both nutritional and toxicological aspects, should be implemented on a large scale. Methylmercury- and POPs-related effects are still the key issues. However, the role

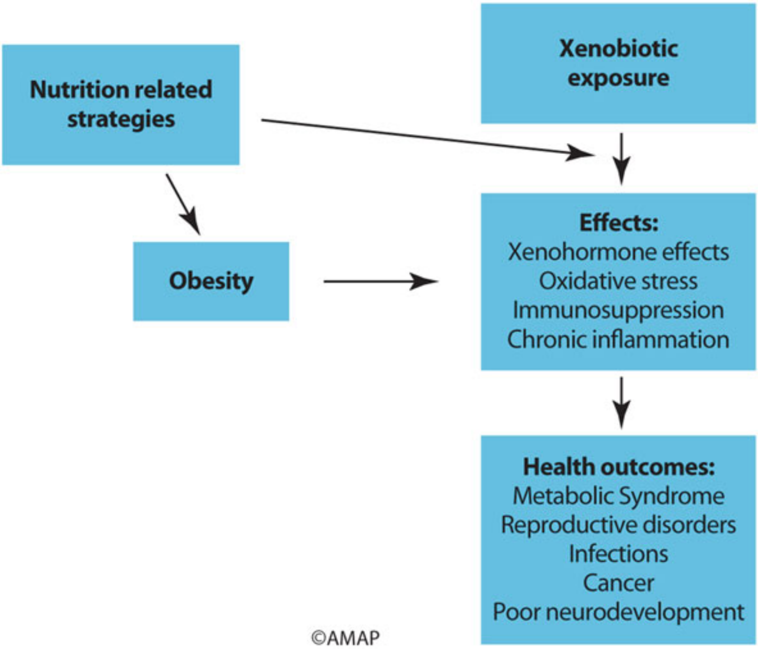

Fig. 7.10 Potential methods for improving public health. *Source* cited in AMAP (2009a)

of newly discovered contaminants, such as PBDEs, polychlorinated naphthalenes, and phthalates, should also be investigated. For exposure estimates, mixtures of contaminants and nutritional benefits of foods should be incorporated in the risk assessment profile. There is a need for better understanding of the interactions between nutrients and xenobiotic compounds, and risks should be evaluated in accordance with this interaction. Figure 7.10 provides a schematic representation of the relationship between nutrition and toxicology in the development of disease prevention strategies targeted at specific exposure populations.

It is essential that a better understanding is achieved of the health consequences of the dietary (and thus, nutritional) transition taking place within indigenous populations and the nutrient/contaminant interaction in Arctic populations, and that this information is communicated to the Arctic populations in a correct and understandable manner.

Public Health and the Effects of Contaminants

General Health of Arctic Populations

Human populations in the Arctic, especially indigenous populations, have comparatively poorer health status than populations from non-Arctic regions of the

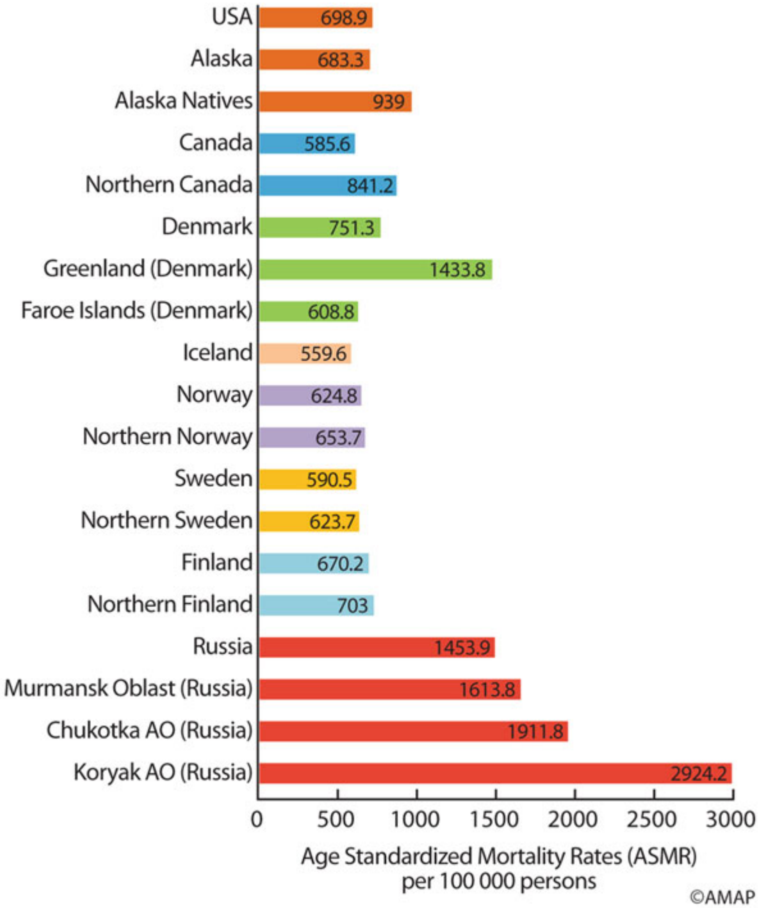

Fig. 7.11 Circumpolar age-standardized mortality rates by cause per 100,000 persons; standardized to European standard population. *Source* cited in AMAP (2009a)

same eight circumpolar countries as can be seen in age-standardized mortality rates in Fig. 7.11. Although infant death rates are lower and population longevity has improved, rates of several chronic diseases have been increasing. These changes are not uniform across the Arctic and are influenced by a number of determinants of health related to socioeconomic, dietary, and cultural influences. It is very likely that the higher prevalence of tobacco use, less active lifestyles, and consumption of more calorie-rich and nutrient-poor store-bought foods among some indigenous populations in the Arctic have contributed to an increasing burden of chronic diseases.

Health intervention strategies and population health in the Arctic will only improve with better information and better cooperation between all players. A full understanding of ethnic-specific health status can only arise if common health status indicators are selected and then monitored, reported, and analyzed consistently. This will require that indigenous peoples' organizations, tribal governments, and other levels of government in each Arctic country work together and support the establishment and/or active utilization of indigenous status identifiers for ethnic-specific public health surveillance.

While contaminants certainly play a role in the current health status of indigenous populations in many areas of the Arctic, it is also certain that other determinants of health are involved, including education, economic well-being, cultural strength, community engagement in shaping its present and future, lifestyle choices, genetic susceptibility, and availability of public health services.

Conducting epidemiological studies in the Arctic is difficult due to a variety of limiting factors: exposure to complex contaminant mixtures, small population size, contaminant–nutrient interactions, genetic factors, confounding factors, and health priorities. For this reason, epidemiological studies conducted in other parts of the world on POPs and metals-induced toxicity should be used as much as possible for risk assessment. However, different factors may limit the applicability of findings from these studies for Arctic populations.

Indigenous populations in the Arctic are exposed to mixtures of contaminants and primarily through food. Investigating the toxicological properties of environmentally relevant mixtures of POPs in laboratory species is essential for determining the biological plausibility of associations identified in epidemiology studies between exposure to these compounds and adverse health effects. It is clear from studies of chemical mixtures that data derived from single chemical experiments cannot be used to predict the risk resulting from exposure to complex mixtures of POPs. Interactions between components of the mixture not only modify the disposition of individual components but also their dose–response relationship for various developmental endpoints. These interactions, coupled with differences in nutrient levels, could explain some of the differences between the findings of the Faroe Islands and Seychelles Islands cohort studies.

New and promising techniques that evaluate the overall xenohormone activity in human blood are useful screening tools and can identify blood samples from population studies which warrant more expensive chemical analysis. The results from xenobiotics studies clearly indicate that the use of single or a few POPs proxy markers cannot alone be used for assessment of either bioactivity or possible adverse effects of POP mixtures on human populations.

Environmental POPs and Health

Many POPs, including PCBs, PCDDs, PCDFs, and pesticides, can mimic hormone activities. As potential endocrine disrupters, they are suspected to be capable of increasing the risk of cancer, birth defects, and reproductive and neuroimmune disorders. To date, no clear evidence for adverse endocrine-related human health effects of POPs has been obtained at the individual or population level. However, data from studies on wildlife species, laboratory animals, and biomarker effects in vitro have strengthened the need for further research to address the potential impacts of endocrine disruptors on human populations.

Although there have been associations found in individual cohort studies between fish consumption or POP exposure and newborn head circumference, birth weight, duration of pregnancy, and infant growth, the relationships observed differ between studies. It is clear that different contaminant levels, different mixtures of chemicals, diet, maternal susceptibility factors, and other confounding factors play a significant role in changing the associations found from one study to the next. It is also clear that exposure to POPs can adversely affect prenatal and postnatal development in human populations.

Results from the PCB studies conducted in the Faroe Islands and Nunavik to date suggest that prenatal exposure to PCBs is related to a relatively specific profile of cognitive impairments in children. Among the cognitive functions assessed, effects have been most clearly demonstrated on executive functions and speed of information processing and those effects can be responsible for the small decreases in IQ observed in most studies. Verbal abilities and visual recognition memory are also likely to be impaired.

Several recent studies in Arctic Canada confirm and support the relationship between exposure to certain contaminants and depressed immunity. Both PCBs and DDE are associated with a higher incidence rate of acute otitis media and respiratory tract infections in Inuit children during the first six months of life. Concentrations of lymphocytes and immunoglobulin A have been found depressed in comparative studies of breast-fed babies and bottle-fed babies. The effectiveness of vaccination programs among Inuit children and children from the Faroe Islands appear to be compromised by perinatal exposure to PCBs (as a marker of POPs). New research with piglets supports these findings and indicates that transplacental POP exposure leads to a reduction in antibody response.

Preliminary findings from a large Russian Arctic cohort adds evidence to the findings identified earlier that higher levels of maternal blood serum PCBs might be associated with more frequent occurrences of low birth weight, premature births, stillbirths, and menstrual irregularities. These possible adverse reproductive health effects of POPs and metals will require more in-depth evaluation. Detailed analyses of all available data and systematic epidemiological studies, which take into account relevant confounders and other contaminants, must be undertaken before conclusive statements can be made.

POP exposures have been suggested as the reason for observed alterations in birth sex ratios in animal populations and occasionally in human studies. New research results with pigs, which have a similar reproductive system to humans, indicate that exposure of sperm to environmentally pertinent organochlorine mixtures in vitro adversely affects oocyte development, polyspermy, sperm fertility, and embryonic development. However, a comparison of existing population studies, one including Arctic countries, did not reveal any definitive or consistent relationship between POPs, sperm X:Y ratios, or male-to-female birth ratios. Emerging data from a relatively small cohort in the Russian Arctic indicate that increasing maternal PCB concentrations may be associated with an initial effect of increasing the male-to-female newborn ratio; however, causality has not been determined and the increase in the ratio appears to disappear in the highest concentration group. The possible effects of other contaminants have not been determined. Systematic epidemiological studies, including all possible confounders and other relevant contaminants, must be performed before any conclusive statements can be made about contaminants and sex ratios in Arctic populations.

A large international study indicated some links between POP exposure and biomarkers of male reproductive function. Associations were found between high PCB153 serum levels and low sperm counts, decreased sperm motility, and damage to sperm chromatin integrity in some of the subpopulations studied. In spite of these effects, fertility was not related to POPs except in Inuit. Definitive conclusions cannot be derived from these studies, in part because the two POP proxy markers measured did not represent the biological activity of the entire POP mixtures. There are also open questions related to the role of genetic background, lifestyle, and/or diet nutrition factors such as trace elements/antioxidants (e.g., selenium) that may interfere with the possible adverse health effects of POPs.

Exposure to POPs may contribute to the development of metabolic syndrome. The endocrine-disrupting properties of several contaminants, especially dioxin-like compounds, can affect glucose and lipid metabolism, which in turn affect the onset of metabolic syndrome. Genetic factors and lifestyle are also important determinants of metabolic syndrome. The dramatic increase in the rate of diabetes among Inuit and Alaskan Natives may be affected by multiple factors; however, the role played by contaminants in obesity, metabolism, and diabetes warrants urgent study.

Environmental mixtures may also be able to affect bone density (diminished bone cortical area and bone mineral content) in male and female rat pups. These findings may be significant for humans as indications of a negative association between PCB plasma levels and quantitative ultrasound bone parameters have been reported in a group of peri- and postmenopausal women from Greenland.

Environmental Metals and Health

The growing fetus and newborn children are especially sensitive to the toxic effects of environmental Hg and Pb. Animal studies indicate that exposure to environmentally relevant mixtures of POPs and metals has significant effects on reproduction. Exposures led to decreases in maternal weight gain, weight gain in offspring, and increased mortality rates in pups prior to weaning. Interactions between MeHg and POPs in mixtures warrants further study as lower levels of MeHg administered alone have been found to lead to more pup mortality than found in mixture studies containing higher levels of MeHg.

Potential neurobehavioral effects associated with MeHg exposure have been found in the Faroe Islands in the domains of verbal function, visuomotor integration, and attention. However, the most consistent marker of prenatal MeHg exposure is delayed auditory processing assessed from brain stem auditory event potentials. Because of the inconsistencies between studies, there is a need for additional well-conducted prospective studies to elucidate the specific growth and neurobehavioral effects of MeHg and to assess the impacts of differences in maternal diet during pregnancy on susceptibility to MeHg exposure.

Recent studies in the Faroe Islands, Greenland, and Nunavik all indicate that Hg can affect circulatory parameters such as pulse pressure, heart rate and heart rate variability, blood pressure, hypertension, and atherosclerosis. Prenatal exposures to MeHg may also affect the development of cardiovascular homeostasis. If these preliminary findings are confirmed, the estimated attributable burden of diseases due to contaminant exposure might increase. Even small relative risks have a large impact on diseases having high incidence and mortality and could affect policy development on safe levels of exposure. Confirmation of these findings in other studies is needed as the current findings have potentially significant implications for Hg intervention policies. In addition, more research is needed to determine the relationship between changes in risk of cardiovascular disease and changes in diet among Arctic indigenous populations.

It is highly likely that oxidative stress is a significant underlying biochemical mechanism in MeHg neurotoxicity. MeHg neurotoxicity can be inhibited by various antioxidants, including Se. New studies in Arctic Canada indicate that Hg exposure may diminish defense mechanisms against oxidative stress by limiting the availability of glutathione, while Se may afford protection by favoring the destruction of hydrogen peroxide. It will be important to continue to assess oxidative stress in adult residents of Nunavik and to further understand interactions which affect the mechanisms of Hg toxicity.

Lead is well known to adversely affect neurodevelopment and behavior in children. Until recently, studies of behavioral effects of Pb in children have only confirmed effects from postnatal exposure, not from prenatal exposure. New studies with children from Nunavik have shown that cord blood Pb concentrations were related to observational measures of inattention even at cord blood Pb concentrations below 10 microg/dL. The new data indicate that behavioral effects of low prenatal Pb exposure are likely to be observed when testing protocols include sensitive measures of behavior.

Some new associations have been reported in Arctic Russia between spontaneous abortions and Hg levels in blood. No negative associations were found between maternal exposure to nickel and the risk of delivering a newborn with malformations of the genital organs. Further study to confirm the associations between Hg exposure and abortion rates is warranted.

AMAP and Risk Communication

AMAP was established by the Arctic Council to investigate the spread and effects of pollution from the industrial regions in the temperate zones to the Arctic. AMAP's main role has been to harmonize the compilation of data to establish the levels and extent of contamination in the circumpolar area and also to initiate research into the correlation between contaminants and the health status of these areas.

Because all Arctic circumpolar areas are represented within the Arctic Monitoring and Assessment Programme, the present assessment provides a reasonably reliable picture of the status of human exposure to heavy metals and legacy POPs within the Arctic. Furthermore, because the exposure measurements have been repeated over time, it is possible to comment on the development of trends with regard to pollution in many areas. Effects studies have also been undertaken. By comparing documented levels of exposure to the effect studies from the Arctic area and the toxicological literature in general, it may be concluded that in several places, it is possible that exposure via food has adverse implications on the health of populations. Studies have demonstrated that the main source of exposure to contaminants in the Arctic is traditional food, especially marine animals.

The benefit/risk management process for traditional foods in circumpolar countries has been challenged by the issue of environmental contaminants entering the Arctic food chain for many decades. Although a framework for risk management has been adopted in many countries, no common process to integrate all benefits and risks, qualitative and quantitative information, and to assess and direct decision making on this topic has yet been developed.

In societies with a wide range of alternative dietary choices and broad cultural means of existence, dietary adjustments are easy to implement. This is not the case in the Arctic area, however, where communities depend heavily on marine food, especially marine mammals. Many populations in the circumpolar area experience real food insecurity. Will there be enough food? Will the healthy food be available?

Marine mammals are the focus for many Arctic cultures. It has been possible to achieve some significant reductions in exposure to heavy metals and POPs by eliminating the most contaminated species and organs from the diet. It is feared, however, that a change in food choices and lifestyle will alter community culture to such a degree that some of the cultures will not survive. Another reason for caution when communicating adverse effects of certain pollutants is that, theoretically, contaminated food also contains compounds that have a positive impact on health, such as marine fatty acids (see Sections "Food, Diet, Nutrition and Contaminants" and "Interactions between Contaminants and Nutrients").

All these considerations constitute "The Arctic Dilemma" that traditional food items have both positive and negative properties and that the management and communication of this dilemma are complicated and dependent on geographic, demographic, cultural, and social conditions (Sections "Food, Diet, Nutrition and Contaminants" and "Interactions between Contaminants and Nutrients").

Dietary guidelines for individual communities are principally the responsibility of the national/local health authorities. The aim of the communication is to provide balanced information allowing people to make informed rational choices about how to reduce their exposure to toxic substances and at the same time ensure that their food has satisfactory nutritional and aesthetic properties. Information alone is not enough, however. There is also an obligation on local authorities to make healthy food available even in small communities, so that people can actually carry out these informed rational dietary choices. This is more of a political and logistic issue.

Because AMAP is able to contribute data and information on exposure status in local areas over time and because such information is of general interest to populations living in the Arctic, it has to some extent initiated and participated in public health assessments of both the beneficial and adverse effects resulting from the various sources of exposure, such as marine mammals.

Communication Strategies and Problems in the Arctic

Information presented in this chapter highlights how risk communication has been conducted differently in different countries and regions. In most countries, risk communication has been undertaken by national authorities, seldom or never by communication professionals. Communication of information resulting from AMAP's activities has been given much consideration from the start, and on some occasions, professional communicators have been used to ensure that the information is communicated as effectively as possible. Nevertheless, the goal of giving clear but nuanced information on the sometimes complicated conclusions arising from its work has not been totally satisfactory in all cases. This may be partly due to none of the Arctic countries having an official strategy for the communication of the results.

In the Faroe Islands, risk communication has had measurable positive effects on both the food choices of target groups (children and young woman) and health parameters. In Norway, Sweden, Finland, and Iceland, human health issues resulting from the accumulation of contaminants from Arctic food chains have been very limited and so the need for dietary advice to the populations has also been limited.

It is communities in northern Alaska, Canada, Russia, and Greenland that are subject to "The Arctic Dilemma" where risk communication has been complicated and often unsuccessful. The difficulty is to convey insights to all citizens of exposed populations that will enable them to weigh the benefits against the drawbacks in their dietary choices.

One particular difficulty has been in identifying the receiver for the risk communication. Is it the health professionals of a given country or is it the entire, affected population? Another difficulty is the form of the communication. A simple message may be easy to communicate, but equally easy to misunderstand, for example, that all seafood is considered contaminated. On the other hand, balanced and targeted risk communication would only reach a specific and narrow receiver population.

Another difficulty has been that toxicological terms are relatively alien to the general population. It is assumed that the majority of a population wants questions about the negative impacts of contaminants answered by a yes or no and not by a number of statistical variations which they can interpret to fit their own personal preferences. However, it is likely that a substantial proportion of the communicated messages with regard to contaminants in the circumpolar area have not been fully understood.

Risk perception research has been valuable in showing the local perspectives and understanding of the issue; surveys and other qualitative assessments have shown that while the majority of the population is aware of the issue of "contaminants," some confusion with regard to the "chemical-only" nature of the use of terms by the research community exists and may create barriers to understanding the messages given out by northern health professionals and researchers.

In the Canadian Arctic, it appears that women of childbearing age have received and understood the least regarding this issue, yet may be the most at risk to contaminant exposure because of the sensitivity of the developing fetus. This is therefore the most important target audience around which to develop risk reduction strategies and messages.

Research conducted in the Canadian Arctic indicates that contaminants are not direct determinants influencing food choice among northern residents; however, indirect impacts of food choice and hunting catch rejection may be taking place in some regions.

A circumpolar assessment of communication efficiency should be established. New studies on risk communication result in new materials for better communication. More work is, however, needed on the effectiveness and effects of risk–benefit communication. The dissemination of AMAP assessment reports and information, both to the public and to health authorities in the Arctic, should be improved.

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Chapter 8
The Role of Estrogens and Estrogenic Metabolites and Male Reproductive Health Disorders

James Gomes and G. Roche

Abstract In the past five decades or so, there has been an explosion of chemical compounds that has entered the market for use in industry, trade, and commerce. The chemical compounds have also permeated household cleaning and other products and also personal care products. It is believed that currently about 400 million tons of chemicals are used in commerce in the European Union for industrial, household and personal care products. Endocrine-disrupting chemicals (EDCs) are synthetic chemicals which can interfere with endogenous sex hormone function leading to a range of developmental and reproductive anomalies. The exposure to EDC is ubiquitous because of the use of these substances in commerce, trade, household products, and personal care products and in food processing and packing. EDCs are used in pharmaceuticals, in agriculture as pesticides, in industry as additives and for their specific activities, in household and personal care products for their antibacterial and antiseptic properties, and in commerce for packaging food and as food items as in natural products (soy). This review examines the exposure to EDC among the males during embryonic and postnatal development. The impacts of exposures during different phases of life on the male reproductive health during embryonic development, adolescence, and adult life have also been explored. Human exposure to EDCs occurs through different routes including inhalation, gastric, dermal, and use of personal products; however, the biggest source of exposure is dietary for all ages. Gestational exposure occurs from maternal exposure during pregnancy, infants

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are exposed through milk ingestion, and adults are exposed through consumption of phytoestrogens. A range of adverse health observed in males exposed to EDC at different points in time is reported to be associated with exposure.

Abbreviations

AR	Androgen receptor
BPA	Bisphenol A
cAMP	Cyclic adenosine 3′ 5′-monophosphate
DAX1	Dosage-sensitive sex reversal, adrenal hypoplasia critical region, on chromosome X, gene 1
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
DEHP	Diethylhexyl phthalate
DES	Diethylstilbestrol
DNA	Deoxyribonucleic acid
EDs	Endocrine disruptors
EDCs	Endocrine-disrupting chemicals
ER	Estrogen receptor
ERα	Estrogen receptor alpha
ERβ	Estrogen receptor beta
FSH	Follicle-stimulating hormone
HPTE	2,2-bis-(p-hydroxyphenyl)-1,1,1-trichloroethane
LH	Luteinizing hormone
PCBs	Polychlorinated biphenyls
PCDF	Polychlorinated dibenzofuran
PR	Progestin receptor
RNA	Ribonucleic acid
SF1	Steroidogenic factor 1
StAR	Steroidogenic acute regulatory protein
TDS	Testicular dysgenesis syndrome

Introduction

The identification and characterization of exogenous endocrine disruptors (EDs) was first reported by Rachel Carson in her book, Silent Spring, first published in 1962 (Carson 1962). In her book, she cautioned the use of certain chemicals with endocrine disrupting properties. These chemicals described as EDs include dichlorodiphenyltrichloroethane (DDT), polychlorinated biphenyls (PCBs), bisphenol A, phthalates, polybrominated diphenyl ethers, and others. The term endocrine disruptor was coined by Dr. Theo Colborn in her book Our Stolen Future (Colborn 1996). Dr. Colborn in her book has described and elucidated pathways through which endocrine-disrupting chemicals (EDCs) could affect the reproductive function, growth and development, and expression of secondary sexual characteristics. Since the publication of these pioneering documents, an immense amount of scientific data has been published arguing for and against endocrine disruption in animals and humans. Currently, observational and experimental studies in toxicology and epidemiology document the exposure and the adverse effects from EDC, and although the evidence appears to be positive, it still remains to be proven conclusively. However, in recent years, a number of mechanisms and modes of action have been elucidated; this text reviews these mechanisms and describes the reported effects believed to be associated with exposure to ED.

Evidence in support of endocrine disruption gained prominence and public interest when reports of treatment of pregnant women between 1945 and 1971 with synthetic estrogen diethylstilbestrol (DES) were published. These reports suggested that exposure to DES increased incidence of cryptorchidism and hypospadias and decreased sperm counts in the sons of these women and clear cell adenocarcinoma of the vagina in the daughters (McLachlan 1981; Herbst et al. 1970, 1971). Genital abnormalities have also been reported in males exposed to DES (Whitehead and Leiter 1981). Since then, a number of scientific studies have reported on the changes in the male reproductive health from exposure to estrogenic substances in the ambient environment and occupational places, lifestyle including dietary habits, and use in trade and commerce (Jensen et al. 1995; Toppari et al. 1995). Adverse health effects including cancer and gestational and birth outcome affects have also been observed in women from exposure to estrogenic substances.

EDCs are chemicals that possess hormone such as activity and exhibit properties similar to endogenous hormones but with adverse consequences. EDCs are known to disrupt mammalian endocrine system: (a) adversely affect the development of embryo and induce changes that may manifest in adolescence and adult life, (b) depending on the period of exposure can change the course of early childhood development, (c) adverse effects are often delayed until the offspring reaches adolescence or adulthood (Colborn et al. 1993). Since the Second World War, large quantities of EDCs have been released in the environment and are still emitted because of their use in commerce and trade. These chemicals (Table 8.1) function through ligand–receptor interaction similar to endogenous hormones and interfere with the functioning of the hormonal receptors. EDCs function as agonists or antagonists of receptors and disturb organ differentiation (Gray 1992; Petersen et al. 1993; Colborn et al. 1993). In addition to mammary glands, fallopian tubes, uterus, cervix, and vagina in the females and prostate, seminal vesicles, and testes in males, and other organs such as brain, thyroid gland, liver, kidney, and immune system in males and females are also targets for steroid hormone action (Colby 1980; McEwan 1981; Leatherland and Sonstegard 1982; Grossman 1984).

EDCs are synthetic chemicals which can interfere with endogenous sex hormone function leading to a range of developmental and reproductive anomalies. EDCs act by altering estrogenic and antiandrogenic activities (Akingbemi and Hardy 2010). The reported adverse health effects from exposure to EDCs are believed to be manifested both at low-exposure levels and at high-exposure levels, but these

Table 8.1 Chemicals in the environment with endocrine disrupting properties and associated hormonal receptors or disruption activities

Type of compound	Name of compound	Hormonal activity	References
Pesticides	2,4-dichlorophenoxyacetic acid	Estrogenic	Gorzinski et al. (1987)
	2,4,5-trichlorophenoxyacetic acid	Antiestrogenic	Lemaire et al. (2006)
	Alachlor	Antiestrogenic	McKinlay et al. (2007); Heydens et al. (1994); Eriko et al. (2003)
	Amitrole	Thyroid disruption	Hongmei et al. (2011)
	Atrazine	Estrogenic	Fan et al. (2007)
	Benomyl	Estrogenic	Morinaga et al. (2004)
	Carbaryl	Thyroid disruption; LH and FSH disruption	Sun et al. (2008), Fattahi et al. (2012)
	Chlordane	Androgen disruption	McKinlay et al. (2007), Lemaire et al (2004)
	Dicofol	Estrogenic	Hoekstra et al. (2006)
	Dieldrin	Estrogenic	Soto et al. (1994)
	Dichlorodiphenyltrichloroethane (DDT)	Estrogenic	McKinlay et al. (2007), Bulayeva and Watson (2004)
	Endosulfan	Estrogenic	Soto et al. (1994)
	Heptachlor	Antiestrogenic	Oduma et al. (2006)
	Hexachlorobenzene	Thyroid and androgen disruption	Foster et al. (1993), Ralph et al. (2003)
	Hexachlorohexane	Estrogenic	Wong et al. (2006)
	Lindane	Thyroid disruption and estrogenic	Rawlings et al. (1998)
	Mancozeb	Cortisol disruption, antiestrogenic	Bisson et al. (2002), Baligar et al. (2001)
	Maneb	Testosterone and thyroid disruption	McKinlay et al. (2006), Manfo et al. (2011)
	Methoxychlor	Estrogenic	Laws et al. (2000)
	Metribuzin	Thyroid disruption	Mnif et al. (2011)
	Nitrofen	Antiestrogenic	McKinlay et al. (2007), Mnif et al. (2011)
	Oxychlordane	Thyroid disruption	Villanger et al. (2011)
	Parathion	Thyroid disruption, estrogenic	Liu et al. (2006), Liu et al (2009)

(continued)

Table 8.1 (continued)

Type of compound	Name of compound	Hormonal activity	References
	Tributyltin	Testosterogenic	Saitoh et al. (2001)
	Trifluralin	Testosterone disruption, estrogenic	Shariati et al. (2008), Rawlings et al. (1998)
	Zineb	Thyroid disruption	Nebbia and Fink-Gremmels 1996
	Ziram	Thyroid disruption	McKinlay et al. (2007), Marinovich et al (1997)
Metals	Cadmium	Estrogenic	Stoica et al. (2000)
	Lead	Antiestrogenic, testosterone disruption	Ronis et al. (1998)
	Mercury	Antiestrogenic, testosterone disruption	Tan et al. (2009)
Other chemicals	Bisphenol A	Estrogenic	Welshons et al. (2006)
	Dioxins	Antiestrogenic	Okino and Whitlock (2000); Mocarelli et al. (2008)
	Ethinylestradiol	Estrogenic	Laws et al. (1999)
	Nonylphenol	Estrogenic	Laws et al. (1999)
	Phenosulfothiazine	Estrogenic	Berthois et al. (1986)
	Polychlorinated biphenyls	Thyroid disruption, estrogenic	Pliskova et al. (2005), Iwasaki et al. (2002)
	Polybrominated biphenyls	Thyroid disruption, estrogenic	Kitamura et al. (2008)
	Pentachlorophenol	Antiestrogenic	Jung et al. (2004), Orton et al. (2008)
	Phthalates	Estrogenic	Takeuchi et al. (2005)
	Styrenes	Estrogenic	Ohyama et al. (2001), Kitamura et al. (2003)

effects appear to be minimal in the mid-level range. Children are at a greater risk of toxicant-related illnesses compared to adults because of their increased vulnerability during growth and development. Many of these effects are mediated by agonists and antagonists of the steroid hormone family of nuclear transcription factors—the estrogen receptor and the androgen receptor (AR) (Toppari et al. 1996a, b; Kelce et al. 1998; Jobling et al. 1995)

A number of studies in recent years have reported on the increasing prevalence of male reproductive disorders in the developed world (Safe 2000; Hendelsman 2000; Ahlborg 1995). However, there are those studies which have failed to observe the increase in the prevalence of male reproductive disorders. These studies have characterized the apparent increasing trend as spurious and are mainly due to

Table 8.2 Adverse reproductive health effects during different phases of life in the males

Embryonic and fetal development	Adolescence	Adult life	Non-specific phase
Cryptorchidism	Development of secondary sexual characteristics	Testicular cancer	Testicular dysgenesis syndrome
Hypospadias	Seminal vesicles	Decreased sperm counts	
	Epididymis	Altered sperm quality	
	Thyroid	Prostate cancer	
	Skeletal		
	Brain		

biased data collection and subjective coding or classification of the disorder (Wolff et al. 1993).

The prevalence of male reproductive disorders has been on the increase in the past couple of decades. These male reproductive disorders have been identified in three distinct phases of male life span: embryonic development, adolescence, and adult life. Although some of these disorders are identified in certain specific phase of life, others have been identified in more than one phase of life (Table 8.2). The male reproductive disorders discussed in this review are altered sperm counts, cryptorchidism, hypospadias, testicular cancers, and testicular dysgenesis syndrome (TDS). However, the debate on the role of exogenous hormones (xenobiotics) in the development of adverse reproductive health effects and other adverse health effects in males and females remains to be conclusive for a number of reasons.

A number of factors including environmental and occupational exposures, lifestyle factors including alcohol consumption, and dietary factors have been associated with the increasing incidence of adverse reproductive health effects The rising trend in the incidence of male reproductive disorders in the developed world is believed to be caused by exposures to EDC from occupational and environmental sources and the use of these substances in trade and commerce and in personal care products and diet. This review examines the different sources of exposure to EDC among the males during embryonic and postnatal development and its impact on the male reproductive health during embryonic development, adolescence, and adult life.

Methodology

The search strategy that was developed for searching the scientific literature consisted of two sets of search terms: (a) outcome terms—male reproductive health, male reproductive disorders, sperm counts, cryptorchidism, hypospadia, testicular descent, testicular cancer, and TDS and (b) exposure terms—estrogens, estrogenic substances,

Table 8.3 Results of data search and number of articles included in the review following inclusion and exclusion criteria

Description	Number of articles	Number of articles excluded
Total number of articles obtained from all databases	1524	
Total number of articles retained after title reading	762	762
Total number of articles retained after abstract review	341	421
Total number of articles retained after review of pdf	113	228
Total number of articles excluded as not relevant	28	
Articles added by hand searching	38	
Total number of articles included in this review	123	

xenoestrogens, antiestrogenic, and estrogenic metabolites. The search strategy was developed in PubMed database, and when the search was optimized, it was used to search other databases including Ovid Medline, Toxline, Embase, EMBO reports, and Cochrane Reviews. Hand searching was also used to search specific scientific articles that were identified in either the bibliography or those obtained by searching the Internet using search engines such as Google and Google Scholar.

The results of search strategy from all the databases were combined in RefWorks, and inclusion and exclusion criteria were applied. Inclusion criteria selected all articles in English and French for all years. The studies with maternal and paternal exposure to xenoestrogens from environmental and occupational exposures; in utero exposures were also included, and scientific articles which contained both the terms such as estrogens and sperm counts, cryptorchidism, hypospadias, and TDS were included. The exclusion criteria exclude articles in foreign languages, androgenic or related exposures, and adverse male reproductive health effects not associated with estrogens or xenoestrogens. A total of 1524 scientific articles were obtained (Table 8.3) and systematic evaluation of the title, abstract, and review of whole article excluded 1413 articles as not relevant. In this review, 111 relevant articles were included, and the data were extracted from all these articles.

The selection of the relevant articles was reviewed by the two coauthors (PO and GR). When there was a disagreement, the issue was resolved by reasoning and discussion; at times, the first author (JG) casted a tie-deciding vote. All the selected articles were exported to RefWorks, and a database was maintained in RefWorks. Each of the selected articles was reviewed, and the relevant data were extracted using a predetermined format. As per this format, information was collected on the citation, population studied, and methodology used in collecting the data (all original research articles were included; review articles were not included in the results but were used in the introduction and discussion sections). The data were extracted by the coauthors independently, and a 10 % sample of articles from each reviewer was re-evaluated by the other reviewer. The concordance between the two reviewers was examined, and a kappa value of 0.7 was observed between the two reviewers.

Endocrine Disrupting Substances

Natural steroid hormones are key enzymes in regulating developmental processes in target tissues such as the reproductive tract and in the development of secondary sexual characteristics. Endogenous hormones accomplish their function by binding to their respective receptors and stimulating or inhibiting various cellular pathways. The hormones bind to the receptor in a ligand–receptor interaction to form hormone–receptor complexes which then recruit coactivators and binding molecules to form a DNA-binding molecule which binds to the DNA leading to a cascade of events and expression of functional proteins. These proteins assist in regulating the developmental process in target tissues. Absence of these proteins lead to alterations in regulation and development of adverse reproductive health outcome.

Environmental chemicals that mimic natural endogenous hormones are pseudohormones that act by: (a) interacting with the receptor and causing an undesired effect, (b) interacting with the receptor without producing any downstream effect, (c) interacting with the receptor synergistically causing an exaggerated and hyperactive effect, and (d) interacting with the receptor and blocking the receptor function with no desired outcome. These substances are known as ED and cause a reproductive adverse health effects in the first or second generation. EDs also known as xenoestrogens act through other mechanisms which may be agnostic or antagonistic and interfere with synthesis, transport, and metabolism of hormones and their metabolites. The list of EDs currently known is long and emerging (Toppari et al. 1996a, b; Crain et al. 2008). However, the determination of the ED is a major problem because of the vast number of chemicals used in industry, trade and commerce, and the diversity of hormonal properties that these chemicals exhibit.

Adverse health effects associated with the male reproductive function and development have been linked with exposure to xenoestrogens. These effects are believed to be mediated by both agonists and antagonists of estrogen receptor alpha (ERα), estrogen receptor beta (ERβ), and AR (Kelce et al. 1998; Jobling et al. 1995). Mutations in ERα and ERβ have been identified with impaired sperm viability and fertility although the sperm numbers are normal in human epidemiological studies and also in animal studies (Eddy et al. 1996). Agonists of the steroid hormone receptors are reported to induce a negative feedback loop on pituitary gonadotropins, luteinizing hormone (LH), and follicle-stimulating hormone (FSH) (Shuipnik et al. 1996) leading to decrease in biosynthesis of testosterone by Leydig cells. Depletion of testosterone levels adversely affects sexual differentiation during prenatal development and development of secondary sexual characteristics and expression of male phenotype postnatally (Akingbemi and Hardy 2001).

Effects Observed in Human from Estrogens and Estrogenic Metabolites

DES has been the most researched compound after it was observed that women who were prescribed DES during gestation produced a range of adverse effects on the reproductive system of the offspring. DES, an estrogenic compound, is reported to induce anatomical malformations and other anomalies (cryptorchidism, hyperplastic testes, epididymal cysts, hypoplastic penis, meatal stenosis, hypertrophy, retained Mullerian remnants, and poor semen quality) (Stillman 1982; Gill et al. 1979; Bibbo et al. 1977; Perez-Herrera et al. 2008). A dose-dependant effect was observed for exposure before week 11 of gestation in human studies (Wilcox et al. 1995; Leary et al. 1983). However, data on infertility and sperm quality are not conclusive (Toppari et al. 1996a, b).

It has been reported that cryptorchidism is associated with testicular cancer. It has also been observed that the incidence of cryptorchidism has been on the rise in humans following exposure to DES. However, no definitive evidence for an increase in the risk of testicular cancer after exposure to DES has been established (Damgaared et al. 2002). Sakkebaek et al. (2001) and Toppari et al. (1996a, b) have suggested that cryptorchidism, hypospadias, semen quality, and testicular cancer may be interrelated. These researchers have indicated that these adverse health effects have common etiological risk factors and perhaps these adverse health effects including TDS and testicular cancer have common and shared etiology. Although it is believed TDS may have genetic etiology, the rapid increase in the rates of TDS suggests that environmental or lifestyle factors may play a role.

Hypospadias and cryptorchidism are believed to be of fetal origin, and cryptorchidism is also related to hypospadias and other complex urogenital disorders (Svenson et al. 1979). A number of epidemiological studies have reported of an association between cryptorchidism and/or hypospadias and testicular cancer (Giwecman et al. 1989; Pinczowski et al. 1991; Moller and Skakkebaek 1997; Depue et al. 1986). Jacobsen et al. (2000) have reported that men with fertility problems are more likely to develop testicular cancer and men with testicular cancer have significantly fewer children than do controls prior to development of tumor. A significant decrease in sperm concentration (113 million/ml to 66 million/ml) during a fifty-year period from 1938 to 1990 has been reported by Carlsen et al. (1992). Reanalyses of the same data taking into consideration the confounders, Swan et al. (1997) reported an even more significant decrease in sperm counts. Several other studies have found similar trends in semen quality over time with geographical differences (Toppari et al. 1996a, b; Andersen et al. 2000; Jorgensen et al. 2001). Jorgensen et al. (2001) have also reported of the regional differences (between Denmark, Finland, France, and Scotland) and seasonal variations in semen quality in healthy men in these countries.

Testicular cancer affects younger men, and in the last 40–50 years, the incidence of germ cell tumors has increased in many countries. The incidence in Denmark is among the highest in the world, and other countries which had lower incidence before are now reporting higher incidences (Finland, USA, and Scotland) (Toppari et al. 1996a, b; Moller et al. 2001; Adami et al. 1994; Boyle et al. 1987; Brown et al. 1986a, b).

Mechanism of Action of Endocrine Disrupting Substances

EDS are mimics of natural hormones and, therefore, copy the natural hormones in their modes of action, transport, and storage, but deviate from function and outcome. EDS behave similar to natural hormones and share receptors and enzymes involved in the synthesis, release, and degradation of hormones. The nuclear hormone receptors are a superfamily of transcription factors and play a significant role in physiological function and disease development. Mechanism of action of only a few of the nuclear receptors has been elucidated and among these are estrogen receptors α and β (ERα and ERβ), AR, progestin receptor (PR), and thyroid hormone receptors. A number of natural and synthetic estrogenic molecules (estrogenic ligands) bind to these receptors and activate or deactivate them. The main function of ER is as a DNA-binding transcription factor that regulates gene expression and subsequent downstream response. Although natural hormones and synthetic hormones bind to the same receptors, the downstream effects are vastly different.

It has been described in some reports that EDS elicit biphasic dose response. These responses are U-shaped and in some cases inverted U-shaped non-monotonic dose responses at low doses and have been described for several EDS (Connoly and Lutz 2004). A number of studies have explained non-monotonic responses as caused by downregulation of receptors at higher hormone levels (Medlock et al. 1991; Tibbetts et al. 1998). It has also been suggested that non-monotonic curves are generated by the integration of two or more monotonic dose–response curves that occur through different pathways with opposing effects (Vandenberg et al. 2009; Soto et al. 1995).

Dolinoy et al. (2007) have reported that the effects of exposure are dependent not only on the type of the chemical and the dose, but also on the time of exposure during life cycle. In utero developmental period is acritically sensitive period of vulnerability. Alterations induced by exposures during this time frame can lead to functional changes that may appear later in life (Yaoi et al. 2008) and contribute to increased risks of adult diseases (Heindel and Newbold 2009). The in utero exposures may act alone or in concert with other environmental exposures. It is believed that among the alterations induced by in utero exposures include changes in gene expression and protein activity and the effects are transgenerational (Schug et al. 2011).

The phenotypic changes induced later in life from prenatal and postnatal exposures are reported to be through epigenetic mechanism, a process that alters the expression of genes and DNA function without altering the DNA sequence. While epigenetic changes may be induced through histone modification, regulation of small noncoding RNAs, and DNA methylation; the DNA methylation changes are believed to be important and significant because of its ability to alter gene function. Environmental EDS are believed to act through DNA methylation pathway which takes place at the carbon-5 position of cytosine in CpG repeats (Klose and Bird 2006). During development, the epigenome cycles through a series of methylation changes and a series of checks and balances are designed to protect the integrity of DNA during these methylation and demethylation cycles. However, during this phase, periodic demethylation and remethylation of the DNA are critically vulnerable to compromised DNA methylation from exposures to environmental EDS. DNA methylation by environmental chemicals can induce epigenetic alterations in both somatic and germ cells leading to functional changes and diseases later in life (Murphy and Jirtle 2003). The mechanism by which EDS alter the epigenome has not been well elucidated and so is the enzymatic machinery responsible for demethylation. Some studies report that activation-induced cytidine deaminase may play a role in DNA methylation in early embryo and in disrupting the action of DNA methyltransferase (Maul and Gearhart 2010).

EDS are believed to inhibit androgen biosynthesis through disruption of LH signaling and suppression of Leydig cell androgen biosynthesis. BPA and octylphenol address LH receptor in Leydig cells and block the formation of cAMP and reduce steroidogenesis (Nikula et al. 1999). Natural androgen is critical to Leydig cell development; there are receptors for androgen and LH in Leydig cells. Binding of LH to receptors in Leydig cells leads to synthesis of cyclic adenosine 3′,5′-monophosphate (cAMP), which initiates a series of events that moves cholesterol into the mitochondria and causes increased steroidogenic enzyme activity and is facilitated by steroidogenic acute regulatory protein (StAR) (Akingbemi and Hardy 2001). Dimethoate blocks transcription of StAR gene in Leydig cells, preventing the movement of cholesterol into mitochondria (Walsh et al. 2000). Other EDS are also reported to inhibit steroidogenic enzyme activity.

Maternal exposure to octylphenol reduces 17α-hydroxylase mRNA and protein levels in Leydig cells, while dimethoate and HPTE reduces P450sec enzyme activity (Walsh et al. 2000; Akingbemi et al. 2000). Animal experiments have shown that exposure to DEHP in rats caused a decline in activity affecting all androgen biosynthetic enzymes (Akingbemi et al. 2001). Sertoli cells are also androgen dependent although the development and maturation of fetal Sertoli cells is stimulated by FSH from the pituitary. The total number of Sertoli cells determines the testicular size and sperm production capacity in adulthood, and Sertoli cells are unable to divide into adult testes. Therefore, a decrease in testicular testosterone reduces the rate of sperm production and promotes germ cell apoptosis.

The Impact of Estrogenic Substances and Estrogenic Metabolites on Male Health

In the past five decades or so, there has been an explosion of chemical compounds that has entered the market for use in industry, trade, and commerce. The chemical compounds have also permeated household cleaning and other products and also personal care products. It is believed that currently about 400 million tons of chemicals are used in commerce in the European Union for industrial household and personal care products (Giwercman et al. 2011). Some of these compounds imitate natural hormones and have the ability to interact with the human endocrine system and disturb normal homeostasis and physiological functioning of the endocrine system. There are a number of compounds with endocrine disrupting capabilities, and they have the potential to alter the functioning of the endocrine system (Giwercman et al. 2011). Most of the EDCs are compounds exhibit estrogenic or antiestrogenic properties and alter the function of natural estrogen and testosterone (Table 8.1).

Many of the xenoestrogens alter gene expression and cellular function by binding to steroid hormone receptors. These ER agonists include pesticides (methoxychlor, dimethoate, chlordane), industrial pollutants (polychlorinated bisphenols, alkylphenols and bispenol A), chemicals used in intensive farming (methoxychlor, 2, 2-bis(p-hydroxyphenyl)-1,1,1-trichloroethane (HPTE)), chemicals used in commerce (alkylphenols) and pharmaceuticals (antibiotics, ethinyl estradiol), and dietary items (genistine, soy, and other phytoestrogens) (Shiupnik et al. 1996). These types of EDCs act through ligand–receptor interactions; however, there are other EDCs that do not bind to receptors but alter hormone-stimulated events and are therefore identified as antiandrogens and antiestrogens. These types of EDCs include vinclozolin and phthalates among others. These substances affect androgen-stimulated male reproductive function, although there is limited information on their mechanism of action (Mylchreest et al. 1999, 2000; Parks et al. 2000; Gomes et al. 2012).

Two subtypes of ER have been reported ERα and ERβ; the former is predominantly expressed in the uterine tissue, whereas the latter is widely distributed and has been identified in central nervous system, cardiovascular system, the immune system, the urogenital tract, the gastrointestinal tract, the kidneys, and the lung. The ER subtypes are abundant in regions of the brain associated with male sexual behavior. Experiments with mice deficient in the ERα gene do not display the normal profile of masculine sex behavior (Ogawa et al. 2000). Androgen action is mediated by AR expressed in a number of tissues and has high binding efficiency for testosterone and dihydrotestosterone (Akingbemi and Hardy 2001). The ERs and the AR play an important role in the hypothalamic–pituitary–gonadal axis and the reproductive tract and are also expressed in the testis, efferent ductules, and epididymis in humans (Shugrue et al. 1998; Enmark et al. 1997; Fisher et al. 1997).

Hormone–receptor complexes formed by binding of natural and synthetic hormones with ERs and ARs bind to hormone response elements on the DNA and activate gene transcription. Synthetic hormones and particularly antiestrogens and antiandrogens suppress ER- and AR-induced natural transcriptional activity. Disruption of ER- and AR-mediated gene expression significantly affects sexual differentiation and function in the males (Carson-Jurica et al. 1990). Endogenous hormones and exogenous hormones act as ligands and upon binding through dissimilar conformations and transcriptional activity produce different pattern of gene activation (Katzenellenbogen 1996; Brzozowski et al. 1993; Pilat et al. 1993).

Some EDCs such as PCB metabolites and lindane do not bind or activate ERs but alter the metabolism of endogenous estradiol, thereby increasing the estrogen content of reproductive tissues (Kester et al. 2000; Bradlow et al. 1996). Antiandrogens such as vinclozolin form two metabolites in vivo that block AR DNA binding and so block AR activation by endogenous androgen (Blok et al. 1992). Some chemicals are reported to interact with both ER and AR in vitro. These chemicals such as DES, PCBs, o,p-DDT, HPTE, BPA, and butyl benzyl phthalate activate ER while blocking AR function (Maness et al. 1998; Sohoni et al. 1998). As a result, the cascade of event that would have occurred in genuine gene activation is altered leading to significant changes in the manifestation reproductive function, fertility, and characterization of secondary sexual characteristics. EDs, therefore, through interaction with ER and AR affect sexual differentiation, neuroendocrine physiology, androgen biosynthesis, spermatogenesis, and the incidence of testicular carcinogenesis.

EDS are ubiquitous for these substances have been used in pharmaceuticals, in agriculture as pesticides, in industry as additives and for their specific activities, in household and personal care products for their antibacterial and antiseptic properties, and in commerce for packaging food and as food items as in natural products (soy). These chemicals are lipophilic, persistent, bioaccumulate, and interfere with hormonal receptors and alter transcription of hormone-responsive genes (Kelce et al. 1995). EDCs interact with hormonal receptors through a ligand–receptor binding process, but it has also been shown that EDC action can be mediated through a membrane-associated or intracellular estrogen receptor; therefore, both interactions need to be considered (Silva et al. 2010). Experimental animal data indicate that EDCs have the potential to adversely affect the male reproductive system, but human data are still inconclusive. It is also to be noted that endogenous hormones are more potent compared to their synthetic counterparts; therefore, the response from different EDC has been wild and unpredictable (Rajapakse et al. 2001).

Availability of appropriate uterine conditions and the presence and functionality of appropriate hormones are essential for the development of the male and the female reproductive tract during fetal life. In males, FSH stimulates primary spermatocytes and is essential for spermatogenesis. Follitropin along with FSH enhances cell multiplication and production of Mullerian-inhibiting enzyme, which leads to the regression of Mullerian duct (Kuroda et al. 1990). FSH also stimulates the production of androgen-binding protein by Sertoli cells which is necessary for the spermatogenesis; FSH is inhibited by estrogen leading to a decrease in Sertoli cells which causes a decline in sperm output. The reduction in Sertoli cells also leads to reduced testicular size and sperm quantity and sperm count (Sharp et al.

1993). Mullerian-inhibiting enzyme is believed to be responsible for suppressing multiplication of germ cells during fetal life; abnormal germ cells cause testicular cancers in later life (Skakkebaek et al. 1987). Cryptorchidism is a risk factor for testicular cancer, and both have a common etiology (Moss et al. 1986). Experimental animals when exposed to ethinyl estradiol increase the risk for gonadal dysgenesis, cryptorchidism, and testicular cancer in the offspring; it also impaired Leydig cells and reduced the numbers of Sertoli cells (Yasuda et al. 1985a, b; Walker et al. 1990).

Estrogenic exposures during pregnancy impedes testosterone production through the impairment of Leydig cells, which is necessary for masculinization of the male embryo and development of external genitalia and testicular descent, and deviation from this path may lead to cryptorchidism and hypospadias (Hutson et al. 1990). It has been reported that alteration of Sertoli cells in early life has impact on testicular size and sperm output (Sharp et al. 1993). Testicular descent is a two-stage process in which the testes descend in the lower part of the abdomen and later on during gestation move through the inguinal canal to the scrotum. Both stages are regulated by Leydig cell hormones, insulin-like factor (INSL3) and androgens. In cryptorchid infants, mutations have been reported in these genes; these findings are supported by studies with experimental animals (Gorlov et al. 2002; Adham et al. 2004).

Epidemiology of Male Adverse Reproductive Health Effects

Human exposure to EDCs occurs through different routes including inhalation, gastric, dermal, and use of personal products; however, the biggest source of exposure is dietary for all ages. Gestational exposure occurs from maternal exposure, infants are exposed through milk ingestion, and adults are exposed through consumption of phyto-estrogens (Tas et al. 1995). Humans are also exposed through different environmental sources for these compounds are persistent and accumulated in the adipose tissues of animals and humans. It has been reported that three quarters of DDT in human milk are in the form of p,p-DDE metabolite and levels are higher in contaminated areas (Simonich et al. 1995). The EDCs by binding to steroid receptors and/or preventing activation of the receptor-mediated pathways or by inderectly decreasing testosterone production by Leydig cells alter normal reproductive function in human males (Akingbemi and Hardy 2001).

Endogenous hormones are important for normal sexual development in utero as well as postnatally. Endogenous estrogens are responsible for arrest of fetal germ cells which then remain dormant until puberty, when pituitary gonadotropins stimulate proliferation (Henderson et al. 1979. However, fetal exposures to exogenous estrogens interfere with the normal process and adversely affect fetal development and manifestation of secondary sexual characteristics during puberty. An increasing number of chemicals are reported to have endocrine disrupting capability, but many of the man-made chemicals have not been adequately tested (Colborn et al. 1993).

Synthetic estrogens are more potent than endogenous hormones because their binding efficiency is better. Synthetic estrogens with estrogenic and antiandrogenic properties are believed to cause demasculinization and feminization of male fish. These chemicals also interfere with systems other than the reproductive tract, thyroid gland, and the adrenal gland. The ligand binds to a number of different receptors including estrogens, androgen, progesterone, thyroxin, glucocorticoid, and the aromatic (aryl) hydrocarbon receptor. It also disrupts the expression of the steroidogenic acute regulatory protein (StAR).

The prevalence of adverse reproductive health outcomes, in most of the developed countries, has been increasing in the past two decades. This increasing trend in adverse reproductive health effects in the males has been shown consistently by several studies reporting on sperm counts, seamen quality, cryptorchidism, hypospadias, and testicular cancer (Main et al. 2010; Jacobsen et al. 2006; Carlsen et al. 1993; Swan et al. 1997). During this time period, it has also been reported that testosterone levels in adult men have also decreased (Travison et al. 2007; Andersson et al. 2007). TDS a condition in which some male adverse reproductive outcomes are believed to be associated and originate from altered testicular development in utero. The TDS development is based on the understanding that genital abnormalities in newborn male babies and adult adverse reproductive outcomes are linked (Skakkebaek et al. 2001). Testicular descent normally occurs during the fetal development in the last few weeks of gestation. This disorder is most common and affects about 2–9 % of boys (Boisen et al. 2004; Kaleva and Toppari 2005). Attributes of the data for the incidence of cryptorchidism suggest involvement of environmental influences (Boisen et al. 2004; Kaleva et al. 2005).

The mechanism of action and etiological relationship for a given risk factor remains to be elucidated, but an etiological relationship between exposure to hormonally active EDC and adverse reproductive health outcomes has been described (Jacobsen et al. 2006). A number of EDCs are believed to ubiquitously present in the environment and commonly used in trade and commerce. EDC with estrogenic and antiandrogenic properties are reported to adversely affect male reproductive system. Geographic variations in male reproductive health outcomes have also been reported. In Denmark, the prevalence of cryptorchidism is 9 % compared to that in other Western European countries where comparable rates are between 3 and 5 % (Pierik et al. 2005; Berkowitz et al. 1993; Thong et al. 1998; Preiksa et al. 2005). It is believed that geographic differences in the prevalence of adverse reproductive health effects are due to environmental exposures and lifestyle and dietary habits. However, the comparison of prevalence rates in different countries needs to be done with caution because of the differences in diagnosis guidelines for certain conditions (Boisen et al. 2004, 2005; Virtanen et al. 2001).

Persistent organohalogen pollutants such as polychlorinated biphenyls, polychlorinated dibenzofurans, and p,p-dichlorodiphenyltrichloroethane have been widely studied for their estrogenic activity. In 1979, in Taiwan when rice oil was contaminated with PCBs and PCDFs, it was observed to cause abnormal sperm morphology and decreased sperm motility (Guo et al. 2000, 2004). However, it has been also reported that exposure to POP has been negatively associated with sperm motility (Richtoff et al. 2003; Hauser et al. 2003). It has been reported that exposure to phthalates interferes with Leydig cell function, thereby altering the intratesticular levels of testosterone (Svechnikov et al. 2008). Different reports have inconsistently suggested that phthalates adversely affect sperm motility as well as sperm concentration (Duty et al. 2003, 2004; Hauser et al. 2005; Jonsson et al. 2005). Some scientific reports also suggest that phthalates may be associated with the decrease in the anogenital distance in male infants (Swan et al. 2005).

A number of researchers have reported that the prevalence of cryptorchidism and hypospadias is significantly higher in Denmark than in Finland (Boisen et al. 2004, 2005; Virtanen et al. 2001). It has also been reported that in infancy Danish boys had lower testes volume, delayed testes growth, and lower sperm inhibin B concentrations compared to Finnish boys (Main et al. 2006a, b, c). The lower testes function in infancy from in utero exposures may have played a role in lower sperm counts. In support thereof, it was observed that Finnish men had bigger testes with higher serum inhibin B, luteinizing hormone and testosterone levels and better semen quality than comparable Danish men (Jorgensen et al. 2002). Population-based studies have also suggested an increase in the prevalence of cryptorchidism in UK (John Radcliffe Hospital Cryptorchidism 1992), Denmark (Boisen et al. 2004), Lithuania, and the Netherland (Pierik et al. 2005; Preiksa et al. 2005).

Sexual Differentiation

The period of embryonic sexual differentiation is a vulnerable time for interference by ED, for these chemicals are believed to cross the placenta and reach the embryo and through breast feeding reach the infant to adversely affect the development (Bern et al. 1992). The timing of exposure to exogenous agents is crucial and critical especially to the male embryo because the expression of steroidogenic factor-1 (SF-1) and dosage-sensitive sex reversal adrenal hypoplasia, adrenal hypoplasia critical region on chromosome X, gene 1 (DAX1) responsible for differentiation of steroidogenic tissue is inhibited by estrogenic substances both endogenously and exogenously, leading to reductions in SF-1 mRNA levels in a time sensitive manner (Majdic et al. 1997). This results into indifferential differentiation of the gonads and disruption of the normal development of the testis (Fisher et al. 1999). Similarly during development, androgens facilitate but estrogen antagonize the action of Mullerian-inhibiting substances, secreted by Sertoli cells, with the result that the Mullerian ducts do not regress adequately (Visser et al. 1998).

Studies with experimental animals have suggested the exposure of male fetuses to higher levels of estrogens during gestation affects differentiation and development of sexual organs. Higher estrogen levels are reported to have smaller seminal vesicles and enlarged prostates (Nonneman et al. 1992). Perinatal exposure to antiandrogens (vinclozolin, p,p-DDE, DEHP) is reported to reduce androgen biosynthesis, decrease anogenital distance, and delay the onset of preputial separation. Studies with experimental animals also suggest higher incidence of hypospadias, ectopic testes, vaginal pouch formation, and nipple retention in exposed rats compared to controls (Mylchreest et al. 1999; Parks et al. 2000; You et al. 1998; Gray et al. 2000; Akingbemi et al. 2001; Wolf et al. 2000). Testosterone is required in fetal life for the development of urogenital system and in adult life for proper functioning of the reproductive system. During these phases, exposure to EDS affects Leydig cell function which disrupts aspects of male reproduction (Abney et al. 1991; Shan et al. 1992). Exposure to EDS that increase estrogenicity or decrease androgenicity of Leydig cells or AR-mediated activity may interfere with many aspects of male phenotype.

Semen Quality and Sperm Concentration

The treatment of pregnant women with DES for pregnancy-related events between 1945 and 1971 led to an increase in the incidence of male reproductive adverse health effects (decreased sperm counts, semen abnormalities cryptorchidism, hypospadias, testicular hypoplasia, and others) (Jensen et al. 1995; Gill et al. 1977, 1979). Similar observations have been made by several other researchers (McLachlan et al. 1981; Whitehead and Leiter 1981). However, recent studies seem to indicate no association between exposure to estrogens in the first trimester and external genital abnormalities (Driscoll and Taylor 1980; Beard et al. 1984; Raman-Wilms et al. 1995). Other scientific studies have reported that sperm concentration, sperm with normal morphology, and sperm quality were all statistically lower in men exposed to DES (Gill et al. 1979; Stenchever et al. 1981), but Andonian and Kessler (1976) were unable to reproduce these findings. Similarly, evidence on the increased risk of testicular cancer in men exposed to DES did not stand statistical scrutiny (Brown et al. 1986a, b; Vessey 1989; Henderson et al. 1979; Depue et al. 1983). Although biologically plausible, the evidence of adverse effects on semen quality and sperm counts remains to be conclusive.

It was Sharpe and Skakkebaek (1993) who first suggested that increased exposure to estrogens in utero was associated with the increasing incidence of male reproductive health abnormalities. It was also suggested around that time that over the past five decades, the sperm counts have fallen by 40 % (Carlsen et al. 1992). However, Fisch et al. (1986) have reported that no significant changes in sperm count or sperm quality were observed among patients at vasectomy clinics in North America. It was also noted that sperm counts from different locations were highly variable, and this has been supported by data from various regions in the USA, Canada, and Nordic–Baltic countries (Jorgensen et al. 2002; Auger and Jouannet 1997; Swan et al. 2003; Younglai 1986). Studies in the general population of young men in Sweden showed no association between sperm counts and PCB exposure (Richtoff et al. 2003). A study among Danish men indicated that men with high or low body mass index had poorer semen quality (Jensen et al. 2004). Therefore, the cause of variability in semen quality and sperm quantity remains to be conclusively for environmental and occupational exposures to estrogens or estrogenic substances and metabolites.

A possible but not significant decline in human semen quality during the last 50–60 years has been reported by Nelson and Bunge (1974) and Bostofte et al. (1983). However, Carlsen et al. (1992) reported a substantial decrease in sperm concentrations among Caucasian men. A French study by Auger et al. (1995) reported a decrease of 2 % in sperm concentration per year. The percentages of motile and normal spermatozoa also decreased substantially. A Belgian study also reported decreased sperm count and motility (van Waeleghem et al. 1994). Overall, it appears that there is a geographic distribution in semen quality and sperm concentration with some countries in Europe having lower values compared to others (Suominen et al. 1993).

Cryptorchidism and Hypospadias

It has been hypothesized that environmental exposures to estrogens and estrogenic chemicals may be associated with increased incidence of cryptorchidism and hypospadias which contribute to increase in problems of the reproductive tract in the males (Sharpe et al. 1993; Skakkebaek et al. (2001). The trends for the incidence of cryptorchidism have been increasing for the past several decades. Although ethnicity has been suggested as a risk factor, the data are not convincing. Heinonen et al. (1977) reported statistically significantly higher rates among whites compared to blacks in the USA, but Berkowitz et al. (1993) could not find the rates statistically significantly different among the blacks and the whites (Heinonen et al. 1977; Berkowitz et al. 1993).

Analysis of the Finnish hospital discharge data from 1970 to 1994 suggested that the incidence of hypospadias has not changed and so also rates of surgical treatment of cryptorchidism in the UK from 1992 to 1998 (Aho et al. 2000; Toledano et al. 2003). A collaborative Finnish–Dannish study has shown that the cryptorchidism rates are higher in Denmark than in Finland (Boisen et al. 2004). These findings suggest that cryptorchidism rates vary by geographic regions and demographic profiles, and these variations are potentially due to environmental factors (Rochleau et al. 2009).

The trends in the incidence of hypospadias have been reported to be increasing in England and Wales (Matlai and Beral 1985); Hungary (Czweizel et al. 1985); Sweden (Kallen and Winberg 1982); Norway, Denmark, Finland, Spain, New Zealand, Australia, and Czechoslovakia (Leung et al. 1985); and the USA (Finlay et al. 1984). In some countries in Europe, the incidence rates are higher than others (Denmark higher than Norway), and in the USA, the rates among the whites are higher than the rates among the blacks (Jensen et al. 1995). It has also been reported that the prevalence of hypospadias worldwide during the past three decades has been on the increase (Safe 2000; Handelsman 2001). In the USA, the prevalence has increased from 20 to 40/10,000 live births from 1986 to 1993 (Ahlborg et al. 1995). Hypospadias has been also associated with exposure to pesticides, and

Lopez-Cervantes (2004) has reported that the risk of hypospadias was 1.4 (95 % CI 1.06–1.84) from maternal occupational exposure and a risk of 1.2 (95 % CI 1.03–1.47) for paternal occupational exposure. Similar results have been reported by Carbone et al. (2006) by indicating that higher pesticide usage was significantly associated with higher prevalence of hypospadias.

However, a study with a birth cohort from Denmark by Zhu et al. (2006) reported that no increased risk of hypospadias was observed in farmers and gardeners. In a similar manner, researchers from the UK having studied chemicals such as pesticides, phthalates, alkyl phenols, and heavy metals from 1980 to 1996 reported that there is no increased risk of hypospadias in offspring of mothers with occupational exposure to these chemicals (Vrijheid et al. 2003). McGlynn et al. (2009) studied PCBs in serum of pregnant women and reported that there were no increased odds between maternal body burden of PCB and hypospadias or cryptorchidism in their offspring. But Ormond et al. (2009) studied 471 cases of hypospadias and reported a 2.4-fold increased odds (95 % CI 1.40–4.17) of hypospadias in male offspring of women with occupational exposure to hair spray. Similarly, vegetarianism during pregnancy increased by more than fourfold the odds ratio (95 % CI 1.81–12.84) of hypospadias compared to non-vegetarianism (North et al. 2000). Maternal diet without fish or meat had a 4.6-fold odds ratio (95 % CI 1.56–13.0) increased the risk for hypospadias in offspring (Akre et al. 2008).

A number of other risk factors of hypospadias have been identified including small gestational age (Pierik et al. 2004); buying market fruits as opposed to homegrown fruits increased the risk fivefold (95 % CI 1.31–19.82) (Giordano et al. 2008); eating fish more than once a week increased the risk twofold (95 % CI 1.03–5.31) (Rejinders et al. 1986); and oral contraceptives during early pregnancy increased the risk 1.7-fold (Norgaard et al. 2009). Exposure to DES during pregnancy has been well studied and reported. The risk of hypospadias among sons of women who had been exposed to DES during gestation was 21 times higher compared to the unexposed (Klip et al. 2002). However, not all studies reported of a positive relationship for Palmer et al. (2009) were unable to document any reproductive adverse health effects in a cohort of women with prenatal exposure to DES.

In recent years, the prevalence of cryptorchidism has been on the increase in Western Europe including the UK (Acerini et al. 2009). Among other estrogenic exposures positively associated with cryptorchidism are maternal exposure to DES and Damgaard et al. (2008) reported a risk ratio of 1.9 (95 % CI 1.1–3.4). However, Toledano et al. (2003) found that there was no increase in the prevalence of hypospadias. Although the relationship between environmental exposures to ED remains to be conclusive, TDS appears to be linked to hypospadias, cryptorchidism, poor semen quality, and testicular cancer and all are associated with exposure to endocrine disruption during fetal development (Skakkebaek et al. 2001).

Data from a prospective cohort study in Finland and Denmark purport to support the observation that TDS is caused by environmental exposures to ED and are also associated with testicular cancer, cryptorchidism, hypospadias, and semen quality (Wohlfahrt et al. 2009). However, Akre and Richardi (2009) and Martin et al. (2008) did not find any relationship between exposure to environmental estrogens and TDS, cryptorchidism, hypospadias, and testicular cancer, although there were some indications of a slightly increased risk for these conditions. This type of contradicting data from different studies makes the verdict on the evidence inconclusive. It is therefore necessary to examine the biological plausibility and conduct further research using both animal and human tissues and obtain definitive data that may help to provide conclusive evidence.

Testicular Cancer

The incidence of testicular cancer is highest in Western and Northern Europe, North America, and Australia, while lower rates are reported for eastern Asia and Africa (Rosen et al. 2011). In the USA, the incidence has increased by 44 % from 1973 to 1998, and similar trends have been reported for Europe, Canada, South America, Israel, and Australia (McGlynn et al. 2003; Chia et al. 2010). It is believed that the increasing incidence rates are likely due to environmental exposure to testicular toxicants. Other factors associated with increased risk are cryptorchidism, subfertility, and first-degree male (father or brother) relative and genetics; however, environmental exposures are one of the most significant risk factors (Meeks et al. 2012; Pharris-Ciurej et al. 1999; Linke et al. 1959; Fossa et al. 2005; Hemminki and Li 2004; Greene et al. 2010). It is perceived that Leydig cells may be particularly sensitive to estrogenic substances causing hormonal stimulation from environmental ED (Safe 2002). The most susceptible period of exposure is believed to be during intrauterine development.

The incidence of testicular cancer is highest in younger men (20–40 years old), and the etiology of this disease appears to be associated with exposure to estrogens in utero (Depue et al. 1983). Studies with dizygotic and monozygotic twins lend support to the observations that increasing incidence of testicular cancer is associated with increasing exposure to estrogens and estrogenic chemicals (Swerdlow et al. 1997; Adami et al. 1994). These observations are also supported by those studies that provide evidence of increased incidence of testicular cancer in men exposed to DES in utero (Strohsnitter et al. 2001). The incidence rates of testicular cancer vary by geographic regions, particularly in Western Europe. A greater than fourfold difference in testicular cancer has been reported between Denmark and Finland, but the factors associated with these observations have not been characterized in the literature, and the causal association remains elusive (Boisen et al. 2004; Moeller 2001).

The trend for the incidence of testicular cancer appears to be increasing for the past several decades (Forman and Moller 1994). Increasing incidence for testicular cancer has been reported in several countries including England and Wales (Pike et al. 1987); Scotland (Boyle et al. 1987); the Nordic countries (Adami et al. 1994); Australia (Stone et al. 1991); New Zealand (Pearce et al. 1987); and the USA (Brown et al. 1986a, b). A 2–4 % increase has been observed in men between the ages of 20–45 in all these countries (Jensen et al. 1995). It has been suggested that the increase in testicular cancer rates are associated with the increased exposure to estrogens and estrogenic metabolites; however, the direct positive relationship remains to be convincingly elucidated. But in the interim cryptorchidism, hypospadias and testicular cancer share a common etiology and exposure to estrogen and estrogenic substances appears to play a role.

Testicular Dysgenesis Syndrome (TDS)

A number of researchers have reported on male adverse reproductive health effects including declining sperm counts and semen quality, increasing incidence of cryptorchidism and hypospadias, and testicular cancer as has been reported earlier (Adami et al. 1994; Carlsen et al. 1992; Swan et al. 1997; Toppari et al. 1996a, b; Paulozzi et al. 1997). It has been also proposed that these disorders represent different symptoms of the same underlying condition called TDS and have a common fetal origin (Skakkebaek et al. 2001; Sharpe 2003). Clinical evidence for TDS in males comes from observations that patients with rare genetic disorder that causes TDS are associated with high risk of testicular cancer often combined with cryptorchidism and hypospadias (Scully 1981; Savage and Lowe 1990). Additionally in patients with testicular cancer, subfertility, or cryptorchidism, dysgenetic changes have been found in the testis (Huff et al. 1993; Hoei-Hansen et al. 2003; Skakkebaek et al. 2003). Experimental animals exposed to EDC have been reported to exhibit similar reproductive disorders, which in humans have been linked to TDS (Skakkebaek et al. 2001; Damgaared et al. 2002; Fisher 2004).

Fertility Issues in Males

Problems associated with the male reproductive health can adversely affect the fertility in males. It has been reported that exposure of males to estrogenic chemicals in utero did not affect fertility in males with no deformities in the external genitalia. The potential effects of in utero exposure to DES were examined in male offspring of mothers who were exposed to DES did not show any decreased fertility (Wilcox et al. 1995). Similar findings are reported by a study among sons of Finnish mothers who were administered pharmacological dose of estrogens and progestins during pregnancy (Hemminki et al. 1999). Studies in Sweden which examined the effects of consumption of fish contaminated with organochlorines observed that there was no association between exposure to organochlorines and fertility (Axmon et al. 2001, 2002).

Adverse Reproductive Health Effects in Male Wildlife and Experimental Animals

Effects Observed in Male Wildlife

Alterations in the male reproductive health in the wildlife include feminization and demasculinization of males, reduced fertility, reduced hatchability, reduced viability of offspring, impaired hormone secretion (activity), and altered sexual behavior (Colborn et al. 1993). Exposure of alligators in Lake Apopka to estrogenic compound (DDT) which metabolizes into a potent androgenic compound (DDE) was associated with demasculinization and reproductive failure (Guillette et al. 1994a, b; Crain et al. 1997). Similar observations have been made in male panthers perinatally exposed to estrogenic substances. Observations of egg shell thinning in different bird species and reproductive problems in Polar bears suggest that pollution with persistent ED is a problem for birds, fish, and mammals. But as has been reported earlier, exposure to EDC has also been a problem in human populations for adverse reproductive and other adverse health effects have been reported.

Ecological studies with the wildlife and environmental pollution and degradation have identified a number of adverse reproductive health effects, especially in the male population. These adverse health effects in the wildlife have been linked to EDC in the environment (Toppari et al. 1996a, b). Hormone-dependent organs in mammals, birds, and fish are observed to be at risk for developmental disorders in offspring because of maternal exposure. These organs include mammary glands, fallopian tubes, cervix, uterus, and vagina in the females and the prostate, seminal vesicles, epididymides, and the testes in males. Organs affected in both sexes include the external genitalia, brain, skeleton, thyroid gland, liver, kidney, and the immune system (Petersen et al. 1993; Colby et al. 1980; McEwan et al. 1981; Leatherland and Sonstegard 1982; Grossman 1984). The effects observed in the males include undescended testes and decreased semen quality in wild panthers (Facemire et al. 1995) and smaller or undeveloped genitalia in alligators (Guillette et al. 1994a, b; 1995).

A number of adverse effects observed in the male wildlife have been associated with exposure to EDC. Abnormal thyroid function has been reported in birds and fish (Moccia et al. 1981, 1986); decreased fertility in birds (Shugart 1980) and fish (Leatherland 1992); decreased hatching success in fish (Mac et al. 1988) and birds (Kubiak et al. 1989); and demasculinization and feminization of male fish (Munkittrick et al. 1991), birds (Fry and Toone 1981), and mammals (Beland 1989). Effects like these have been reported in many areas where the exposure is high to a range of toxicants including industrial chemicals and pesticides (Colborn et al. 1990). These effects have also been predominantly observed in the Great Lakes area, and although much of these data come from these areas, the levels of contamination in the Great Lakes are similar to the other major drainage basins in the USA (Phillips and Birchard 1990).

Adverse Health Effects Observed in Male Experimental Animals

The male reproductive health effects induced by exposure to ED in wildlife have been reproduced in experimental animals. Male offspring exposed in utero are reported to exhibit epididymal cysts, testicular hyperplasia, cryptorchidism, hypospadias, micropenis, prostate hypertrophy, cellular atypia, retained Mullerian remnants, poor semen quality, and subfertility (Newbold and McLachlan 1985; Yasuda et al. 1985a, b; Luthra and Hutson 1989; Walker et al. 1990; Newbold 2001; Thayer et al. 2001). Sertoli cell and Leydig cell hyperplasia, interstitial testicular tumors, squamous metaplasia of the seminal vesicles, and adenocarcinoma of the testis have also been reproduced in experimental animals (Walker et al. 1990; Yasuda et al. 1988; Newbold et al. 1986).

Peripubertal male mice and rats treated with antiandrogens (vinclozolin, linuron, methoxychlor, and di-n-butyl-phthalate) have been shown to delay puberty. Exposures to low doses of antiandrogens (vinclozolin, procymidone, linuron, and p, p-DDE) are known to reduce anogenital distance and appearance of areolas. Comparable experiments with medium doses induced retained nipples and hypospadias and cryptorchidism and epididymal agenesis at higher doses (Gray et al. 2001; Viguier-Martinez et al. 1983). Other researchers have reported that exposure to phthalates induced urogenital malformations and affect reproductive function by inhibiting fetal testosterone synthesis (Shono et al. 2000; Parks et al. 2000; Foster et al. 2001).

Estrogenic Substances in the Environment

The deposition of DDT into the environment began with the widespread use of DDT in the 1940s, but regulations on the use of DDT introduced since 1972 have been effective in reducing the levels in the environment (Colborn et al. 1990). The effects of DDT contamination that were documented in birds and by Bald eagle in particular through egg shell thinning, embryonic and chick survival, and mating and hatching abnormalities are now a thing of the past. Similar effects observed in fish and wildlife have now declined, although industrial effluents, rich in ED, fed into rivers and other water bodies continue to cause hormone-related adverse health effects. Apart from DDT, other known EDs that have been identified in the environment are chlordane, dieldrin, polychlorinated biphenyls, dicofol, kelthane, and methoxychlor (Colborn et al. 1993).

In humans, cryptorchidism and hypospadias have been most reported and have been associated primarily with environmental exposures to ED, although other causes have been identified. Environmental and occupational exposures to ED are believed to be the prime suspects because these agents are commonly present in the environment, and their residues have been found in higher amounts in individuals with adverse reproductive health. Apart from environment and occupation as risk factors of cryptorchidism and hypospadias, genetic factors which include genetic mutations and polymorphisms among others have also been reported (Kalfa et al. 2009; Virtanen et al. 2007). Although EDC can manifest its activity in four different ways, in this review, estrogenic and antiandrogenic substances and their metabolites have been examined. A number of endocrine-disrupting chemicals have been detected in human reproductive tissues (Thomas 1992). These lipid-soluble compounds are reported to accumulate in fatty tissue in the body (Jensen and Slorach 1991). Although some of these substances are banned from commerce and trade, they are still present in the environment because of their persistence and bioaccumulation.

Scientific literature suggests that a range of chemicals used in agriculture, industry, personal care products, and trade and commerce are capable of binding to intracellular estrogen receptors either directly or after in situ conversions to an active metabolite. Methoxychlor is demethylated to a more estrogenic compound, and DDT, chlordane, and nonylphenol exhibit binding preference to estrogen receptors. Reviewing the lessons learnt from DES exposure and maternal exposure estrogenic chemicals, the following observations male adverse reproductive health outcomes appear to be associated with increased exposure to estrogenic chemicals: (a) increased incidence of cryptorchidism and hypospadias in the past three to four decades (Group JRHCS 1986), (b) decrease in sperm counts (Carlsen et al. 1992), and (c) increased incidence of testicular and prostatic cancer (Sharpe and Skakkebaek 1993). The effects of exposure to estrogens are not only observed during embryonic or early childhood development but also in adult men. Exposures to estrogenic substances have been implicated in the etiology of prostate hyperplasia (vom Saal et al. 1993; Ghandian 1983).

Pesticides as Estrogenic Substances

Epidemiological studies have reported on the increased risk of genital malformations in areas of intensive agriculture (Garcia-Rodrigues et al. 1996), greenhouse workers, and farmers (Pierik et al. 2004; Weidner et al. 1998; Fernandez et al. 2007). Congenital cryptorchidism is reported to be significantly positively associated with maternal exposure to organochlorine pesticides through breast feeding (Damgaard et al. 2006). Increased risk of cryptorchidism and higher than average amounts of pesticide residues were reported in mothers who worked in greenhouses (Andersen et al. 2008). These offsprings were also reported to have smaller genitalia and lower serum concentration of testosterone and inhibin B. Fernandez et al. (2007) have reported that boys with genital malformations had elevated levels of DDT and pesticides (lindane, mirex, and endosulfan). Increased risk of testicular cancer has been reported to be associated with exposure to organochlorinated pesticides, DDE, and chlordane (Hardell et al. 2003; McGlynn et al. 2008).

Chemicals Used in Industry and Commerce

Phthalates are used as plasticizers in flexible polyvinyl chloride (PVC) products, toys, consumer products (soaps, shampoos, and perfumes), and medical devices. The most commonly used phthalate is di-(2-ethylhexyl) phthalate (DEHP); the monoester form of DEHP is reported to be 10-fold more potent in its toxicity to Leydig cells and Sertoli cells compared to DEHP. Detectable levels of phthalates have been reported in 75 % of the population in the USA (Silva et al. 2004; Koo et al. 2002). An association between phthalate exposure and male genital development has been reported by Swan et al. (2005). Prenatal exposure to phthalates has been reported to induce testicular dysgenesis in experimental animals (Welsh et al. 2008). Similar observations have been made in humans, and alteration of pituitary–gonadal axis has also been reported in three-month-old boys from exposure through breast milk (Main et al. 2006a, b, c). Other researchers have reported development of hypospadias from in utero exposure to diethylhexyl phthalate (Lu et al. 2009). Reduced anogenital distance has been described in infant boys exposed prenatally to phthalates. Boys with short anogenital distance are reported to have a higher risk of cryptorchidism and hypospadias and smaller penis size (Hsieh et al. 2008). While the adverse effects of in utero exposure to phthalates in infant males appear to be conclusive, similar adverse effects in adult males are not conclusive. In adult males, the only observed effects are decreasing semen quality and alterations of hormone levels; however, these observations are not consistent (Duty et al. 2004, 2005; Murature et al. 1987; Meeker et al. 2009).

The exposure of the fetus in utero is of particular concern because it is classified as a potential developmental toxin by US administration (Borch et al. 2006; Kim et al. 2004). Phthalates are believed to have profound effects on testicular cell types such as Sertoli cells and gonocytes (David et al. 2006). These studies indicate that human male adverse health effects are possibly through the interference by phthalates with Leydig cell, and the adverse effects on development and manifestation of secondary sexual characteristics through interference with Sertoli cells (Ge et al. 2007). Leydig cells are primary source of testosterone in the male; therefore, differentiation of Leydig cells in the testes is an important event for the males. For the differentiation and characterization of male secondary characteristics, testosterone production is critical (Huhtaniemi et al. 1992). Insulin-like growth factor 3 (IGF3) produced by Leydig cells binds to the leucine-rich repeat-containing G protein-coupled receptor 8 (LGR8) in the gubernaculum and together with testosterone induces scrotal descent of testis (Scott et al. 2005; Adham et al. 2000). Therefore, interference with the development of fetal Leydig cells and alterations in testosterone productions may lead to the manifestation of cryptorchidism.

Fisher et al. (2003), Ema et al. (1998, 2000) and Mylchreest et al. (1998a, b) have suggested that in utero exposure to phthalate esters induces cryptorchidism, hypospadias, impaired spermatogenesis, and reduced male fertility in rats. In studies in human populations, it has been observed that male genital development is compromised in newborns exposed during gestation to phthalates. A reduced anogenital distance has also been reported from exposure to monobenzyl phthalates and monoisobutyl phthalates (Swan et al. 2005). Adverse effects have also been reported in adult Leydig cell development. Pups born to mothers exposed to DEHP at 100 mg/kg/day have reduced serum testosterone levels (Akingbemi et al. 2001). Neonatal exposures to phthalates via mother's milk are reported to have adverse reproductive health outcomes. Contamination of milk by phthalate monoester has an adverse influence on the postnatal surge of reproductive hormones in newborn boys (Main et al. 2006a, b, c). Pubertal exposures to phthalates are reported to produce adverse health effects which are both duration- and dose-dependent. Exposure to DEHP in experimental animals in a time- and dose-dependent manner increased testosterone and serum LH levels and in some cases testicular atrophy and inhibitory effects on Leydig cell function (Akingbemi et al. 2001, 2004).

Main et al. (2007) have reported that mothers who gave birth to boys with cryptorchidism had higher concentrations of polybrominated diphenyl ethers (PBDEs), which are used as flame retardants. PBDE congeners 47, 153, 99, 100, 28, 66, and 154 were higher in breast milk for those mothers who gave birth to sons with cryptorchidism (Main et al. 2007). It is believed that exposure to these types of substances alters testosterone:estrogen ratio in the fetus (Main et al. 2010). Other researchers have reported an association between exposure to PBDE and other persistent chemicals and cancer of the testes. Higher levels of PCBs, DDE, HCB, chlordanes, and PBDEs were found in mothers of sons with testicular cancer (Hardell et al. 2003). Other observed effects include longer waiting time to pregnancy following exposure to perfluorinated chemicals and decreased sperm counts with normal morphology and exposure to perfluoroalkyls (Fei et al. 2009; Joensen et al. 2009). Studies with biopsy samples from orchidopexy have reported higher levels of dichlorodiphenyltrichloroethane (DDE, a metabolite of DDT), toxaphene, pentachloroanisole, pentachlorobiphenyl, and chlorinated cyclodienes (Hosie et al. 2000).

Summary of Male Reproductive and Developmental Health

EDS are believed to contribute to many adverse reproductive health effects. Epidemiological data in the past fifty years appear to indicate an increasing rate of reproductive health effects that correlate with increasing amounts of EDS use (Delbes et al. 2006). EDS have been linked to (a) disrupted reproductive function (poor semen quality and infertility); (b) altered fetal development (urogenital congenital anomalies including hypospadias and cryptorchidism); and testicular germ cell cancer (Vos et al. 2000; Meeker et al. 2008). Some researchers believe that cryptorchidism, hypospadias, and poor semen quality are risk factors for and are predictive of testicular germ cell cancers; together, these conditions are described as TDS (Dupe et al. 1979; Skakkebaek et al. 2001). TDS is suggested to have developmental origin due to reduced androgen levels and has a negative impact on the Sertoli and the Leydig cells; TDS is strongly associated with environmental exposure to EDS.

With the exception of a few well-documented cases, the association between environmental EDS and adverse reproductive health has been difficult. Environmental EDS are substances that mimic natural endogenous hormones and, are therefore, able to interfere with the endocrine system and either block or alter the receptors, thereby altering the downstream effects of ligand–receptor interactions (Sikka and Wang 2008). The net results are malformations of reproductive system, hypospadias, retained nipples, and reduced anogenital distance (Foster 2006). Androgens are essential to the proper development and functioning of the Sertoli cells; EDS interference with the Sertoli cells during fetal development results in reduced fertility later in life (Sharpe 2010).

Epidemiological studies suggest that reduced semen quality is associated with exposure to phthalates, polychlorinated biphenyls (PCBs), dioxins, and organochlorinated pesticides in the North American population, but these findings are not consistent. Poor sperm motility is reported to be associated with the level of exposure to monobutyl phthalates (Duty et al. 2003) and semen quality on the timing of exposure (Mocarelli et al. 2008). Studies on occupational exposures to pesticides have also been reported to be associated with reduced semen quality (Lifeng et al. 2006; Oliva et al. 2001; Padungtod et al. 2000; Abel et al. 2000). Inverse association between urinary concentrations of pesticides and sperm concentrations and motility in men has been described by Meeker et al. (2004). Although these studies show the presence of a relationship between EDS and adverse reproductive health in human males, further studies are required to characterize the functional identity of these compounds and the mechanism of action in the induction of these adverse health effects. The traditional ligand–receptor interaction synthetic hormones and natural receptors and alterations in genes through genetic and epigenetic mechanisms need to elucidate the mechanism of action of EDCs. The fetal developmental origin of the adverse health effects in human males needs to be studied further with the development of good animal model so that irrefutable evidence can be presented.

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Chapter 9
Indigenous People of the North and the Globalized World

Frank Sowa

Abstract In an increasingly interconnected world, there are more ecological, social, cultural, political, and economic changes than ever before. These various processes, which are summarized in the social sciences under the label of "globalization," lead to a very complex connectivity. Taking the Kalaallit, the Greenlandic Inuit, as an example of indigenous people of the North, it can be stated that global interactions affect daily life in Greenland. The aim of this article is to approach these complex interactions on the basis of *Greenlandic foods*, which is called *kalaalimernit* in Greenlandic. Globalization can be interpreted as imposing a pressure on *Greenlandic foods* because of economic, political, and environmental influences. Nevertheless, globalization gives the actor "Greenland" the opportunity to become a self-confident player in the global politics of power. The global flows of ideas such as "human rights" and "democracy" enable Greenland's Self-Rule Government to push ahead a successful nation-building process. Central elements of the distinctive Greenlandic culture are the Greenlandic language and the *Greenlandic foods* of the hunting complex. The argument of this contribution is that *Greenlandic foods* are suitable to establish a distinctive group with a collective Greenlandic identity because through the consumption of this kind of foods it is possible to construct cultural boundaries: Inside the country, local foods are used to mark a separation from Danes, and outside the country, *Greenlandic foods* work as a conscious distinction from the rest of the world. Foods function as a predicament of its own for the Greenlandic culture that it is necessary as well as risky to keep on eating *kalaalimernit*.

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Introduction: Talking About Foods

The 2002 Arctic Winter Games took place from March 17 to March 22, 2002, in the Greenlandic capital Nuuk and set a new milestone in the history of the Games. Not only were the Arctic Winter Games for the first time cohosted by two Arctic communities, Iqaluit in Nunavut and Nuuk in Greenland; furthermore, this event took place for the first time in Greenland ever. The participating Arctic regions were Alaska, Alberta, Chukotka, Greenland, Magadan, the Northwest Territories, Nunavik, Nunavut, and Yukon. This outstanding *circumpolar* event was more than a sport event for young people, and it was a cultural event as well. In that way, the Arctic Winter Games attract athletes, cultural performers, and spectators from all over the Arctic to celebrate the circumpolar peoples and their Northern sports and cultures. During this week, there were a lot of shows, performances, dances, and throat singing, as well as non-artistic presentation as a harpooning contest and a sealskin flensing contest by local hunters beside the sport competitions. There was also the possibility to taste the wide range of local foods, e.g., in attending in one of the official banquettes. The *Arctic Kitchen* near the snow arena offered daily local foods for the hungry visitors and spectators in a huge snow house (what Westerners would call "igloo"). The catered *Greenlandic food* specialities were reindeer and seal meat turning this event to a unique Greenlandic event expressing Greenlandicness.

This article deals with indigenous people of the North and the globalized world. One group of indigenous peoples in this circumpolar north are the Greenlandic Inuit (Sowa 2014e), who call themselves *Kalaallit* (singular *kalaaleq*), which means Greenlanders (Sowa 2014b). From a sociological perspective, the object of investigation is the political representation of Greenlandicness in the global era by using foods with local origin. In my view, it is not a coincident that Greenlanders present their own food culture to a (world) audience during the Arctic Winter Games. For them, it is a possibility to express the uniqueness and vitality of the Inuit culture. In practicing this strategic staging of their own culture, it is possible to mark a cultural boundary in an active way. During these occasions, *foods*, which are an inconsiderate part of the everyday life of the Greenlanders, turn to *Greenlandic foods* as glorified symbol for the Greenlandic culture.

The globalized world has an impact on Greenland. In an increasingly interconnected world, the actor "Greenland" becomes the opportunity to be a self-confident player in the global politics of power. The global flows of ideas such as "human rights" and "democracy" enable Greenland Self-Rule Government to push ahead a successful nation-building process (Sowa 2013d). This requires a political representation of a distinctive Greenlandic culture whose central elements are the Greenlandic language and *Greenlandic foods* of the hunting complex. This article argues that *Greenlandic foods* are suitable to establish a distinctive group with a collective Greenlandic identity because through the consumption of this kind of foods it is possible to construct cultural boundaries: Inside the country, *Greenlandic foods* are used to mark a separation from Danes, who are living and working in Greenland. Outside the country, it works as a conscious distinction from the rest of the world. In this way, Greenlandic foods can be seen a strong marker of distinction and can be able to form a cultural group in the global context.

At the same time, globalization can be interpreted as imposing a pressure on *Greenlandic foods* because of economic, political, and environmental influences. Therefore, several global flows (Appadurai 1990) have serious impacts on the local foods in Greenland, for instance, the global flow of consumer goods that lead to economic pressure, the global flow of ideas as environmental or animal rights ethics have political pressures as a consequence, and finally, the global flow of pollution and problem situations confronted *Greenlandic foods* with environmental risks.

This article is structured as follows: In the next chapter entails a description of the motivation, fieldwork, and scientific challenges (Section "Motivation, Fieldwork and Scientific Challenges"). The following chapter focuses on globalization as imposing a pressure on *Greenlandic foods* caused by the integration of Greenland in the global net of worldwide interconnectedness and interdependency. The analysis of the *Greenlandic foods* shows that these kinds of foods are under pressure (Section "*Kalaalimernit* Under Pressure") due to a global flow of consumer goods (Section "Global Flow of Consumer Goods: Kalaalimernit and Economic Pressure"), a global flow of ideas (Section "Global Flow of Ideas: Kalaalimernit and Political Pressure"), and a global flow of pollution (Section "Global Flow of Pollution: Kalaalimernit and Environmental Risks"). However, *Greenlandic foods* are also able to function as a marker of difference in a globalized world (Section "*Kalaalimernit* in a Globalized World: Food as Marker of Difference"). *Kalaalimernit* are seen as special due to the name (Section "Name of Greenlandic Foods"), the places where they are found (Section "Places, Where Greenlandic Foods Are Found"), the quality attributed to them (Section "Quality of Greenlandic Foods"), the sign and symbol character (Section "Food as Sign and Symbol"), the distinctive cultural practices (Section "Food as Distinctive Cultural Practices"), and the connection to the Greenlandic idealized self-image (Section "Food as a Connection to the Greenlandic Idealized Self-image"). Through presenting *Greenlandic foods* to a world society, the actor "Greenland" gets more possibilities to create its own future. The conclusion is that it functions as a predicament of its own for the Greenlandic culture that it is necessary as well as risky to keep on eating *Greenlandic foods* (Section "Conclusion").

Motivation, Fieldwork, and Scientific Challenges

In the course of time, each researcher discovered probably a subject area or a research topic that inspires him or even ties him up. I have been working on the influences of processes of globalization on a local indigenous culture since the year 2000. As an example of an indigenous people of the North, I chose the Greenlandic Inuit or *Kalaallit*. All in all, during the period of writing my master thesis and my dissertation (Sowa 2000, 2014c), I travelled three times to Greenland, staying about eleven weeks in 2000 and nine weeks in 2002 for doing research, as well as another two weeks for attending a PhD course at the *Ilisimatusarfik* (University of Greenland) in 2002, 22 weeks in total. It was a long journey from getting in touch with Greenlanders, conducting qualitative face-to-face interviews, analyzing the rich data (interviews and documents), reading theories and other empirical studies, revising first interpretations and heuristics, writing thick descriptions, and putting the final results down on paper until the research diffuses in the scientific discourse (Sowa 2013a, 2014c, 2015b).

My main visited location to gather experiences and to conduct interviews was the Greenlandic capital Nuuk (Fig.9.1). Nuuk is a modern—on the first glance Danish like—city with some huge supermarkets, shops, fast-food restaurants, pubs, and bars (Sowa 2004a). The whole range of European products such as drinking yoghurt, deep frozen pizza, ready-to-serve meals, instant coffee, fresh fruits and vegetables or hot dogs, cheeseburgers, French fries, and beefsteaks is available. In a Greenlandic household, there are often a fridge, a freezer, an oven, and a microwave oven. And even the kitchen utensils are bought on a *Tupperware* party. Hence, one could say "life there is pretty much as it is in other places in the modern world" (Thomsen 1996: 265).

The second visited location was Oqaatsut, a tiny place in the Disko Bay with 50 inhabitants and 179 sledge dogs. The idea of doing fieldwork in Oqaatsut was due to the narrations of my interviewees in Nuuk who claim that the "real," "authentic" Greenlanders were living in the small villages along the coast (Sowa 2012a, b). I organized a family stay in Oqaatsut and lived there for three weeks. With the help of a Greenlandic translator, a friend of a friend, I conducted four interviews about the daily life of villagers.

The topic of *Greenlandic foods* was often a part of conversations. During my stay in Nuuk, I once had a conversation with a German natural scientist who lived in Greenland for years. We talked about the present decline of native animal species and he said "Actually Greenlanders do not have to hunt anymore." Following the argument of the German informant, every Greenlander can buy the daily needs in the supermarket; therefore, *hunting is not necessary to survive* in modern Greenland anymore. It is not a hunter-gatherer society the Greenlandic Inuit are living in. Greenland is part of a globalized world.

This connection has been the subject of scientific challenges. Globalization can be defined as "the intensification of worldwide social relations which link distant localities in such a way that local happenings are shaped by events occurring many miles away and vice versa" (Giddens 1990: 64). The development of modern systems of information, communication, media, traffic, and transport has led to a growing worldwide interconnectedness and mutual influence among nations, societies, and cultures. This means that sovereign national states are still the main actors, but new forms of transnational actors such as non-governmental organizations or governmental bodies with shifting prospects of power, orientations, identities, and networks gain more influence (Beck 2000). This interconnectedness has been influenced the Arctic for many centuries. Scientific investigations focus on the question whether Arctic societies and cultures and their cultural practices are

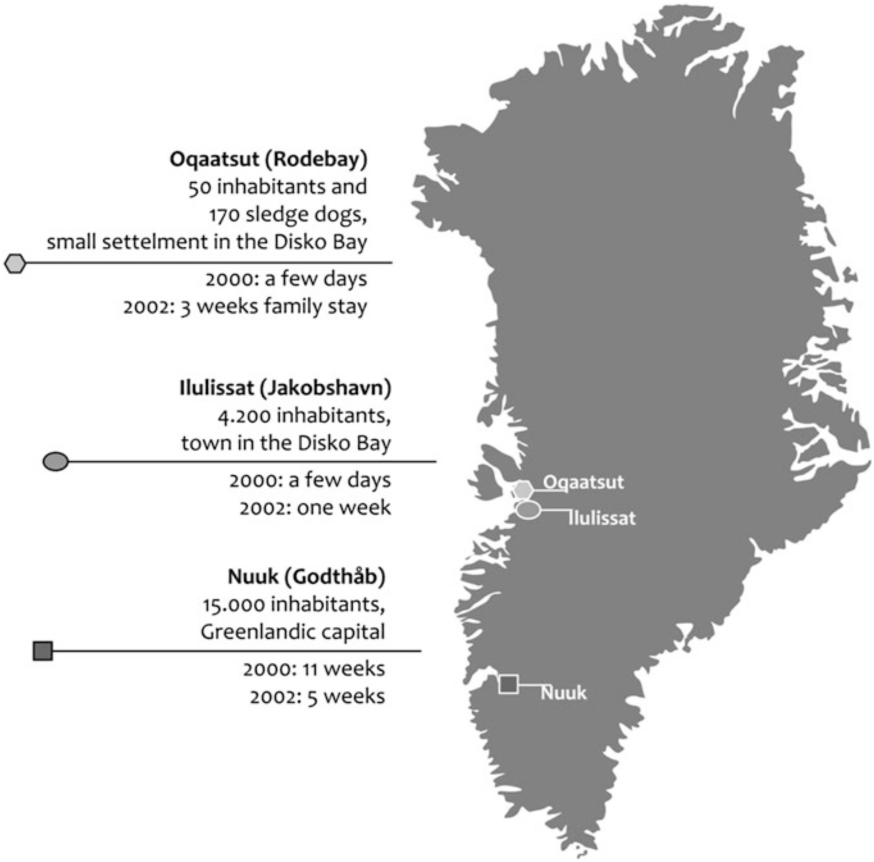

Fig. 9.1 Locations of fieldwork in Greenland

threatened by social disruption or can persist social and economic changes and whether there are cultural discontinuities or continuities of indigenous or aboriginal culture (Angell and Parkins 2011; Csonka and Schweitzer 2004; Sowa 2016). Today, Inuit live in a mixed economy of informal (i.e., subsistence) and formal activities and patterns of persistence and continuity in the "traditional" activities, cultural practices, and identity formations, and changes and fractures of these are identified (Einarsson et al. 2004; Marquardt et al. 1999; Nuttall 1998; Oosten and Remie 1999; Sowa 2012a, b; Stevenson 1997; Wenzel 2000). In order to make a living in a mix of wage-based economy and hunter-gatherer economy, many Northern residents are still engaged in subsistence activities, "defined as harvesting natural, renewable resources to provide food for one's own household, for gifts for others or to exchange outside the market economy" (Poppel and Kruse 2009: 39).

My interviewee from Germany used a concept of culture which implies that members of this culture share a set of collective attitudes, meanings, values, and practices. In times of globalization, these in this way conceptualized cultures come under threat through the spreading of a homogenized, westernized, consumer mass culture (Tomlinson 2000), This development is understood as an expanionist, one-way development from core countries to periphery countries. A perception which is described as the “McDonaldization of society” (Ritzer 1993), the evolution of a “McWorld” (Barber 1996), the Coca-Colonisation of the world (Macionis and Plummer 1998), or “the Madonna economy” (Group of Lisbon 1995). In this perception, members of cultures are constructed as passive recipients influenced by commercialism or Western thinking. This view does not take into account that members of cultures can be also seen as active recipients, who reinterpret, reinvent, or contextualize global consumer goods (Friedman 1990) and have also an impact on developments on the centers of Western societies (Abu-Lughod 1998; Beck 2000; Giddens 1999; Hall 1998a, b; Hannerz 1992; Nederveen Pieterse 1994; Robertson 1995).

Inside contemporary societies, there is a plurality of ways of belonging and being. Therefore, numerous research approaches in the cultural and social sciences understand that the “vocabulary of cultural description and analysis, needs also to be opened up to divergence and multiplicity, to the non-coincidence of kinds and categories” (Geertz 2000: 246) and reflect the conception of culture as shared by all of its members (Breinig and Lösch 2002; Matthes 1992; Straub and Shimada 1999; Welsch 1999). According to Welsch, the classical concept of culture was first defined as an individual culture through a specific demarcation from other cultures, and second, culture was always regarded as the culture of one people—where the latter was seen as the bearer and producer of this culture—and finally, third, culture had a strict internal standardizing function, that is, “every culture is supposed to mould the whole life of the people concerned and of its individuals, making every act and every object an unmistakable instance of precisely this culture” (Welsch 1999: 194). In Wolfgang Welsch’s view, the classical concept of culture is—to labor the point somewhat, as he concedes—“a sort of cultural racism” (Welsch 1999: 195) because cultures are conceived as autochthonous islands or hermetically separated ghettos that are uniform and distinctive. The three processes of modernization, the homogenization of forms of life, and the emergence of cross-cultures lead to hybrids, permeations, and the overlapping of cultural forms (Welsch 1999).

In spite of the fact that plural affiliations are calling the concept of culture into question through intracultural differentiations, there are attempts to reformulate the concept of culture. Here are Stuart Hall’s ideas helpful (Hall 1992). Hall challenges the conception of homogeneous cultural containers. Consequently, the idea of cultures as self-enclosed islands whose constituents can be defined unambiguously—whether in ethnic, traditional, or indigenous terms—is an illusion. Nevertheless, there is such a thing as the sense of belonging of the members of a society. Cultural or national collective identities are perceived as uniform and homogeneous only as long as they manage to represent themselves as such. Thus, I understand culture in a networked world as a (powerful) identification offer that is maintained through the representation of collective identities, often in relation to other collective identities. These notions of culture have implications for the present contribution because the point cannot be to study the constituent parts of Greenlandic culture. Rather, the point will be to examine existing, legitimate representations of Greenlandic culture, which develop in relation to other cultural collective representations—on the assumption that "cultures" are defined and demarcated in contrast to other "cultures."

Remembering my interviewee's argumentation that in a globalized (consumer) world, it is not necessary to continue hunting, there is no understanding about culture in the global era. These subsistence activities as a part of a Greenlandic culture are not characterized by common practices which are taken place on a regular basis rather by processes of identification. Studies have shown not only that subsistence activities are of economic and nutritional significance to a substantial number of Arctic households, but also that hunting is essential as a marker of identity (Poppel and Kruse 2009; Rasing 1999; Searles 2002). Through participating in hunting, fishing, herding, trapping, and sharing activities, Greenlanders can relate to their Greenlandic identity and identify themselves with the collective Greenlandic culture.

Let us now focus on Greenlandic foods which are called *kalaalimernit*. These foods include local game products that are served raw, dried, boiled, frozen, or smoked. Among the used species are marine species (seals, whales, and fish) and terrestrial animals (reindeer, polar bears, and various species of birds). *Greenlandic foods* are identified by the raw material, emphasized origin, as well as the method of preparation, with particular emphasis on "non-preparation" or "naturalness" (Petersen 1985; Sowa 2015a). In a globalized world, there are different pressures regarding *kalaalimernit*: First, it is the global flow of consumer goods (economic pressure), second the global flow of ideas (political pressures), and third the global flow of pollution and problem situations (environmental risks) which have a serious impact on *kalaalimernit*.

Kalaalimernit Under Pressure

Global Flow of Consumer Goods: Kalaalimernit *and Economic Pressure*

Considering food, the increasingly interconnected world leads to an expansion of foreign foods and cooking in Greenland. As a consequence, *kalaalimernit*—often characterized as "traditional food" (Pars and Mulvad 2002; Pars et al. 2001)—has to compete with a variety of imported foods and the consumption of *Greenlandic foods* is declining. According to a survey in 1994, 78 % of the people said that they are consuming Greenlandic fishing and hunting products once a week, but only 31 % do it daily (Böhmert and Poppel 1994: 12–15). Another population-based health interview survey in Greenland shows that imported products such as potatoes, cheese, and fruit syrup are used daily by more than 75 % of the respondents, whereas seal meat is eaten daily by 21 %, fish by 17 %, and wildfowl by 10 % of the respondents (Pars et al. 2001: 25). The data from a countrywide population survey in 2005–2009 show that traditional food items are consequently being replaced by imported food: Among the young Greenlanders (18–24 years) traditional foods made up only 12.8 % of the daily diet, among the older generation (60 years and older) the share is also low (28.3 %) (Bjerregaard and Mulvad 2012). There might be several reasons for this development. Firstly, imported foods are perceived as less expensive. The Danish eskimologist Sejersen notices that the reason for this perception is in fact the different quality of Greenlandic or Danish foods (Sejersen 1998). Danish foods are often in a lower quality and therefore very cheap. Some Greenlandic products have even no equivalent among Danish foods in view of quality. Furthermore, when people go to buy *kalaalimernit* at the open-air market *kalaalimineerniarfik,* there are larger quantities bought than in the supermarket. Sejersen concludes that however, as a principle, buying *Greenlandic foods* is considered as wrong by Greenlanders (Sejersen 1998). Secondly, the consumption of *kalaalimernit* is declining because of the wish for variety. Many current cookbooks promote also creative ideas for Greenland households (Kleivan 1996). In particular, the young Greenlanders like to vary their diet and enjoy the whole range of food products (Fig. 9.2). Pars and others diagnose: "By eating modern imported food, young people signal their interest in being a part of the modern world" (Pars et al. 2001: 29).

Global Flow of Ideas: Kalaalimernit *and Political Pressure*

Over the last few decades, the Arctic has been again and again a scene for international and intranational conflicts concerning the management of living resources especially that of seals and whales (Lynge 1992; Nuttall 1998; Osherenko and Young 1989; Wenzel 1991). The principal focus of attention is the hunting complex and therefore also the product of the hunting complex, the *kalaalimernit*. Within scientific studies, the Greenlandic hunting activities are blamed for the decline of individual animal populations (Boertmann 2007; Jensen and Christensen 2003; Kankaanpää 2001; Lisborg and Teilmann 1999; Pinngortitaleriffik 2000). A wider international audience was informed by a publication titled *A farewell to Greenland's wildlife* (Hansen 2002) and publications by the *World Wide Fund for Nature* (Hjarsen 2003, 2005). Due to the dissemination of a global model of nature (Sowa 2013c) including ideas of establishing a world environmental regime consisting of environmental management institutions and organizations, there are conflicts over the management of natural resources between scientists and hunters within Greenland (Sowa 2004b, 2014a), but also from an international perspective as conflicts over management between numerous non-governmental organizations and nation-states.

On the one hand, acting from many miles away, Western non-governmental organizations and international associations are interested in the hunting complex because they want to prevent the extinction of species, to save and protect the animal rights or to regulate the hunt. Geographic distance seems to have no significance anymore. On the other hand, Greenlanders want to continue hunting, sealing, and whaling and started to use the global flow of ideas (e.g., human rights) to establish their own non-governmental organizations and argue on an international level for their concerns. Here, the global model indigeneity is critical implying that the Greenlandic Inuit have been recognized as an indigenous people and that the Greenlandic Inuit are given special rights reserved for indigenous peoples (Sowa 2013c). In that sense, we are faced with a multidirectional worldwide traffic having old actors, as national states, and new actor, as the *Inuit Circumpolar Council* or other non-governmental organizations, involved.

Globalization in this context led to several direct consequences on the hunting complex in Greenland in the last twenty years. The most important were—to name only a few—the ban of sealskin, the use of special arms to hunt, and the hunting quotas on whales. In addition, the self-perception of Greenlanders, especially those who are involved in the hunting, changed through the intensification of globality, the awareness of a global world, and possible influences which shape the perceptions and the actions of the local actors. Following Kalland, the Western environmental organizations were very successful in establishing the whale as *the* symbol for environmentalism in the Western societies (Kalland 1993). This development led to a binary division of the world in those who save the whales and others who kill the whales, in other words: in good and bad people. One informant told me after the interview the following statement: "*And tell them we are not bad people*" Male, Nuuk, Greenland (personal communication, 2000). In this way, the worldwide interconnectedness has brought direct influences, e.g., hunting bans, modify in methods or quotas, as well as indirect influences, e.g., that Greenlanders think that they are perceived as ‶bad people' by others in the Western world.

Global Flow of Pollution: **Kalaalimernit** *and Environmental Risks*

Finally, the globalization of environmental risks concerns Greenland fundamentally. The German sociologist Ulrich Beck talks about a new *risk society* where there can be stated a tendency toward a globalization of the risks of modernity (Beck 1992). The first example of this new kind of global risk was the catastrophe of Chernobyl. The new quality of these environmental risks is that they are produced by human agency and that their scale is not local but global. It makes no difference where we live, or whether we are rich or poor. We are sitting in the same boat. According to Beck, "*The multiplication of risks causes the world society to contract into a community of danger*" (Beck 1992: 44).

Through the global flow of environmental risks, even the Arctic has to deal with contamination, which does not originate in the Arctic. These contaminants came originally from industrialized countries to the Arctic by wind and ocean currents. Hence, the Arctic ecosystem faces serious environmental pressures, including contaminants, oil, radioactivity, underwater noise, and habitat degradation (AMAP 1998, 2003, 2004, 2009; Dietz 2001; Macdonald et al. 2003). *Kalaalimernit* is also

Fig. 9.2 Greenlandic student in the Greenlandic capital Nuuk (own photography, taken in 2002, Sowa 2014c: 299)

affected directly by the global flow of environmental risks because of the presence of high levels of contaminants in the food chain (Hansen et al. 2008; Mulvad et al. 2007; Pars and Mulvad 2002). Through this accumulation in the food chain—this phenomenon is called *biomagnification*—all marine mammals contain levels of heavy metals, as cadmium or mercury, and *persistent organic pollutants* (POPs) that far exceed those that are regarded as acceptable (AMAP 2005, 2011).

There is evidence that *Greenlandic foods* are the major source of exposure for POPs and heavy metals including mercury (AMAP 2011; Hansen et al. 2008; Johansen et al. 2004, 2007; Mulvad 1997; Pedersen et al. 1999). These substances were detected in the blood and organs of Greenlanders, and the concentrations were above international guidelines (Bjerregaard and Mulvad 2012). The dominant contributors of contaminants in the "traditional" diet are seal muscle, seal liver, seal kidney, seal blubber, and whale blubber (Johansen et al. 2004). It is assumed that POPs and mercury could have several toxic effects, especially on developing fetus and the adult cardiovascular system (Bjerregaard and Mulvad 2012). Hence, it is getting more dangerous for human health to consume *kalaalimernit*, especially for those parts of the population that consume large quantities of freshwater fish or marine mammal tissues (AMAP 2011), despite the positive effects of consuming Greenlandic foods, as there are protein richness, long-chain n-3 fatty acids, and micronutrients (Bjerregaard and Mulvad 2012; Mulvad et al. 2007; Pars and Mulvad 2002).

In his speech “Stop the pollution but keep eating the food (for now),” the former president of the Inuit Circumpolar Conference (ICC), Aqqaluk Lynge, describes the dilemma Inuit are confronted with. On the one hand, more and more scientific research reports detect contaminants in the food chain with a grave impact on its consumers. On the other hand, “Inuit food” is nutritional: “Our food is healthy. Seal meat is nutritious and has been shown to fight disease. Whale maktak helps us fight colds and infections. And it does it naturally (…). Land animals such as caribou are freshly killed and consumed in a healthy and life-giving manner. Inuit food is good. Inuit food is healthy” (Lynge 1999). For Lynge, the alternatives such as buying only store-bought foods are not acceptable. Finally, he connects the consuming of *kalaalimernit* with culture: “Let’s remember that we have eaten these foods for thousands of years and through this we have developed our culture and celebrated our spirituality” (Lynge 1999). Aqqaluk Lynge refers to the link between food and culture. *Kalaalimernit* is seen as a very vital part of the Greenlandic culture. Among other things, it is used as a marker of difference and a very powerful tool to express a unique Greenlandic Inuit culture, as the 2002 Arctic Winter Games showed. The focus on that topic will be developed in the following chapter.

Kalaalimernit in a Globalized World: Food as Marker of Difference

The apparent contradiction is that on the one hand, people are not living in a strictly divided “*cultural container*” anymore. As theorists of concepts of hybridization, creolization, syncretism, and mélange effects argue, we have never lived in such units. But, on the other hand, people still relate to these categories of nation, region, and culture and furthermore—as I argue—they have to relate to these categories to be accepted by the “world society.” In the global reference system, people have to refer to generally accepted terms and standards. Hence, if disadvantaged groups will be successful in their claims of land or more rights, they have to present a unique (or sometimes “authentic”) culture. Therefore, a few elements of the peoples’ everyday life, or *lebenswelt*, are glorified and presented to the outside world. These elements may not be the most relevant elements of peoples’ everyday life, but the most distinctive to express successfully the uniqueness and otherness. In this light, globalization can be seen as opportunity for the actor “Greenland,” because it is on its way to build up a nation (Sowa 2013d).

Starting point of this nation-building process are developments after the Second World War. By the modernization of Greenland in the 1950s and 1960s, there was a taking over of cultural elements and institutions (Kleivan 1969/70a), as well as Danish foods and language (Kleivan 1969/70b). It is described that Greenlanders became "*Northern Danes*" (Caulfield 1997: 36; Dorais 1996: 29) and "*increasingly felt like strangers in their own land*" (Caulfield 1997: 36). Furthermore, the payment of the workers occurred after birthplace criteria, what meant that Danish workers got preferential treatment to Greenlandic workers. The process of *Greenlandization* (Breinholt-Larsen 1992; Nuttall 1992), a process to gain more equality in Greenland and more independence from Denmark, has led to Greenland Home Rule Government in 1979 and the establishment of a Greenland Self-Government in 2009 (Kleist 2010; Nuttall 2008; Thomsen 2013). In the contemporary interconnected world with a lot of power contests, it is more and more important to formulate a position using a distinctive culture. Therefore, it becomes a political strategy to strengthen the Greenlandic culture and to focus on a few aspects of everyday life. This is what others would call "*strategic essentialism*" (Spivak 1988).

To explain this strategy in contemporary Greenland, it is helpful to borrow Pierre Bourdieu terms of "distinction" and "taste." Bourdieu investigated people in France in the 1960s and 1970s and uses tastes to function as marker of "class" in a society where no grave class differences are obvious. Following his argument, the class system is still alive, but the differences are only slight. The lifestyle underlines difference and defines the class position in the social space. Bourdieu distinguishes the legitimate taste, the taste of the ruling classes, the middlebrow taste, the taste of the middle classes, and the popular taste, the taste of the lower classes. According to Bourdieu, "*[t]aste is what brings together things and people that go together*" (Bourdieu 1984: 241). The ruling class decides through symbolic struggles what the legitimate taste is. Hence, there is a possibility of change, and theoretically, almost everything could get the status of the legitimate taste. In the effort of keeping the pole position, the ruling classes try to use taste as distinction to the other classes. Though tastes can function as distinction leading to a class formation, "Tastes (i.e., manifested preferences) are the practical affirmation of an inevitable difference. It is no accident that, when they have to be justified, they are asserted purely negatively by the refusal of other tastes" (Bourdieu 1984: 56). According to Bourdieu in the day-to-day practices, the effort by the upper class to form a distinctive group can be witnessed, e.g., in considering the food sector.

This food sector is also suitable to form cultural groups, taking the restriction into account that there is a limitation of what is able to be considered as *Greenlandic foods*. However, through the "good taste," Greenlanders are able to dissociate themselves from Danes in the country. Powers and Powers agree when they are arguing that "[f]ood is capable of symbolizing the manner in which people view themselves with respect to insiders and outsiders of society" (Powers and Powers 1984: 86). In other words, the person belongs to the group when he or she proves his or her "good taste." The Greenlandic eskimologist Robert Petersen describes it as follows: "[I]t is the common understanding that in order to have a Greenlandic identity the person must eat dried meat, dried fish, raw mattak, etc. Not only should the person be able to eat it, he or she should also like it (which is not so difficult). In a lot of places it has become difficult to get these kinds of food for every-day consumption. Often they are served at festive occasions, giving them cultural and social significance and making them a symbol of a Greenlandic celebration" (Petersen 1985: 299).

Kalaalimernit function, as well as the Greenlandic language, is a marker of difference (Sowa 2004c). This distinction could also be analyzed in a Japanese whaling community where the consumption of whale meat is used to strengthen the identity of the inhabitants of the whaling town (Sowa 2013e, 2014f). *Greenlandic foods* are both suitable for building up a Greenlandic community and as a marker of a cultural boundary between Greenlanders and Danes on the one hand and Greenlanders and the whole world on the other hand. It could be a strategic possibility to express indigenousness when Inuit try to secure indigenous rights as international whaling quota from the *International Whaling Commission* (IWC) (Sowa 2012/2013, 2013b, c, d, f, 2014d). In times of the first contacts between Europeans and Greenlanders, foods also played an important role in separating the civilized people (the Europeans) from the primitive savages (the "early" Greenlanders). The missionaries Hans Egede and David Cranz wondered about rotten and stinking seal meat (Cranz 1765: 190; Egede 1986 [1741]: 363), as well as the uncivilized behavior, e.g., the lack of table manners (Cranz 1765: 191–192; Egede 1986 [1741]: 359) and lack of fixed meals (Cranz 1765: 192; Egede 1986 [1741]: 363). In the perception of the Western observers, Greenlanders were seen as guided by instincts and emotions who do not act according to human reason (Cranz 1765: 245). Maybe this quality of food is also shown not least by the mistaken translation of "Eskimos" as "eaters of raw meat" (instead of "snowshoe-netter") still used to this day (Damas 1984: 6).

What makes *Greenlandic foods* as a means of distinction in a cultural perspective? There are six important points: First, it is the special name, second the places where it is found, third the extraordinary quality, fourth the unique sign and symbolic character, fifth the cultural indoor and outdoor practices that are involved, and sixth the connection to the idealistic image Greenlanders have about themselves which altogether establish a distinctive group with a collective identity. These six points strengthen the Greenlandic identity and lead to the permanent production and reproduction of cultural boundaries to Danes and the world. This does not mean that these boundaries are stable. For example, it is obvious that today more non-Greenlandic foods are consumed than the *Greenlandic food*. Hence, Greenlanders have to communicate and practice the cultural boundaries again and again.

Name of Greenlandic Foods

The Greenlandic expression for *Greenlandic foods* is called kalaalimineq, or in plural *kalaalimernit*. Literally, it means "piece of a Greenlander" because *kalaaleq* means "a Greenlander" and *mineq* "a piece of" (Roepstorff 1997). Here lies a central difference to imported food, which is called *qallunaamineq*, in the translation "food of a Dane" because *qallunaaq* means "a Dane" (Berthelsen 1997) or "a non-Inuit/a white man" (High North Alliance 1997: 7). To put it in a wider context, one could say *qallunaamineq* means "food of a white man." The Greenlandic terms for food show a deep linguistic identification of the Greenlanders with their own food.

Places Where Greenlandic Foods *Are Found*

Today, not everybody in Greenland has a direct connection to the hunting complex because of the job structure. In 1997, there were only 2579 full-time hunters in Greenland, while the spare-time licenses increased to 6878 (Greenland Home Rule Government and Statistics Greenland 1999: 56). Hence, there are certain places where it is possible to get *kalaalimernit*, e.g., in the local supermarkets where special zones are established for Greenlandic specialities. However, normally people in Nuuk buy fresh meat at the local open-air market where hunters sell their daily catches. This open-air market is called *kalaalimineerniarfik*, which means, "*place where Greenlandic foods are sold,*" and every town has this kind of market. These markets were founded in the eighteenth century to guarantee the meat support for Danish, and later Greenlandic staff are employed by the church and workers of the Royal Greenland Trade Company (Marquardt and Caulfield 1996). Following one of my informants, people run to the open-air market when there is fresh whale meat available and it is sold out in a very short time (Female, Nuuk, Greenland, personal communication, 2000).

Quality of Greenlandic Foods

The third point deals with the quality of the *kalaalimernit.* Here, it should be indicated that when talking about *Greenlandic foods,* it is not a thing of quantity but always of quality. While the quantitative consumption of *Greenlandic foods* is decreasing, a scientific study proves the enormous qualitative value of the foods considered by Greenlanders (Pars et al. 2001). Through all age groups, the considered value of *Greenlandic foods* (4.67 points) is significantly higher than the value of imported foods (4.10 points). The scale used in the study is between 1 point (= I do not like it) and 5 points (= I like it very much) (ebd.). Other scientific studies reveal that Greenlanders support the quality of their own foods because it is with low fat and a lot of proteins and rich in vitamins compared to imported foods (Freeman et al. 1998; Helms 1997 [1983]). As another survey shows, eating meat of seals and whales is considered as very healthy. 90 % of the questioned have the opinion that *kalaalimernit* is good for their health (Bjerregaard and others 1995: 38, quoted by Kleivan 1996: 150).

Food as Sign and Symbol

There is also a symbolic character of *kalaalimernit*. People consider the meat as high quality because the animals come from their *homeland* (Nuttall 1992) and this is certainly a pure and healthy land (Sejersen 1998). With the help of consuming *Greenlandic foods,* one is able to express his or her Greenlandicness. Whenever somebody likes to show other people that one relates or belongs to Greenland, one offers *Greenlandic foods*. A good example is also the celebration of the national holiday on the 21st of June. Days before, one notices the specific smell of whale in the air, and on the day itself, one could taste it from hunters at certain places in Nuuk, for example, at the "*house of the Greenlanders*"—which is a very old and famous ruin outside the city. There, seal meat was offered in a traditional way as seal soup boiled with rice and potatoes. A lot of families go out in the nature to have a picnic or barbecue with whale or seal meat. Also on a *kaffemik*, which is the Greenlandic term for a celebration, one has to serve *Greenlandic foods*. There is an important point to make: A private birthday party takes the character of a national celebration. People show their Greenlandic identity and distinguish themselves from the Danes who live in their country.

Food as Distinctive Cultural Practices

The fifth important point is the fact that there are cultural practices involved (Fig. 9.3). A significant activity is the sharing. This sharing activity among hunters is important, e.g., after the beluga hunting where several hunters are involved. Through sharing, a communal solidarity and traditional values are reproduced and confirmed (Dahl 1990, 2000). Marine mammals are also shared in wider networks as a gift expressing the relationships people have with each other. In this way, sharing cements bonds of kinship and close social association (Nuttall 1991). Furthermore, sharing activities also take place on several occasions where, e.g., dried fish or raw *mattak* is offered. When such a snack is presented, one cuts off a piece of the fish or *mattak* and gives it to the neighbor while talking together. Frank Sejersen noticed that "Greenlandic snacks are *shared*; Danish snacks are *served*" (Sejersen 1998: 62).

A special outdoor activity is the consuming of *Greenlandic foods* in nature (Sowa 2012a, b). For some Greenlanders, they consider "*being in nature*" as one aspect of their culture. To have a close relation to nature is considered as a part of Greenlandic culture: One of my informants said: "*It's also important to have a tight relationship to the nature. Of course we have not, unfortunately we have no boats right now but now we use to use the bike to near by the airport or go to the nature and eat or make seal meat*" Male, Nuuk, Greenland (personal communication, 2000). Another informant added: "*I think it's always been important in summer time you go out in nature, be free in the nature. To live, to eat seal or birds or fish. Many Greenlanders when they go out sailing it's the time when they are feeling free*" Female, Nuuk, Greenland (personal communication, 2000).

Fig. 9.3 Arctic Winter Games: sealskin flensing contest (own photography, taken in 2002, Sowa 2014c: 185)

Another special outdoor activity is the hunting itself: The animals are hunted directly. Hence, there is a concrete relationship to the whole animal. In addition, when large animals are hunted, there are all sorts of special rules, for example, even for how a whale is to be shared after the hunt. In small places in Greenland, a communal solidarity and traditional values are reproduced and confirmed by hunting beluga whales (Dahl 1990; Sowa 2012/2013). Also, the celebration of the first shot seal is a sign for the continuation of the hunting complex as Mark Nuttal argues (Nuttall 1992: 26).

Food as a Connection to the Greenlandic Idealized Self-image

In the perspective of the urban Greenlanders, they have the chance to get in touch with their idealized self-construction of the "real" Greenlander by consuming *kalaalimernit*. The idealized self-image describes Greenlanders living in the small communities going hunting and fishing. In the Greenlandic capital Nuuk, Greenlanders are talking often about Greenlandic villages where the "authentic" Greenlanders should live. It is remarkable that many urban Greenlanders who are "real" Greenlanders do not consider themselves as "real" Greenlanders. However, according to my research, these urban Greenlanders can perceive themselves as Greenlanders when they are consuming *kalaalimernit*. Furthermore, as examples from other Arctic regions show, that food plays an important role in the construction of an Inuit identity. The following quotations express this perfectly: "*I am still Inuk because I grew up on Inuk food*" (Freeman et al. 1998: 39) or "*My body as an Inuk is not fully Inuk without eating animals*" (Langlais 1995: 55, quoted in Freeman 1996: 50), and finally, "*Seal blood is in all Inuit who (eat) animals. Seal blood gives us our blood. Seal is life-giving*" (Inuit Circumpolar Conference 1996: 55). For urban Greenlanders who know they are not perfectly Greenlandic (Female, Nuuk, Greenland, personal communication, 2000), they could fulfill their own self-image by eating *kalaalimernit* at festivals or at least once in a month. It is a common understanding that if one wants to be a real Greenlander, one has to eat and speak as Greenlandic.

In addition, many Greenlanders who live in Denmark also feel the need to eat as Greenlander. The only way to satisfy this need is to visit the Greenland House (*Kalaallit Illutaat*), where they can buy frozen and dried meat, as well as fish products from Greenland. There are also celebrations where, e.g., boiled seal meat in soup and blubber is offered. *Kalaalimernit* is an important and a strong symbol of Greenlandic identity: "The food brings back the memories of situations in Greenland, and the enjoyment of the special Greenlandic food while socializing with other Greenlanders and speaking Greenlandic are imported ways of maintaining their sense of being Greenlanders" (Kleivan 1996: 155).

Kalaalimernit is a very strong mean of distinction because of the special name, the place, the considered quality, the sign and symbol character, the practices, and the connection to the idealized self-images. It is used as a marker of difference on special public events to express the uniqueness of the Greenlandic culture to the world society. Globalization in this perspective can be seen as the opportunity to present the special Greenlandic way of life to fulfill the requirements of the global reference system; consequently, all cultures are different and separated. In establishing an accepted Greenlandic culture, the actor "Greenland" is able to push ahead the nation-building process and to get accepted by the world.

Conclusion

My aim of this article was to present globalization as a phenomenon of pressure as well as opportunity. Considering *Greenlandic foods,* I offered three "threats" to *kalaalimernit*, the global flow of consumer goods, the political pressure concerning the management of living resources, and the globalization of risks. On the other hand, *kalaalimernit* is a strong marker of distinction and can be able to form a cultural group in the global context. Through strategic staging of their own culture, e.g., during the Arctic Winter Games in the year 2002, it is possible to mark a cultural boundary in an active way and to construct a difference to Danes in the country and to others outside the country. This finally enables Greenland to become a self-confident player in the global politics of power and to influence their own future. It is a real predicament that for the Greenlandic culture, it is necessary as well as risky to keep on eating *Greenlandic foods.*

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Chapter 10
Human Security in the Arctic: The IPY GAPS Project

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During the usual rush of "Arctic" themed conferences in the fall of 2014, human security in the Arctic was on the agenda. The Transatlantic Science Week in Toronto, Canada, arranged a panel that looked at non-state security challenges in the Arctic, ranging from search and rescue issues to health, food, identity, and

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environmental security.[1] The Gender Equality in the Arctic Conference in Akureyri, Iceland, devoted a plenary session to human security and gender, and the Arctic Circle conference in Reykjavik, Iceland, included both full panels as well as numerous individual papers that addressed human security issues in the Arctic.[2] Many of the invited presenters were scholars that have been the initial drivers behind the exploration of the relevance of the human security concept in the Arctic (including some of the authors of this chapter). This chapter seeks to show how our IPY project—"Impacts of Oil and Gas Activity on Peoples of the Arctic using a Multiple Securities Perspective" (GAPS)—has facilitated this growing attention to human security in the Arctic and outline some of the project's overarching contributions.

Ten years ago, applying the concept of human security to the Arctic context was largely unheard of. The idea was nevertheless being explored in 2004, when researchers from different disciplines ranging from international relations/political science, to anthropology, ecology and ecotoxicology, met at the University of Tromsø, Norway, to discuss whether or not the concept of "human security" was at all relevant to the Arctic context. There was a lot of scepticism in the room back then, particularly by researchers generally used to employing the concept in contexts of the global South. On the one hand, it could be argued, bringing the human security concept to the Arctic meant a cooptation of the concept that was supposed to further expose inequalities and hardships experienced in weaker or failing states. Bringing human security to the Arctic could mean devaluing the suffering of populations in the global South, giving the global North priority and attention where it is not needed. On the other hand, however, exploring the meaning of the human security concept in the global North could potentially reduce the "virtuous imperialism" embedded within the concept when applied to the South; where potentially noble and virtuous help is delivered from North to South, often on terms determined by the North, and for Northern security interests (Hoogensen Gjørv 2014).

The debate over the relevance of the concept to the North and more specifically the Arctic, continues, but there is increasing acknowledgement and application of the concept to the region. In fact, it is arguable that increasing acknowledgement of the complexity of security in the Arctic region, not least including human security perspectives, has become both relevant and necessary to better understand the complex concept of security.

The linkages in the Arctic between environment, human social and economic activity and their associated values and energy dependency are increasingly clear. Until very recently there has been a marked increase in proposed activities regarding energy and mineral resource development in the Arctic region. Even with the current downturn in prices for oil and gas, and the recent United Nations Framework Convention on Climate Change (UNFCCC) COP 21 Paris agreement,

[1]Transatlantic Science Week 2014: http://www.cvent.com/events/transatlantic-science-week-2014/agenda-1f6f78d182a64bddb793d86ed0214781.aspx.

[2]Gender Equality in the Arctic Conference: http://www.arcticiceland.is/en/program; Arctic Circle 2014: http://arcticcircle.org.

there are many analysts who claim that oil and gas (particularly in the Arctic) will still have a role to play, not least replacing dependencies on coal (Topdahl and Stokka 2015). The tensions between economic security, energy needs and energy security, and environmental security have thus been heightened with the increasing global attention and scrutiny over such extractive industry and their potential impact on global climate change, habitat degradation, community health and welfare, as well as apprehensions over offshore drilling that arose with renewed vigour after the 2010 Gulf of Mexico oil spill.

The University of Tromsø—the Arctic University of Norway, in cooperation with the Fram Centre (FRAM—High North Research Centre for Climate and the Environment, a combination of the former Norwegian Polar Institute and other research institutes), York University (Canada) and the Ural Division of the Russian Academy of Sciences (RAS), cooperated on the IPY GAPs international project that attempted to address the question of human security in the Arctic in the context of oil and gas development. GAPS investigated these linkages by focusing on the dynamics of human/community needs in the Arctic. GAPS claims that human security provides a potentially powerful analytical tool for both understanding and analysing individual and community security within different contexts, including in the Arctic. However, making human security meaningful requires a "democratization" of security and demands attention be paid to what communities themselves value in the contexts under examination.

The following chapter provides an overview of the roads travelled in the IPY GAPS project process, taking its departure point from the original project application as well as an earlier co-authored article (Hoogensen et al. 2009a, b), combined with additional contributions from key partner participants, updating research accomplishments and highlighting the ways in which the IPY GAPS legacy lives on in current research.

State Security and Human Security and the Spaces Inbetween

In general terms, security is about the reduction or absence of fear (Liotta and Owen 2006). Security combines the elements of actors, values, practices, survival, and the future. All of these are linked by fear. Fears are often identified based on the extent to which something is valued—for example most people value their life, and in some contexts (like war or domestic violence) people are in fear of their life being taken away. If they did not value their lives, this would not result in fear or an issue of security. Security is a process that moves towards its goal (feeling secure) by identifying causes of fear (based on values) and providing measures to mitigate against these fears (protecting that which we value) and ensuring our fears are alleviated for the future. Anyone can engage in this process, and individuals, communities, states, and the global community do, but the study of security, at least that which has in the last century informed policymaking on security, has focused very much on only one dimension—and that is the security of the state. Stephen Walt argues that security studies are about the phenomenon of war and that it can be defined as “the study of the threat, use, and control of military force” (Walt 1991). The process of securitization, or bringing an issue or agenda into a security framework, requires the state to act in an extraordinary way, using means and measures that we normally do not wish to employ otherwise (Buzan et al. 1998). These measures usually refer to the use of force, and in particular the use of the military. The decision to use force and mobilize the military is associated with “high politics” or the top priority of the state and does not include action at the political or local levels (such as social security or economic policy) or “low politics” (Hough 2008). It is largely due to this special relationship of security to the state and military, or “high politics” concerns, that many in the traditional security community argue vehemently against a widening of the concept, whereby security might be recognized as having something to do with communities and individuals (known popularly as “human security”) as well as the state.

State-based security has been reified as the “traditional” approach to security, but this is a misleading and ahistorical reading of the term (Fierke 2007). There is no particular logic that supports that claim that security can only be the purview of the state, and we must not forget that individuals and communities will always have a significant role to play in the provision of security. In fact, there is no possible way that the state can provide security in the multiplicity of ways that contribute towards the protection of individual and community values—much of these are carried out at the individual and community level. However, there are significant differences between what is and can be done at each level. The identification of threats and the correlative means by which such security is created differs according to scale. Whereas the state might be most preoccupied (rightly or wrongly) with threats of a military nature, the threats and processes of providing security at the more “local” level may amount to something quite different and be rooted in cultural, food, gender, environmental, economic, health and/or political security. Various actors identify threats and create security on the basis of different priorities or values, as seen for example in the differences between individual concerns about economic security, community values about identity, or a state-based preoccupation with terrorism. Who is right, or rather, whose security “counts”? That depends in part on how we envision the role of the state, and it is this role that either recognizes other actors or not.

In the Arctic, the state/military-based language of security has been actively employed for decades (Heininen 2004; Huebert 2006; Borgerson 2008). Security in the Arctic has focused on issues of power, resource exploitation and territory, and these issues continue to influence government policy in the region. As we have noted in earlier articles (Hoogensen et al. 2009a, b), the initial move towards examining the relevance of the human security concept to the Arctic context was inspired by the agreement signed between Canada and Norway committing these states to the concept called “human security”, which focuses on individuals and communities and can be very loosely defined as “freedom from fear, freedom from want” (UNDP 1994). This Canada–Norway document, known as the Lysøen agreement, formalized their dedication to issues threatening individual well-being, such as landmines, the employment of child soldiers and threats against women and children in conflict. The agreement also included, however, a commitment to strengthen "Arctic cooperation" between the two nations. The links between the Arctic and human security were not fleshed out in this document, but it became one of the springboards in the creation of what was called the Human Security Network, which expanded beyond Canada and Norway to include such countries as Jordan, Thailand and Costa Rica, and which therefore made such possibly rhetorical issues between the two nations relevant on a global scale.

The Lysøen document generated questions about what human security can and does have to do with the Arctic. In the Arctic, it becomes very clear that the "categories" we use to identify security, from military to human security, are very fluid, and by and large very closely linked. Economic security cannot be completely isolated, for example, from environmental security, energy security or political security (Hoogensen 2009). The Arctic, a region that is subject to various geographical and political interpretations (Einarsson et al. 2004), is a region shared between eight sovereign states (Russia, Canada, USA, Norway, Sweden, Denmark (Greenland), Finland and Iceland) though not composed of eight complete states, whereby almost all of the eight states that govern the Arctic are partially located outside of the Arctic region and have their political centres far to the south of the region (ibid). It is also predominantly ocean surrounded by land (Verhaag 2003). The region is characterized by distinctive differences in climate, culture and politics across geographies, combined with shared challenges and interests. These include the vulnerability of the environment to human activity, great distances between human communities, and the wealth of diverse natural resources, which are often extracted or hunted to the benefit of powers south of the region.

Due to, among other things, the military baggage of the narrow security concept, there are some who argue that the notion of human security in the Arctic is misplaced, if not a post-colonialist imposition of "Southern" governments and scholars upon Northern peoples (Griffiths 2008). It is true enough that the concept of human security is a western construction meant to be employed by "Northern" (meaning global North, such as Canada, USA or European countries) states towards the global South (Hoogensen Gjørv 2014). Human security has been criticized as a tool of the North (Duffield and Waddel 2006). These criticisms are valid, as are any that suggest that communities are forced to comply with state or elite-imposed programmes. As noted by Maria Lvova in her GAPS research, in general individuals and communities do not discuss their "human security" concerns when on their neighbourhood walks, not least because the term "security" has significant social meaning due to linkages with the military and the state. However, human security is a concept that has the potential to enable us to include and listen to the concerns of non-state actors (like individuals and communities) where they have been previously excluded, and to understand the relationships between different values, actors and levels of security creation (Duffield and Waddel 2006).

We argue that security has two sides—it clearly exposes vulnerabilities and sources of fear, but it also speaks to capabilities and enabling—people, societies, groups and states to ensure their security by a variety of means (including the military, but also by other means), to ensure that life continues, to even make sure a good life can be found.

There are many ways in which one might be able to theorize both the positive and negative dimensions of security. Individuals and communities will endeavour to seek security, not just in relation to avoiding threats, but also with the goal of building their capacities. Taking our departure point from the popularized definition of human security in the 1994 UNDP Human Development Report (UNDP 1994), and further work on a multiple actor framework (Hoogensen Gjørv 2012), we can try to understand a multi-scale/actor, democratic and context-based definition of security in the following way: Security is achieved when individuals and/or multiple actors have the freedom to identify risks and threats to their well-being and values (negative security), the opportunity to articulate these threats to other actors, and the capacity to determine ways to end, mitigate or adapt to those risks and threats either individually or in concert with other actors (positive security). By "actors" we mean individuals and communities, researchers and research communities, governmental and non-governmental organizations, media, business/industry, militaries and policymakers. All of these "actors" have a role to play in identifying threats and creating security.

The Arctic, and Arctic Oil and Gas

Over centuries, Arctic peoples have learned to adapt and thrive in an uncertain, harsh environment. Change, from environmental to economic to political, is occurring in the Arctic at increasing rates, placing great pressure on local peoples' capacity to cope. Such change can strain many factors that are necessary for human well-being in the Arctic. These include the health of the environment, the supply of traditional foods, community health, economic opportunities, and political stability. The relationship between the challenges faced by individuals and communities and the ways in which they meet these challenges are not well understood by the political institutions (in other words, the state) that have been traditionally "designed" for confronting the challenges that can threaten the security of individuals and communities. In fact, the state is not and has never been able to confront all challenges—and yet, the capacities and concerns of individuals and communities are often minimally, if at all, taken into account (as noted by many indigenous political movements).

Since the early 1970s, oil and gas development has come to dominate the industrial sector in the Arctic. The pace of resource development has accelerated significantly in recent years and for the longest time prices were on the rise, motivating industry to travel further north to extract fossil fuels for global consumption. Increasing pressures from various governments—Russian, Norwegian, Canadian and American—requires the Arctic to be open for business, as well as sends a clear message about who has control over significant energy resources. Even though questions are now arising regarding the dependency on oil and gas due to dropped prices, the likelihood that these extractive industries will be central to economies and energies for the foreseeable future is still high. Arctic communities are still tied into the global market for oil and gas, putting more pressure on their individual and societal capacities to cope with change, participate in resource management decision-making, and secure any possible economic and social benefits.

Oil and gas activity poses critical challenges as well as significant opportunities to the security of communities, affecting local economies, traditional livelihoods and identities, health, food, and the environment (Deiter and Rude 2005). Furthermore, many of the current drivers of change in the Arctic are only expected to intensify in the future (IPCC 2014). Such large-scale alterations of the environment interfere with local peoples' capacity to adapt by putting access to resources—and the resources themselves—at risk (AMAP 2009, 2014). While GAPS focused its analysis on the effects of oil and gas development in the Arctic on local communities, it is impossible to separate such activity from an overall context of change—particularly, climate change—as these processes interact and overlap in ways that can make potential effects even more acute.

However, acute does not always mean negative, at least from a human systems perspective. For example, increased oil and gas use has already been implicated in increased climate change, particularly in the Arctic region. From an environmental–ecological perspective, the consequences of climate change can be destructive, or at least drastically alter ecosystems such that both human and non-human species either must significantly adapt or be wiped out (polar bears, e.g. or in the case of humans, migration from areas affected by sea rise). On the other hand, the melting of the sea ice, a consequence of climate change, increases access to oil and gas

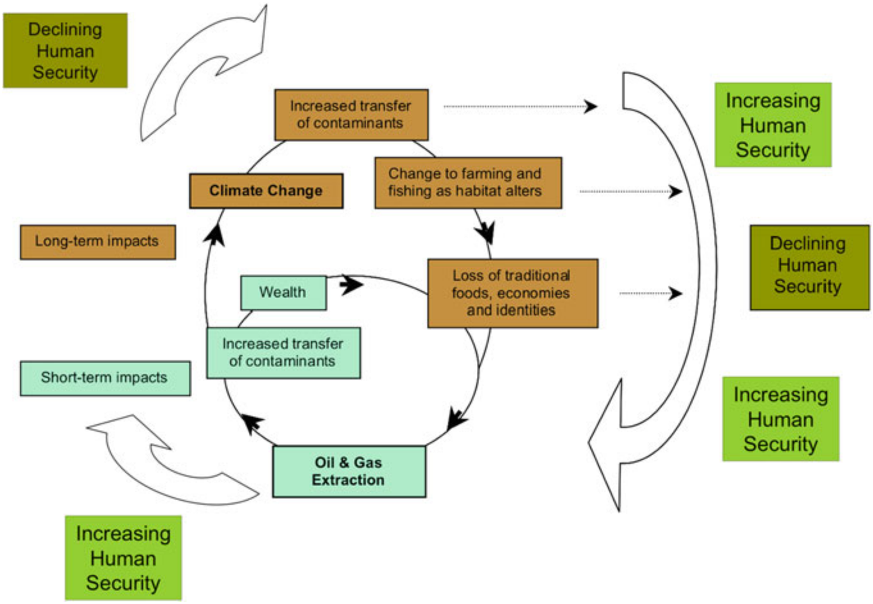

Fig. 10.1 Potential consequences of the Arctic fossil fuel industry. These consequences may result in human security increasing or decreasing depending on local community response, adaptation and resilience, and the values and visions of the future that inform community approaches

reserves in the Arctic, as well as opens up transport potential through both the Northwest and Northeast passages, translating into economic opportunities that could support some Arctic communities. At the same time, oil and gas thriving communities have the potential to devastate fisheries communities or reindeer herding as the wealth moves from one resource to another. How local communities respond to the emergence of factors associated with the longer-term impacts of fossil fuel industries may then feed into decisions about how to manage and develop these resources in the shorter term (Fig. 10.1).

The past decade has seen an increase in the use of the notion of "energy security" and it remains on centre stage, whereby governments are interested in securing energy resources (maintaining supply) at a reasonable price, not least to both exert power over those who are dependent upon these resources (Yergin 2006; NATO 2009; Ciuta 2010; Umbach 2010). The tension created by the recent downturn in oil and gas prices demonstrate that many states, not least Norway, Russia, Canada and the USA, are interested in increased ease of cheap transport and access to as well as sale of domestic resources, as climate change alters the Arctic landscape and increases accessibility of these resources. However, as stated above, the burning of these fuels to satisfy global energy demands will further accelerate climate change, impacting not only states that which to ensure energy security, but also humans and their human security. Consequently, oil and gas plays an increasing role in security debates by both increasing sought-after revenues for Arctic states, and also providing for a reduced dependency upon Middle Eastern sources. As oil and gas activity continues, and it will even under the constraints of COP 21, it is critical that we understand its effects on communities in a comprehensive way, so that we can explore all the factors that contribute to a sense of well-being or human-based security. Through collaboration and communication with communities, we can examine the risks, threats and opportunities that oil and gas activity presents to human security in the Arctic.

Understanding Security, Understanding Security in the Arctic

The GAPS project contributed significantly to the development of a multi-actor security framework that plays a central role in our understanding of security in the Arctic. The creation of security and insecurity, and by whom (actors), is very much dependent upon the contexts being referred to. However, not enough knowledge exists regarding creation of security and insecurity, and the identification of threats at non-state levels while state-based security discourses attempt to maintain their dominant position on what can and cannot be called "security". The GAPS participants were ready to explore the ways in which that human security had potential for bringing complex, multi-level and multi-actor Arctic challenges and solutions into sharper focus. The project was international and interdisciplinary from the beginning and had two main goals. The first was to determine the meaning of security in the

Arctic context using human security as a central lens by which to understand the Arctic. The second was to identify and document the impacts of oil and gas development on Arctic communities through a security lens.

It was clear early on that a multi-actor approach was necessary. Initially, the actor categories in focus were "Arctic communities", "research", and "policy/policymakers". The "policy" category included the military which in combination represented so-called traditional state security perspective as that is always present. Our inclusion and privileging Arctic communities in Arctic security, however, ensured that a central activity of the project would be identifying the factors that contribute to (in)securities as articulated by community members and then further define the relationship between traditional and human security in the Arctic context. This process would include identifying potentials for local adaptability/capabilities to ensure human security in communities as well as determine linkages and disjunctures between research-based and community-based identification of threats and capabilities. This theoretical objective would be complemented by the second goal, that to explore empirically the impacts of oil and gas activity on Arctic communities and their environments.

A multi-actor security approach is further illuminated by the concepts of positive and negative security, which reflect the tensions between the ways security has been conceptualized, who provides security and how, and how scholars and practitioners themselves place a "value" on security. Negative security relates to the treatment of security as a concept we wish to avoid, a concept that should be invoked as little as possible. We value it negatively because it represents the use of force. In the Arctic context, the perceived military build-up of different Arctic states and the potential threat of use of force that such build-up implies would fall into the category of negative security—this type of a fearful dynamic of mistrust between states is ideally to be avoided. On the other hand, the concept of security has also been known to represent something that is positively valued, or as something that is good or desired, providing a foundation to pursue our needs and interests and enjoy a full life (Roe 2008; Hoogensen Gjørv 2012). In the Arctic context, positive security would refer to sustained livelihoods (traditional or market economy based), access to clean food resources, gender equality, long-term environmental health and sustainability, and thriving and inclusive community identities and political representation (Deiter and Rude 2005; Hoogensen Gjørv et al. 2014; Sweet 2014). Positive security reflects a different, but also central, foundation for security based on a foundation of trust. Trust is established more easily through non-violence means, through negotiation, compromise and dialogue (Hoogensen Gjørv, forthcoming). In an environment of trust, security actors (individuals, communities) can further build (enabled) upon those values and priorities that contribute to their own understanding of what makes them secure. Understanding the core assumptions within positive and negative security allows us to observe and assess the field where multiple actors play a role in diverse settings. The increase of acknowledged actors as well as the complexities behind how state authorities interact with other actors and how they are perceived become crucial to understanding security.

There are distinct relations of power between these actors, where certain actors have been more marginalized than others. Policymakers and the military have traditionally played dominant roles in the articulation of security, while communities and individuals have gone been greatly marginalized as security actors, if not ignored completely. Within the model lines of communication connect them all but power affects communication realities. A discussion of the multiple security challenges of climate change in the Arctic in Norway might include very different actors than the same discussion taking place in Russia, where actors in each case are affected by power and constrained in different ways. The security perspectives of each set of actors will not avoid conflict, and at times one category of actors may, in some ways, create insecurity for another(s). The purpose of the model is to make visible competing security perspectives (instead of marginalizing or ignoring some) within a given context, upon which possible compromises and solutions can be based.

Thus in our framework, Arctic communities express their perceptions of threats and assess their capacity to cope with them. These expressions are not isolated from those threats identified by Arctic researchers (e.g. contaminants in traditional foods), and the interactions between these two communities (Arctic peoples/communities and Arctic researchers) are explicitly linked. Other actors like the media, business, the military and policymakers contribute to the actor-based security “hexagon” and generate a multiple securities approach. This does not mean that government and policymakers are obliged to intervene at all levels of identified human (in)security in the Arctic. However, they can act as one (in concert with media, research, etc.) important conduit for the facilitation of knowledge between communities, as well as responding to human (in)securities when communities or other actors can no longer respond effectively to threats.

Case: Human Security, Oil and Gas in Norway

A contextualized understanding of human security in the Norwegian context was provided by one case examining the impacts of the oil and gas industry on developments in Lofoten region of Norway (Dale 2011). During the first decade of the 2000s, there were decisive steps taken towards including this northern region in what has been called “the Norwegian petroleum fairytale”. The construction of the Melkøya LNG processing plant based on the findings of natural gas at the Snøhvit (Snow White) exploration field had secured the petroleum industry in what seemed to be a success story that would convince decision-makers of the importance and potential of high north petroleum development, also for regional development. By 2008 the area outside the regions Lofoten, Vesterålen and Senja had already been high on the oil and gas agenda for a few years, and national, regional and local actors sought to push the case for further exploration. Politically, the case was seen as potentially threatening to the reigning Stoltenberg coalition cabinet (consisting of three political parties, two of which were very sceptical to potential petroleum development in the area). Through a combination of document and media analysis, conference participation, local dialogue and information meetings, Brigt Dale focused on the relationships between local and national understandings of risks and threats connected to the potential petroleum development of these areas, and how these could be understood in the light of debates on risk, security, knowledge production and—ultimately—questions of power and rule. His main focus was to show how both local proponents and opponents to future petroleum production in the sea area outside Lofoten experienced a divide between their concerns and discussions, and those conducted by media, politicians and scientists on the national level, a divide which ultimately influenced their sense of (in)security.

Dale's work contributed not only to a broadening of the general debate on petroleum development in the high north, but also to the theoretical debate within the security studies. He argued that the debate, as it has developed mainly after the cold war, had failed to sufficiently recognize the need for both a broadening (including more sectors of focus into the realm of the security studies) and a deepening of the concept (including more security referents beyond the state, requiring also a methodological reorientation). Dale expanded on a human security approach with the engagement of Foucauldian notions of security, biopolitics and governmentality, refocusing concerns towards matters of power, knowledge and governmental practice (Marlow 2002a, b; Elden 2007; Dean 2010). The development of the concept ontological security clarified the ways in which the relationships between identity construction, community building and security were constructed (Giddens 1990; Marlow 2002a, b; Hawkins and Maurer 2011). These contributions to security theory were further compared to the developments in the risk concept, providing a parallel debate on the relationship between individuals, the state where knowledge production has been important (Douglas 1982; Jasanoff 1986; Beck 1992).

On the basis of these theoretical considerations, Dale demonstrates how arguments concerning petroleum development were held together as coherent, logical frames of reference (Dean: op.cit) based on an adherence to specific identities. Individuals and communities ascribe ontological importance to specific actions, symbols and frames of reference (be they "in nature" or "from culture"). In other words, the way debates over a possible petroleum production has stirred discussions and reflections about who "we" are and who "we" might become, oil or not, has been particularly interesting. Identity can in this way be understood as both ontological practice (Giddens 1990) and symbolic ordering (Cohen 1985) of the world and was relevant in Lofoten for how people positioned themselves in the petroleum debate—if they were opponents, proponents or undecided—and for the way they defined how to be and what it meant to be secured. This governmentality approach to ontological security enabled an analysis of the way in which power is embedded in management structures and technologies of security, aimed at securing population, and that these efforts also creates *insecurity*. The governmental efforts of securing a future for a welfare state and thus for the Norwegian population, at least partially financed by new petroleum resources from the sea areas outside Lofoten, Vesterålen and Senja produced insecurity locally and regionally (and perhaps, some would claim, globally, as new findings would mean more climate changing emissions) as the risks involved in drilling could potentially threaten the *post-petroleum* potential of the region (Kristoffersen and Dale 2014).

A further study conducted by GAPS in Norway took a different focus, examining some of the direct ecological impacts that could in turn affect human security in local communities. This part of the project looked more broadly at the impacts of climate change, and the potential it has to expand the conditions under which toxic endophyte-infected grasses grazed by reindeer thrive. Such expansion is likely to have serious negative effects to reindeer husbandry, which is an important industry to Sámi people in Northern Norway, impacting economic, societal (identity) and environmental security for these communities. Invasive and erupting species, both indigenous and introduced, can alter how systems function at the community and ecosystem level (Myers and Bazely 2003) and have substantial social and economic impacts (Mooney and Hobbs 2000). In a warming climate, some species that spread northwards will, undoubtedly, include a number that will significantly disrupt ecosystem functioning (Mooney and Hobbs 2000), thereby threatening human well-being. One such species group that have such potential are endophytes of grass species. Grasses are a key group of plants that contain most of the world's major food (cereals) and forage (grasses) species. Many temperate and Arctic grasses can be infected by microscopic fungal endophytes which render them toxic to herbivores (Bazely et al. 1997) and also make the grass host more drought and flood resistant, consequently making the grasses able to invade new habitats. Although grass-fungal endophyte–herbivore interactions have been recognized as extremely important elsewhere (they cause huge productivity losses to the USA livestock industry), they are little studied in northern Europe (Ball et al. 1993) in spite of their importance to moose, and reindeer.

Reindeer husbandry is an important industry to Sámi people in Northern Norway and especially Finnmark. Projected climate change will likely expand the conditions under which toxic endophyte-infected grasses grazed by reindeer thrive. Because Finnmark naturally has climatically steep gradients from coast to inland and from west to east (Hanssen-Bauer 1999), presence of toxic endophyte-infected grasses along these gradients can be used as indicators of future climatic changes on plant invasibility.

Several hundred individuals of grass species from a total of 98 areas, covering 15 reindeer summer districts were sampled. The initial data obtained spoke to infection frequencies of endophytes in general. Infection frequencies of endophytes were low, and we could not document ecological patterns of infection. Hence, we found no indications of that the probability of invasibility of endophyte-infected grasses will increase in response to climate change. However, we cannot outrule that species infected with toxic endophytes show ecological patterns indicating invasibility in response to climate change. On the basis of this research, a method article critically assessing the use of the Agrinostics field immunoblot assay has been published in *Methods in Ecology and Evolution* (Jensen et al. 2011).

Case: Human Security and Extractive Industries in Russia

We cannot assume that human security is experienced and understood in identical ways across the Arctic. As in Norway, the GAPS research in Russia focused on local community experiences and perspectives, further illuminating the ways in which human (in)securities reveal power dynamics between different actors. Maria Lvova applies a multiple actor security approach to the Russian Arctic, with a special focus on the Murmansk region. Lvova's work goes to the heart of the theoretical premises for the GAPS project, in that she explores existing asymmetries within security perspectives that primarily focus on the state rather than on people, generating various difficulties related to the application of the security concept in general. Lvova examines the ways in which security is practiced and applied, often resulting in, for example, "virtuous imperialism" where security is established on the assumptions of powerful (usually state) actors, thus overlooking the "non-conventional" practices and excluding non-state actors, who have not had the power to have their voices heard, known as "silence security dilemma". Research that explores such power dynamics in the Russian Arctic demands more awareness on the context, the multiple actors involved in the security issues and not least an awareness of the methodology of research, which allows one to grasp the complexity and to show the dynamics of the processes taking place.

The project consists of two main parts. The first is designed to map the problem of energy security policy in Russia and development of the energy sector in the European North of the Russian Federation in particular. She further explores how the focus on energy affects human security considerations. While revealing the main factors influencing Russian policy in the energy sector, Lvova simultaneously explores the intersection of various understandings of the security concept. She focuses mostly on official policy documents, but also illustrates some positions through quotations from interviews. While critically examining assumptions embedded within official documents and thus shaping the security policy towards the energy sector and the North, Lvova enters the second part of her work and looks at what is left unproblematized, and to what degree interview respondents (including local populations) reflect upon gaps in policy and practice. Lvova attempts to identify those elements which are out of the official/visible picture: how are subjects constituted within energy security policy? Who is understood as responsible for the "problem"? Who is excluded and why? Why do particular explanations of security issues and security concepts take place? In Russia, security is strongly characterized by the military or state domain and therefore dominates the discourse that determines the language tools available for people, including the word "security" itself. It might be difficult for non-state actors (local populations) to express their views on security or to reflect upon their practices as a result of statism or state dominance over all spheres of peoples' life. The energy sector is strongly connected to state security which is governed by all typical connotations attached to it (military, war, treason) that people feel a lack of appropriate language and no possibility to enter the space of a discussion on security. Are there any alternatives? Do people talk about security? How then interaction is happing between the state and the individual in Russia?

Lvova's work tries to explain difficulties identified in the development of energy security thinking and the way in which "security" is operationalized in Russia by taking a step into the field of philosophy and taking a holistic view on security in general. Lvova draws insights from the work of two key figures of post-structuralism: Gill Deleuze and Michel Foucault. She assumes that existing knowledge is fragmentary and that the application of pre-made theory is not necessary. This helps to overcome debates within the security studies on the place and role of human security and also opens a new perspective on the nature of the concept of human security: it cannot be addressed based on one conventional definition, but has various forms and can as well establish various connections with other security issues and approaches. Lvova argues that this sort of human security perspective can discover the "silences" or lacunas excluded from the dominating perspective on security. With this approach, Lvova creates the theoretical tools to assess and analyse her empirical findings, enabling her to articulate security assemblages and to reflect on the nature of human security in the Arctic, through the example of the Russian case.

Lvova's work investigated security discourses in environments where the concept itself is highly problematic, enhancing theoretical work through empirical evidence in northwest Russia. At the same time, another angle investigating human security was conducted by an interdisciplinary team engaging in initial examinations into the potential health security risks associated with oil and gas activity in the Komi Republic of Russia, learning about community perspectives and how these relate to other security perspectives. Komi is a region that produces approximately 3–4 % of the oil production in Russia (Stuvøy 2011). Oil production is largely located north in the republic, but it dominates the economic development of the region, creating disparities between north and south (ibid). The northern city of Usinsk, founded in 1966 due to the location of large oil reserves in the region, is the centre for oil production, as approximately 75 % of the oil produced in Komi comes from around the Usinsk area (Ketkina 2010). In 1994, the Usinsk area was subject to a significant oil spill (in comparison with the leaks that had occurred over a period of years beforehand), which had both contaminated the region as well as increased awareness about the negative impacts of oil production to local communities (Stuvøy 2011).

In the interest of initially assessing the perceived impacts of oil and gas activity in the Komi Republic in relation to the 1994 oil spill as well as afterwards, a number of surveys and interviews were conducted in 2009 and 2010 by the Russian research team led by Dr. Evgeny Bojko and researcher Olga Ketkina at the Institute of Physiology for the Ural Division of the Russian Academy of Sciences (Syktyvkar) and analysed by Olga Ketkina and Kirsti Stuvøy (Norwegian University of Life Sciences, Norway). One of the two surveys/interviews specifically asked about impacts of oil and gas production and was conducted in two locations in the Usinsk district: Ust-Usa and Mutny Materik, covering 95 respondents. These two regions were classified as "conventionally polluted" as they were directly affected by the 1994 oil spill (Ketkina 2010). A different survey was conducted in both the "conventionally polluted" regions of Ust-Usa and Mutny Materik, as well as in "conventionally non-polluted" regions in the Izhma district (in the villages of Sizyabsk and Petrun) to assess the overall perceptions of health in these two districts (Ibid). Izhma district depends considerably upon reindeer husbandry. The reason for comparing these two districts was both to understand perceptions according to proximity to the oil and gas industry, but also to ascertain if reindeer herding is affected by migration routes that are interrupted by oil and gas installations. The initial results of both of these surveys highlighted a number of important trends occurring in the local perceptions of health and oil and gas activity impacts.[3]

The survey responses indicated almost unanimously that oil spills significantly affect the daily lives of people living in the oil and gas development regions of the Komi Republic. Some respondents cited personal health problems, others focused more on societal-level impacts. Oil spills were perceived as occurring frequently. Some of the respondents said they occurred annually, although there were great variations from respondents regarding the exact date of the last oil spill (some said just recently, last week, some last year). Respondents also felt that the media reports about spills more frequently now than earlier, but that they report mainly on major oil spills. Information about oil spills were nevertheless mostly obtained through eye witnesses, personal experiences or rumours.

Respondents expressed both positive and negative perceptions of oil and gas development. The employment opportunities afforded by the presence of the oil and gas industry was cited as the clear benefit of such activity, particularly for individuals. (Stuvøy 2011). In the villages of Ust-Usa and Mutny Materik, the "polluted" towns, many of the respondents noted that the benefits of oil and gas activity accrued to those in cities but not for those in remote villages. The responsibilities for security and the environment were additionally perceived to be shared between authorities, industries and to an extent the individual her or himself, but generally respondents emphasized an almost sole responsibility of business. This illustrates relative consensus on who should take responsibility in people's views. Negative effects of oil production that were identified by respondents vary between personal health impacts such as weakened immune systems and environmental degradation as experienced through bad fishing conditions and poor crops. The primary negative impact of oil production that was mentioned was almost unanimously the environmental impact, but this was also closely matched by reflections about the impacts on health, which were also perceived as negative: "The presence of the oil industry and the observed pollution effects imply that adverse health impacts are a constantly present insecurity" (Stuvøy 2011). The negative perceptions about general health were confirmed by the second, unrelated survey that was conducted in both of the "polluted" (Usinsk district dominated by oil and gas) and "unpolluted" villages (including Sizyabsk and Petrun in the Izhma district that is dominated by reindeer husbandry).

This initial work conducted through these surveys demonstrate the importance of including local perceptions "from the ground", in assessing the values and hopes that people in the Komi Republic expect to carry into their futures. These local perceptions must also be discussed and analysed against context and history (natural, social and physiological), against research and policy. The data we have thus far obtained are as follows: "expose how security, when assessed by individuals, incorporate both assessments of individual opportunities and negative

[3]The following presentation of the survey data is based upon the analyses conducted by Ketkina (2010) and Stuvøy (2011).

social consequences generated by pollution or unequal distribution of wealth" (Stuvøy 2011).

GAPS lastly contributed to other IPY projects, in this case MODIL-NAO (Monitoring of development of traditional indigenous land use areas in the Nenets Autonomous Okrug, Northwestern Russia) where a report was compiled based on questionnaires submitted to traditional land users in Russia (primarily reindeer herders) in the Nenets region.[4] The GAPS project contributed to the report by briefly analysing different issue areas highlighted by the respondents, in the light of human security. Part of the intention of MODIL-NAO was to provide the people of Nenets with solid data from which they can better articulate their interests and have their voices heard in the political decision-making processes. This in turn assists improved accountability on the part of the state and oil companies towards indigenous populations in this area, particularly with regard to shared land use between oil and gas activities and reindeer herding and other "traditional" economy activities. On the basis of the data obtained through the MODIL-NAO interviews, the report's chapter on human security evaluated processes of security and insecurity through the following indicators: legislation, consultation/participation, environment, quality of life and culture, economics and energy. These factors were derived from the results of the interviews themselves as those which were identified as some of the most prevalent values for these communities. The analysis therefore weighs the various statements and positions of the different NAO respondents, the oil industry, researchers and the state (through legislative practices) to arrive at an understanding of the security dynamics in the region, and what this potentially means for the future local security of the region. The dialogue between researchers and community members, and their subsequent interactions with industries and state, informed the direction of the analysis. The report can be found on the MODIL-NAO Website (see Footnote 6).

Moving Forward Across Disciplines Within a Human Security Framework

The momentum created by the IPY GAPS project contributed to a widening interest to engage (if not entirely agree) with the human security concept in the Arctic. The edited volume, "Environmental and Human Security in the Arctic" published by Routledge in 2014, had its roots in the initial meeting in Tromsø in 2004 and drew on work from a wide range of contributors including authors involved in GAPS early on, but also contributors beyond the GAPS project itself. This book is an introduction to some of the initial ideas around the potential and challenges of a more complex, widened understanding of security in the Arctic. The book serves as a forum, in which a diverse group of authors contribute their thoughts, experiences

[4]MODIL-NAO Website: http://ipy-nenets.npolar.no/main%20pages/frame.html.

and research results that have a bearing upon understanding security in the Arctic. The contributors to this book come from different disciplines ranging from political science and philosophy to engineering and ecology; different nationalities including Russian, Canadian, Norwegian, American, Finnish, as well as indigenous and non-indigenous. This results in a combination of works that understand security from different vantage points. Some authors concentrate mostly on the concept of security itself, while for others it lies in the background of the topic they are exploring. Taken together, the book provides one glimpse into an emerging security dynamic that is developing in the Arctic and how this is discursively conceptualized and practically developed, organized and delivered by multiple security actors, both state and non-state.

The book takes on such diverse topics as a securitization theory approach to analyse the ways in which Russian security politics has developed in relation to the Svalbard archipelago, demonstrating that Russian attitudes have not changed much despite the fall of the Soviet Union, to arguments for a holistic picture of security in the Arctic that is accompanied by human and environmental security perspectives. The concept of human security itself and its possible relevance to the Arctic is also challenged noting assumptions embedded within this concept that have led to a type of "virtuous imperialism" where northern states were perceived as having the capacity to both identify as well as rectify human security concerns in the global South, endowing the South with the benefit of northern skills and knowledge. To prevent Arctic human security falling into this typical trap, it is argued that we can learn from indigenous scholars and feminist debates in the region. To this end, the book touches on the relevance of self-government, practices of local governance and co-management of resources in indigenous communities, as well as explorations into links of human security to political representation, communication versus consultation, and women's voices, both in indigenous communities and non-indigenous, including insights into violence against women, democracy development on the local level and attitudes towards sex (particularly through prostitution and trafficking) in Russia.

Health security is highlighted as an important aspect of Arctic human security, where technological innovations such as telemedicine to improve access for people living in remote communities. Cooperation in the Arctic around health can still be a challenge, however, due to different cross-cultural perceptions of the notions of "health risks", "health care" and "treatment" demonstrating the "virtuous imperialism" problem with the human security concept if the concept refers to a so-called competent security provider (developed country) providing services and skills to an assumed "security recipient" (developing country).

Further chapters examine the intricate linkages between social systems and ecosystems demand a rethinking of the way that we understand and guide or govern these systems if our end goal is sustainability and examines the vulnerability and adaptability of social systems to abrupt changes in climate, as well as linking changes to the environment in the Arctic to adaptations and improvements in engineering and technology. Drawing on the experience of the GAPS project, the book also addresses the interdisciplinary nature of human and environmental security concepts, discussing the ways in which the human security concept can be useful to natural scientists to make their work relevant to the social and political world. As the authors note, there is no question that there remains significant challenges in the ability for social scientists and natural scientists to collaborate effectively, but suggest that a human security framework that recognizes the interconnections between social systems and ecosystems provides a solid foundation for interdisciplinary cooperation and research.

The accumulating body of research into human vulnerabilities and adaptations to changing environments in the natural and social sciences has, many argue, failed to be transferred into effective action on the part of policymakers (Shellenberger 2004). "It appears that we care very little about the health threats that pollution of the Arctic represents … research on the Arctic receives little attention" (Anderssen and Gabrielsen 2005) Directing research knowledge towards an existing and specific policy community (human security), therefore, is essential and integral to effective policy action. GAPS not only challenges predominant, state-centric conceptualizations of human security, but it also addresses security issues across a range of disciplines and approaches human well-being from a variety of interlinked and overlapping angles. The human security framework demands input from a variety of academic disciplines; thus, the GAPS research team included experts in ecology, community health, ecotoxicology, geography, sociology, physiology and political science. The IPY GAPS project demonstrated that it is imperative to be interdisciplinary in our research approach in order to integrate the wide range of factors that comprise human security, e.g. economic, food, health, environmental, personal, community and political security (UNDP 1994).

By taking the community as our starting or referent point, GAPS adopted a bottom-up approach that aims to effectively involve Arctic communities, as well as explicitly articulate the need for interdisciplinary research on the impacts of change in the Arctic: "She sits at the back of the hall, listening to experts from far away talking in a language not her own about the fate of the bush she has roamed all her life" (Weber 2006). Research cannot be composed of "far away experts". The GAPS research team has made communicating and collaborating with communities and local organizations an integral and self-evident part of the research process and pays active attention to the well-articulated need for comprehensive, participatory research on the impacts of oil and gas development in Arctic communities. Among lessons learned in the GAPS process:

1. We have conceptualized human security as a democratizing, process with multiple local outcomes.
2. We have brought human security thinking to the invasive species and ecotoxicology fields showing that non-indigenous, potentially invasive species are already present in the region, as well as continue to learn about the relationships between human health and industrial toxins.
3. We have learned how and why interdisciplinary collaborative research is uncomfortable and how there are different kinds and types of interdisciplinary collaborations (often these key distinctions are NOT made by researchers who

consider themselves to be doing interdisciplinary, collaborative research) and that the language of our disciplines and methodological assumptions can be large barriers to collaboration.

4. We learned about how interdisciplinary, collaborative team based on knowledge movement and open access thinking can support goals of human security.
5. We have been contributing ecological thinking to the group research.[5]
6. We have increased our commitments to open access in support of knowledge as a nutrient for human security.
7. We have expanded the intersection of ecology–policy–politics relating to climate change adaptation and human security frameworks.

Arctic oil and gas development and its impacts on peoples and the environment, when analysed from a securities perspective, can be seen as having both positive and negative attributes, generating risks and threats as well as opportunities. The human-/individual-based security framework provides a vehicle for the expression of threats and vulnerabilities and adaptabilities from the grass-roots level. As the GAPS legacy moves forward, not only are predicted and predictable, discipline-based findings emerging, but also new kinds of knowledge and awareness are emerging as a consequence of framing engagement with different communities explicitly within the human-based security framework.

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[5] Note that this subsequent thinking is being published in newer book chapters by Bazely et al. (2015)

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Chapter 11
IPY BearHealth: Polar Bear (*Ursus maritimus*) Circumpolar Health Assessment in Relation to Persistent Pollutants and Climate Change

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Keywords Polar bears · Arctic · Persistent contaminants · Effect studies · Climate change

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The polar bear, its exposure to contaminants and the possible effects was the main focus of BearHealth during IPY 2009–2011.

Introduction—Purpose and Background

In addition to mercury (Hg), an array of chlorinated, brominated, and fluorinated persistent organic pollutants (POPs) have since the 1850s and 1940s, respectively, been transported to the Arctic where they accumulate in biota (de March et al. 1998; de Wit et al. 2004; Dietz et al. 2009). More specifically, the 1850s marked the period of early industrialization and was associated with elevated emissions of Hg, whereas around the 1940s marked a period when large-volume worldwide use of industrial chemicals and pesticides commenced. These POPs and Hg are transported via global atmospheric and oceanic pathways and processes that result in deposition in the Arctic and are found in Arctic endothermic top predators, in particular in polar bears. Because POPs are lipophilic, and because lipids constitute an important energetic factor in polar biota, POPs are biomagnified in the long Arctic marine food chains. As a result, POP levels are very high in polar bears in spite of their relatively high ability to biotransform known lipophilic POP compounds (Letcher et al. 2010; Sandala et al. 2004; Verreault et al. 2005). These long-range transported POPs possess known or suspected toxicities including endocrine disruption leading to, e.g., immune and reproductive effects that may affect survival and reproduction of polar bears (Letcher et al. 2010; Sonne 2010; Sonne et al. 2012a, b). However, not all POPs necessarily exert "toxic" effects (known or unknown), and even if a POP is known to exert biologic effects, it is not necessarily toxic effects.

The background for the IPY BearHealth project was based on multiple reports that the high levels of POPs and Hg in polar bears could affect the health and reproduction of polar bears. About 10 years ago, high levels of POPs in male polar bears from Svalbard had been linked to reduced levels of plasma concentrations of testosterone (Oskam et al. 2003), while in female Svalbard polar bears, positive association between plasma levels of PCBs and progesterone had been reported (Haave et al. 2003). Also, negative associations between plasma levels of POPs and cortisol had been reported in both male and female Svalbard polar bears (Oskam et al. 2004). In East Greenland male polar bears, negative relationships between various groups of POPs and testes length and baculum length/weight and baculum bone mineral density had been reported (Sonne et al. 2004, 2006, 2007). In females, negative relationships were reported between various groups of POPs and ovary length/weight and uterine horn length (Sonne et al. 2006). Associations between plasma levels of PCBs and plasma thyroid concentrations had also been reported (Skaare et al. 2001; Braathen et al. 2004), as well as a negative association between PCBs and plasma levels of retinol (vitamin A) (Skaare et al. 2001). Furthermore, experimental field studies had strongly indicated that high levels of legacy POPs disrupted the immune system of polar bears (Lie et al. 2004, 2005). It had also been reported that the bone mass density in skulls of polar bears from East Greenland sampled in the period of POP use (1966–2002) was lower than that in skulls sampled prior to this period (1892–1932) (Sonne et al. 2004) and that POP levels were associated with kidney lesions in East Greenland polar bears (Sonne et al. 2005). Finally, a decade ago, it had been suggested that in the Svalbard population of polar bears, a low number of females older than 15 years with cubs could be due to reproductive impairment of females, lower survival rates of cubs, or increased mortality of reproductive females caused by the high levels of POPs in this population (Derocher et al. 2003).

Polar bears are distributed throughout the circumpolar region, have unquestionable importance to northerners both culturally and economically, and can thus be considered as an important sentinel or monitoring species for POPs and Hg. The IPY BearHealth project was based on the fact that levels of POPs are generally high in the polar bear, making it an ideal wildlife receptor for biomonitoring of spatial and temporal trends, distribution, dynamics, fate, biomagnification and potential effects of Hg and legacy and emerging POPs. Furthermore, tissue samples of polar bears were readily available in a circumpolar context due to their distribution range, the indigenous hunting, and live captures for satellite tracking.

The aim of the IPY BearHealth project was to conduct a more recent circumpolar health assessment of polar bears in relation to POP exposure. This was strengthened and complemented by the fact that polar bear scientist collaborations through the

IUCN Polar Bear Specialist Group and Arctic Monitoring and Assessment Programme (AMAP) resulted in early and continued trans-Arctic monitoring and assessment of contaminants and have served as an exemplary collaborative model for research and monitoring on other species as well.

Arctic Warming

Arctic ecosystems face multiple challenges at local and regional scales. Among these are the potential stress caused by climate changes and exposure to anthropogenic chemical contaminants. An indicator of the urgency of these combined changes is the rate at which Arctic POP data, modeled along with climate data, have been produced during the past 15 years. During the 1990s, contaminants were identified as the major risk to ecosystems and northern food security (de March et al. 1998). More recently, the warming of the Arctic has been signaled by the loss of multiyear sea ice and thawing of permafrost and accelerated coastal erosion (Olsen et al. 2011). However, depending on the region, Arctic warming also affects the pathways, transport, uptake, food chains, migration routes, as well as storage and mobilization of stored contaminants within the Arctic ecosystem (Jenssen et al. 2015; Macdonald et al. 2003, 2005; Macdonald and Loseto 2010: McKinney et al. 2015). Other factors of importance include overfishing, hunting, habitat destruction, increased traffic, and increased interest in resource exploitation (e.g., oil and gas) as potential future consequences of change.

Over the last decade or so, it has become widely recognized that combined, POPs/Hg and climatic warming are putting Arctic ecosystems at risk (Macdonald et al. 2005; Macdonald and Loseto 2010). Despite national bans of the use of POPs beginning in the 1970s and later, the ratification of the UNEP Stockholm Convention in 2004, which curtailed or restricted primary emissions of POPs including pesticides and industrial organohalogen compounds (OHCs), we continue to see cycling within Arctic systems of almost every chlorinated POP released in significant amounts during the past 50 years as a result of industrial and agricultural activity. One example of increased POP exposure around year 2000 was for East Greenland polar bears where there was a documented shift in diet from ringed seals (*Pusa hispida*) to greater consumption of more highly POP-exposed harp (*Pagophilus groenlandicus*) and hooded seal (*Cystophora cristata*) (Dietz et al. 2013a, b; McKinney et al. 2013). Likewise, significant quantities of Hg released as a result of human activities have ended up in the Arctic (AMAP 2011). OHCs and Hg are two types of contaminants that behave very differently in the Arctic environment, although both tend to put top predators at risk including polar bears, other wildlife, and native northerners who depend on aquatic food webs for much of their diet (AMAP 2010, 2011).

Geographic Trends of Contaminants

POPs

During the IPY period (2007–2009) and until present, IPY BearHealth studies have examined more recent POP spatial trends. For example, McKinney et al. (2011a) investigated several brominated flame retardants (BFRs) and legacy contaminants in adipose tissue of bear (2005–2008) from 11 circumpolar subpopulations spanning Alaska east to Svalbard. For legacy OCs and polybrominated diphenyl ethers (PBDEs), such trans-Arctic comparisons had previously been conducted by Norstrom et al. (1998), Verreault et al. (2005) and Muir et al. (2006). Although 37 PBDE congeners, total-(α)-hexabromocyclododecane (HBCDD), 2 polybrominated biphenyls (PBBs), pentabromotoluene, pentabromoethylbenzene, hexabromobenzene, 1,2-bis (2,4,6-tribromophenoxy(ethane), and decabromodiphenyl ethane were the screened BFRs, only 4 PBDE congeners (BDE-47 and BDE-153 and lesser proportions of BDE-99 and BDE-100), total-(α-) HBCDD, and BB153 were consistently detected. Geometric mean ΣPBDE (4.6–78.4 ng/g lipid weight (lw)) and BB-153 (2.5–81.1 ng/g lw) levels were highest in East Greenland (43.2 and 39.2 ng/g lipid weight (lw), respectively), Svalbard (44.4 and 20.9 ng/g lw), and western (38.6 and 30.1 ng/g lw) and southern Hudson Bay (78.4 and 81.1 ng/g lw). Total-(α)-HBCDD levels (0.3–41.1 ng/g lw) were lower than Σ_4PBDE levels in all subpopulations except in Svalbard, consistent with greater European HBCD use versus North American pentaBDE product use. ΣPCB levels were high relative to these BFRs and other legacy contaminants and increased from west to east (1797–10,537 ng/g lw). Σ-chlordane (ΣCHL) levels were highest among legacy OC pesticides and relatively spatially uniform (765–3477 ng/g lw). ΣDDT levels were relatively low and spatially variable (31.5–206 ng/g lw). However, elevated proportions of *p,p′*-DDT to ΣDDT in Alaska and Beaufort Sea relative to other subpopulations suggested fresh inputs from vector control use in Asia and/or Africa.

Among polar bear subpopulations we investigated the relative effect of dietary differences versus the regional geographic influence the OHC accumulation and exposure patterns (McKinney et al. 2011b). Dietary variation between Alaska, Canada, East Greenland, and Svalbard subpopulations was assessed by muscle nitrogen and carbon stable isotope (δ^{15}N, δ^{13}C) and adipose fatty acid (FA) signatures relative to their main prey (ringed seals). Western and southern Hudson Bay signatures were characterized by depleted δ^{15}N and δ^{13}C, lower proportions of C_{20} and C_{22} monounsaturated FAs and higher proportions of C_{18} and longer chain polyunsaturated FAs. East Greenland and Svalbard signatures were reversed relative to Hudson Bay. Alaskan and Canadian Arctic signatures were intermediate. Between subpopulations, dietary differences predominated over interannual, seasonal, sex, or age variation. Among various brominated and chlorinated contaminants, diet signatures significantly explained variation in adipose levels of Σ_4PBDE (14–15 %) and legacy ΣPCBs

(18–21 %). However, dietary influence was contaminant class-specific, since only low or nonsignificant proportions of variation in organochlorine pesticide (e.g., ΣCHL) levels were explained by diet. Hudson Bay diet signatures were associated with lower ΣPCB and ΣPBDE levels, whereas East Greenland and Svalbard signatures were associated with higher levels. It was concluded that understanding diet/food web factors is important to accurately interpret contaminant trends, particularly in a changing Arctic.

The Arctic marine environment has the capacity to store large amounts of all sorts of contaminants and that the manner in which these compounds enter the ocean or food webs depends strongly on ice cover. As recently reported in McKinney et al. (2012), OHC dynamics in arctic marine food webs may be impacted by current climate-induced food web changes including increases in the transient/subarctic seal species and polar bears. Food web OHC transfer was quantified in the Cumberland Sound arctic marine food web in the presence of transient species using species-specific biomagnification factors (BMFs), trophic magnification factors (TMFs), and a multifactor model that included δ^{15}N-derived trophic position and species habitat range (transient versus resident) and also considered δ^{13}C-derived carbon source, thermoregulatory group, and season. Transient/subarctic species were found to have higher tissue OHC levels and greater BMFs likely due to higher energetic requirements associated with long-distance movements or consumption of more contaminated prey in regions outside of Cumberland Sound.

There is an urgent need for updated data on circumpolar spatial distribution of previously investigated and new and emerging environmental pollutants in polar bears that include the Russian subpopulations. Because of the high capacity of polar bears to biotransform lipophilic compounds, polar bears also have high levels of metabolites of POPs (Sandala et al. 2004; Verreault et al. 2005; Letcher et al. 2010; Bytingsvik et al. 2012a). Paradoxically, these water-soluble metabolites are often more toxic than their mother compounds and may thus have toxic effects, particularly on the thyroid hormone system and on toxic processes induced via oxidative stress that is related to higher metabolic rates (Brouwer et al. 1998). There is a lack of information on to which extent these metabolites contribute to the adverse health effects in the Arctic Inuit population and wildlife.

Mercury

Approximately 200–300 tons of mercury (Hg) is annually transported to the Arctic from lower latitudes via the atmosphere and large-scale ocean currents (de March et al. 1998; AMAP 2005). The Arctic acts as a sink for global emitted Hg due to spring mercury depletion events (Berg et al. 2001; Lindberg et al. 2001; Lu et al. 2001; Schroeder et al. 1998). As for POPs, Hg concentrates in the long Arctic marine food chains (de March et al. 1998). Although mercury is a naturally occurring element, Dietz et al. (2009) showed that up to 94 % of the mercury in Greenland polar bears is of anthropogenic origin. Recently, liver and kidney histology and mercury concentrations in East Greenland polar bears have been examined, which have indicated a number of linkages with histopathological changes.

Studies of polar bears from East Greenland have shown that they are among the most Hg-contaminated species in the Arctic (de March et al. 1998; AMAP 2005). Under the IPY BearHealth project, investigations between 2005 and 2008 of bears sampled from a range spanning the Bering Sea to Greenland (Routti et al. 2011) showed that the Beaufort Sea and adjacent Lancaster/Jones Sound and Gulf of Boothia in the Canadian Arctic had the highest liver concentrations of total Hg (THg). The observed geographic pattern of Hg levels in these polar bears was robust and may have been caused by a number of, as yet, undetermined factors. An obvious candidate for the source of high Hg in marine mammals from the Beaufort Sea is the Mackenzie River, which is estimated to deliver 2.2 tonnes of THg and 15 kg methyl-Hg (MeHg) annually to the estuary (Leitch et al. 2007). However, food web structure and feeding behavior cannot be neglected as potential contributors for the observed geographic patterns for Hg in polar bears (Cardona-Marek et al. 2009; Horton et al. 2009). Therefore, there remains an enigma which of these factors contribute to the robust observation of high Hg in bears of the western Arctic (North America). Recent change in sea ice climate for this region further complicates prediction and understanding.

Based on the measurements of $\delta^{15}N$ (Routti et al. 2012), THg concentrations in polar bear liver increase with apparent trophic feeding level, which is consistent with previous studies reporting biomagnification of Hg for other wildlife (Gaden et al. 2009; Loseto et al. 2008; Muir et al. 2005). Adjusting trophic position of the polar bears reduced the observed geographic variation in liver THg concentration among subpopulations to the point that many of the apparent geographic differences were no longer statistically significant (Routti et al. 2012). What this implies is that food webs, and not regional sources of Hg, likely provide much of the cause for geographic variation. Interestingly, Routti et al. (2012) showed that liver THg concentrations in bears were negatively correlated with muscle $\delta^{13}C$ ratios, suggesting that river inputs of terrestrial carbon, with associated Hg enrichment, may also provide a component of regional variation.

Temporal Trends of Contaminants

POPs

As part of the IPY BearHealth project, Dietz et al. (2012a) determined legacy organochlorine contaminants in adipose tissues from polar bears sampled in East Greenland between 1983 and 2010. Of twenty major legacy contaminants and congeners (ΣPCB, 4 PCB congeners (CB-153, CB-180, CB-170/190), ΣDDT (*p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT), α- and β-hexachlorocyclohexane (HCH), HCB, octachlorostyrene, dieldrin, oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, heptachlor epoxide, and BB-153), nineteen showed statistically significant average yearly declines of −4.4 % (range −2.0 to −10.8 %/year) among subadult polar bears (i.e., females <5 years, males <6 years). Similar but fewer statistically significant trends were found for adult females and adult males likely due to smaller sample size and years. The authors concluded that: "*Despite declines as a result of international regulations, relatively high levels of these historic pollutants persist in East Greenland polar bear tissues*" (Fig. 11.1).

As part of the IPY BearHealth project, Dietz et al. (2012b) determined several BFRs in adipose tissues from the East samples between 1983 and 2010. Significant linear increases were found for Σ_4PBDE (BDE-47, BDE-99, BDE-100, and BDE-153), BDE-100, BDE-153, and HBCDD. Average increases of 5.0 % per year (range 2.9–7.6 %/year) were found for the subadult polar bears. BDE-47 and BDE-99 concentrations did not show a significant linear trend over time, but rather a significant nonlinear trend peaking between 2000 and 2004. Similar but fewer statistically significant trends were found for adult females and adult males likely due to smaller sample size and years. These increasing concentrations of organobromine contaminants contribute to complex POP mixture, already causing health effects to the East Greenland polar bears (Fig. 11.2).

McKinney et al. (2013) reported on how rapid climate changes that are occurring in the Arctic are having substantial repercussions on the diets of upper trophic level consumers and specifically for polar bears from East Greenland. Quantitative fatty acid signature analysis (QFASA) and fatty acid carbon isotope (δ^{13}C-FA) patterns were examined to assess diets of East Greenland polar bears over a three-decade period (1984–2011). QFASA-generated diet estimates over the study period

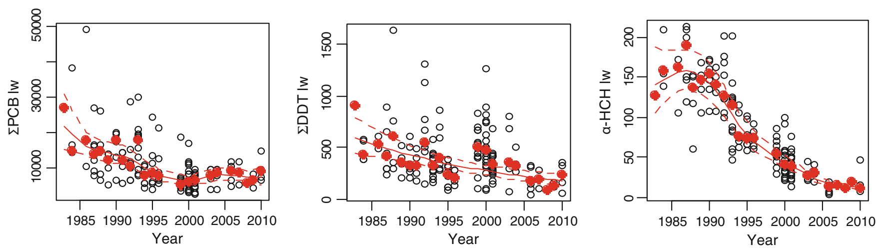

Fig. 11.1 Log-linear temporal trends of selected legacy organochlorine contaminants from juvenile East Greenland polar bears (see Dietz et al. (2013a) for further information; data in ng/g lipid weight). The *filled red dots* are median values. *Red lines* indicate significant trends and dotted line the 95 % confidence intervals

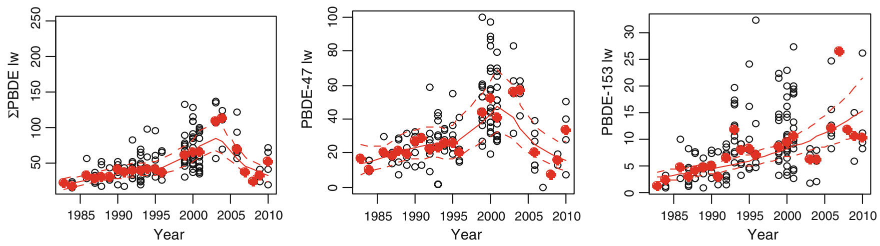

Fig. 11.2 Log-linear temporal trends of selected brominated flame retardants from juvenile East Greenland polar bears (see Dietz et al. 2013b for further information; data in ng/g lipid weight). Graphics described in Fig. 1

indicated that these polar bears mainly consumed Arctic ringed seals (47.5 ± 2.1 %) and migratory subarctic harp (30.6 ± 1.5 %) and hooded (16.7 ± 1.3 %) seals. Bears rarely, if ever, consumed bearded seals, narwhals, or walruses. Over the 27-year period, ringed seal consumption declined by 14 % per decade and hooded seal consumption increased 9.5 % per decade. The declining δ^{13}C-FA ratios supported a shift from more nearshore/benthic/ice-associated prey to more offshore/pelagic/open water-associated prey. The QFASA-generated diet estimates were consistent with diet changes predicted by climate changes, where hooded seal and ringed seal consumption increased and decreased, respectively, and occurred during years when the North Atlantic Oscillation (NAO) Index was lower. Presently, we are in the process of linking further changes in the polar bear health scenario to climate parameters in the East Greenland area. Also under IPY BearHealth, we showed that sea ice, dietary changes, and OHC contaminant patterns change linkages for legacy OCs and newer PBDE POPs in polar bear from the western Hudson Bay subpopulation (McKinney et al. 2009, 2010).

In the first years of the IPY BearHealth project, Dietz et al. (2008) determined subadult polar bears within the period 1984–2006 for PFASs (i.e., PFSAs including PFOS [perfluorooctane sulfonate]) and perfluorinated carboxylates [PFCAs]). Linear regression analysis of log-transformed median concentrations showed significant annual increases for PFOS (4.7 %), PFNA (perfluorononanoic acid, 6.1 %), PFUnA (perfluoroundecanoic acid, 5.9 %), PFDA (perfluorodecanoic acid, 4.3 %), PFTrA (perfluorotridecanoic acid, 8.5 %), PFOA (perfluorooctanoic acid, 2.3 %), and PFDoA (perfluorododecanoic acid, 5.2 %). By 2006, concentrations of ΣPFASs exceeded the concentrations of all conventional POPs, of which several have been documented to correlate with a number of negative health effects. In addition, the rapidly increasing concentrations of PFASs are likely to cause cumulative and combined effects on the polar bear, compounding the already detected threats from OHCs. By including polar bear samples collected during the IPY years in combination with recent ringed seal samples, Rigét et al. 2013 documented updated time series of PFASs in East Greenland polar bears, and East and West Greenland ringed seals were updated in order to deduce whether a response to the major reduction in perfluoroalkyl production in the early 2000s has occurred.

Previous studies by Bossi et al. (2005) and Dietz et al. (2008) had documented an exponential increase of PFOS in liver tissue from both ringed seals and polar bears from Greenland. However, Rigét et al. (2013) documented that PFOS concentrations increased up to 2006 with doubling times of approximately 6 years for the two ringed seal populations and 14 years in case of polar bears. Since then, a rapid decrease occurred with concentrations showing half-lives of approximately 1, 2, and 4 years, respectively. In polar bears, perfluorohexane sulfonic acid (PFHxS) and perfluorooctane sulfonamide (PFOSA) also showed decreasing trend in recent years as do perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnA), and to some extent perfluorooctanoic acid (PFOA). For the West Greenland ringed seal population, PFOA, perfluorononanoic acid (PFNA), PFDA, and PFUnA appeared to have peaked in the mid-2000s, whereas PFNA, PFDA, and PFUnA in the East Greenland population have been stable or even increasing in the recent years. The peak of PFASs in Greenland ringed seals and polar bears occurred at a later time than in Canadian seals and polar bears (Smithwick et al. 2005; Butt et al. 2007), considerably later than observed in seal species from more southern latitudes. Rigét et al. (2013) suggested that this could be explained by the distance to emission hot-spots and differences in long-range transport to the Arctic.

In Svalbard polar bears, levels of POPs in mother–cub pairs in 1998 and 2008 were compared. In BearHealth, we documented that levels of ΣPCBs, ΣOH-PCBs, and most PFOS were significantly reduced from 1998 to 2008. In lactating mothers, plasma levels of Σ_{21}PCB, Σ_6OH-PCBs, and Σ_2PFSAs (i.e., PFHxS, PFOS), respectively, were approximately 55, 65, and 28 % lower in 2008 than in 1998 (Bytingsvik et al. 2012a, b). In suckling cubs-of-the-year, plasma levels of Σ_{21}PCB, Σ_6OH-PCBs, and Σ_2PFSAs, respectively, were approximately 59, 50, and 21 % lower in 2008 than in 1998 (Bytingsvik et al. 2012a, b). In addition, levels of two the short-chained PFCAs, PFHpA (perfluoroheptanoic acid) and PFOA, were lower in 2008 than in 1998 (Bytingsvik et al. 2012b). However, on the contrary, levels of most PFCAs (i.e., PFNA, PFDA, PFUnDA, PFDoDA [perfluorododecanoic acid], and PFTrDA) were detected in higher concentrations in 2008 than in 1998. Thus, plasma levels of Σ_7PFCAs were approximately 39 and 50 % higher in mothers and cubs, respectively, in 2008 than in 1998 (Bytingsvik et al. 2012b). Because the level of several PFASs in mothers and cubs from both sampling years exceeded the levels associated with health effects in humans (see Bytingsvik et al. 2012b), the findings raise concern on the potential health effects of PFASs in polar bears.

Mercury

To investigate the increase of Hg in polar bears over the last 100 years, Dietz et al. (2011a) analyzed THg in polar bear hair, as part of the IPY BearHealth project, from Northwest Greenland polar bears hunted during 1892–2008. THg concentrations showed yearly significant increases of 1.6–1.7 % ($p < 0.0001$) from 1892 to 2008, and the two most recent median concentrations from 2006 to 2008 were

23- to 27-fold higher, respectively, than the baseline level from 1300 A.D. in the same region. This indicated that during the 2006–2008 period, Northwest Greenland polar bear THg exposure was 95.6–96.2 % anthropogenic in its origin. The authors could not find a change in trophic levels over time, so change in feeding pattern could not explain the change in Hg exposure. The neurochemical effect level of 5.4 μg THg/g dw proposed for East Greenland polar bears was exceeded in 93.5 % of the Northwest Greenland polar bears. It was concluded that these results call for detailed effect studies in main target organs such as brain, liver, kidney, and sexual organs in the Northwest Greenland polar bears.

The polar bear THg time trend results reported by Dietz et al. (2011a) were in accordance with an earlier review study by Dietz et al. (2009). The latter study examined the literature concerning the long-term changes of Hg in humans and selected Arctic marine mammals and birds of prey since preindustrial times (i.e., before 1800 A.D.), to determine the anthropogenic contribution to present-day Hg concentrations and the historical timing of any changes. The authors found a consistent change trend of THg, where concentrations increased by an order of magnitude in hard tissues of Arctic marine animals beginning in the mid- to late-nineteenth century and with the rate of increase accelerating in the twentieth century. The median man-made contribution to present-day Hg concentrations was 92.4 % ranging from 74.2 to 94.4 %. No generally consistent trend was evident across tissues and species from the circumpolar Arctic during the last 30 years or so (Rigét et al. 2011). However, there was a clear west-to-east gradient in the occurrence of recent increasing Hg trends, with larger numbers and a higher proportion of biotic datasets in the Canadian and Greenland regions of the Arctic showing significant increases than in the North Atlantic Arctic. Most of the increasing datasets were for marine species, especially marine mammals (Rigét et al. 2011) (Fig. 11.3).

As already discussed, the food web provides further complexity when Hg (temporal) trends in high trophic animals such as polar bears are examined. Very recent temporal trends for THg in polar bear livers have been shown to differ among populations in the circumpolar Arctic (Routti et al. 2011). However, this is true also from previous studies, where for example, for Beaufort Sea bears showed a slight increase between the 1980s and 2002 (Rush et al. 2008) and with no change from 2002 up to 2008 (Routti et al. 2011). A slight increase in THg was observed in bears from other subpopulations and areas of the Canadian Arctic when comparing data from 2002 with the 1980s (Rush et al. 2008). The most recent data indicate that Chukchi and Canadian Arctic (excepting the Beaufort Sea) subpopulations have declined relative to levels in 2002. Subpopulation like the Lancaster Sound has shown dramatic increases, which is not the case for the Hudson Bay or Svalbard (Dietz et al. 2012c).

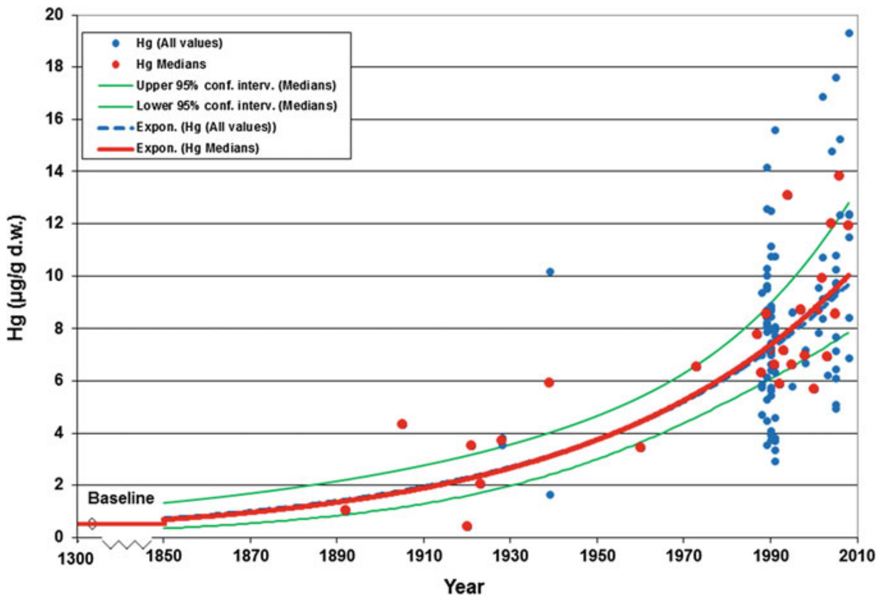

Fig. 11.3 Mercury concentrations showed yearly significant increases of 1.6–1.7% ($p < 0.0001$) from 1892 to 2008 and the two most recent median concentrations from 2006 and 2008 were 23- to 27-fold higher respectively than baseline level from 1300 A.D. in the same region (Nuullit). This indicates that the present (2006–2008) Northwest Greenland polar bear Hg exposure is 95.6–96.2% anthropogenic in its origin. (*Source* Dietz et al. 2011a)

Effects of Contaminants

In the Arctic, there is concern about the high POP and Hg levels reported in indigenous people who consume blubber and meat of marine mammals and in endothermic top predators such as polar bears, glaucous gulls (*Larus hyperboreus*), ivory gulls (*Phagophila eburnea*), white whales (*Delphinapterus leucas*), narwhals (*Monodon monoceros*), and killer whales (*Orcinus orca*). In humans, high levels of POPs, such as dioxins, PCBs, and organochlorine pesticides (OCPs), have been associated with reduced birth weights, thyroid hormone imbalance, and adverse effects on learning, cognitive, and motor abilities (Zoeller 2005). In wildlife, similar effects have been reported on thyroid imbalance, as well as effects on reproductive hormones, on reproduction per se, and probably also on survival (Jenssen 2006). Thus, the concerns that populations and ecosystem functioning may be severely affected by anthropogenic pollution are scientifically based. In polar bears, levels of POPs are reported to correlate with a number of ecologically relevant biomarker endpoints such as bone density and pathology, histology of immunological organs, renal lesions, liver morphology, immune function, thyroid hormones, reproductive hormones, and vitamin A status (see Letcher et al. 2010; Sonne 2010 and references therein), abd links to exposure assessments and human health perspectives from long-range transported pollution can have more far-reaching consequences. For example, Sonne et al. (2006) suggested that the size of reproductive organs in East Greenland polar bears is reduced by POP exposure. Coincidently, Tiido et al. (2006) had reported a similar correlative relationship in Inuit people from Greenland.

Recent data, and mainly results from the IPY BearHealth project, continue to show that relative to other regions, polar bears from Svalbard and East Greenland have the highest contaminant loads. Therefore, most effect studies have been on bears from these exposure "hotspot" areas. At Svalbard, the extensive tagging programs on the protected polar bears have revealed important information from free-ranging polar bears. In East Greenland, there is a unique opportunity to collect samples from the entire animal including internal organs via the subsistence hunting (see photograph below), which is not possible at, e.g., Svalbard where the polar bear is protected.

In Scoresby Sound, East Greenland, the Inuit have a quota of 35 polar bears that can be hunted every year. These bears have been monitored for contaminants over the last three decades, which constitutes the best Arctic time series.

POPs

Letcher et al. (2010) and Sonne et al. (2012a) have very recently reviewed the body of effect studies of contaminants on East Greenland polar bears. In terms of effects, it was concluded that bone density seemed to decrease as a function of time and POP concentration levels. Furthermore, T-scores for adult males indicated risk for osteoporosis, and the size of sexual organs likewise decreased with increasing OHC concentrations. Physiologically based pharmacokinetic (PBPK) modeling on reproduction showed that the risk quotients were ≥ 1 for ΣPCB, dieldrin, and PFOS, which indicate an increased risk of prenatally reproductive pathology (Sonne et al. 2009). Dietz et al. (2015) carried out a risk quotient (RQ) evaluation on OHC-exposed polar bears harvested from 1999 to 2008 and from 11 circumpolar subpopulations spanning from Alaska to Svalbard. Here the total additive RQ from all OHCs ranged from 4.3 in Alaska to 28.6 in East Greenland bears for effects on reproduction, immune health and carcinogenicity, highlighting the important result that the toxic effect threshold (i.e. RQ > 1) was exceeded for all polar bear populations assessed.

IPY BearHealth studies included skull BMD and length size measurements in relation to POP levels in East Greenland polar bears over the period of 1892–2010 (Sonne et al. 2013). Although the exact mechanisms are not known, studies have suggested a decrease in, e.g., dieldrin-mediated disruption of cortisol, parathormone, sexual hormones, and vitamin homeostasis (Bechshøft 2011; Bechshøft et al. 2012a, b; Sonne 2010; Sonne et al. 2004, 2012a, b). These disruptions could affect the BMD through increases in bone resorption and decreased bone formation as seen in other species followed similar long-term OHC exposures (Lind et al. 2003, 2004; Roos et al. 2010). It is therefore likely that OHC exposure is one of the driving factors behind the BMD decrease in East Greenland polar bear males during the period studied. Regarding CBL, the decrease may reflect a modulation on skull growth and development (Lind et al. 2004; Manolagas et al. 1995; Pertoldi et al. 2009; Selye 1973; Sonne 2010; Sonne et al. 2004, 2012a, b).

During the IPY BearHealth study period, thyroid gland histology was examined in twenty East Greenland polar bears (Sonne et al. 2011). The examinations revealed that twelve of the bears (ten males aged 3–19 years and two females aged 4–7 years) had normal thyroid tissue, while eight bears (40 %) of varying ages and genders (three males aged 3–9 years and five females aged 4–25 years) showed clear histological lesions including parafollicular C cell proliferation, nodular hyperplasia, and interstitial fibrosis. No significant differences were found in the prevalence of thyroid gland lesions between males and females. Similarly, no marked difference was found in mean age between individuals with and without lesions. There was no significant difference in OHC mean concentrations between males and females or between individuals with and without lesions. Despite no documented relationship to OHC concentrations in adipose tissue, it is worth noting that the lesions were similar to those of OHC-exposed laboratory and wildlife contaminated mammals. Since the lesions were not associated with age or gender, other environmental factors such as energetic stress and autoimmunity/genetic predisposition also need to be considered. It is therefore possible that OHCs, in combination with other environmental and intrinsic factors described in the literature, may interfere with the hypothalamic–pituitary–thyroid axis (HPT axis) resulting in endocrine perturbations in East Greenland polar bears.

IPY BearHealth also undertook the first study of temporal developments in liver and kidney lesions (Sonne et al. 2012a, b). Lesions were studied in liver and kidney samples from East Greenland polar bears taken over the 12-year period from 1999 to 2010. Seven liver and seven kidney lesions were observed of which six were age-related. Controlling for this, the analyses showed that hepatic steatosis and renal cell infiltrations, glomerular sclerosis, and tubular hyperplasia decreased over the investigated time period. Similarly, hypertrophy of hepatic Ito cells, renal glomerular capillary wall thickening, and interstitial fibrosis increased over the study period. Lesions were both negatively and positively correlated with OCPs and Hg, respectively. These results suggested that specific liver and renal lesions have decreased or increased over time and that long-range transported OHCs and Hg may be among the cofactors responsible for these observations. These relationships are important to take into account when biomonitoring health and pollution in wildlife species such as polar bears. In conclusion, polar bears are susceptible to long-range transported chemicals that may have various adverse effects on multiple organ systems such as the reproductive and immune system.

Mercury

As part of the IPY and the AMAP Hg assessment, Dietz et al. (2011b, 2012c) conducted a critical review based on the available Hg data in Arctic marine biota, including the polar bear and the Inuit populations, relative to toxicity threshold values. In particular, marine top predators exhibited concentrations of Hg in their tissues and organs that are believed to exceed thresholds for biologic effects. In the Arctic, investigations of histopathological lesions in liver tissue from Arctic wildlife have focused on polar bears (Fig. 11.4), but information is also available from pilot whales, bowhead whales, beluga, and ringed seals (Woshner 2000; Woshner et al. 2002; Sonne et al. 2007, 2010). Liver lesions were found in these species, and where statistically significant associations were found between histochemical endpoints and Hg concentrations, and the histopathological changes found were similar to those observed in other Hg-exposed marine and laboratory mammals. Kidney lesions have been reported in polar bears and some whale species from the Arctic (cf. Woshner 2000; Woshner et al. 2002; Sonne et al. 2007, 2010; Rosa et al. 2008), which resemble those reported for gray seals (*Halichoerus grypus*), ringed seals, and bottlenose dolphins living in the heavily metal and POP-polluted regions such as the Baltic Sea (Lavery et al. 2009; Bergman et al. 2001) (Fig. 11.5). In the lower brain stem, Hg-associated decreases in NMDA receptor levels and DNA methylation was found (Basu et al. 2009). However, beluga whale (*D. leucas*), pilot whale (*Globicephala melas*), and hooded seal (*Cystophora cristata*) were also

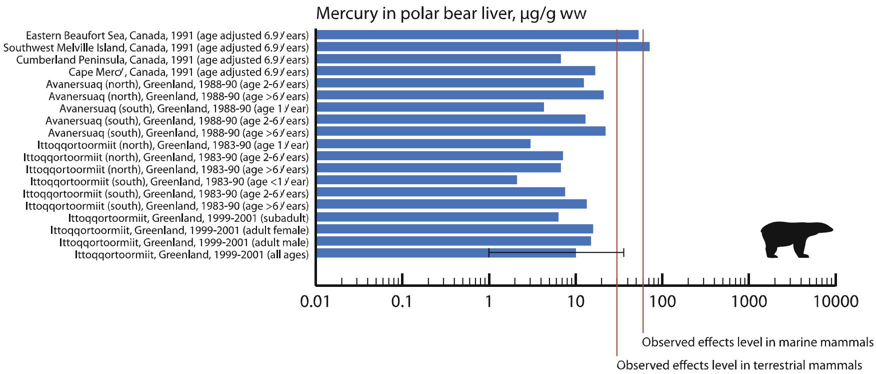

Fig. 11.4 Mercury concentrations in the liver of polar bears from selected regions of the Arctic and collected during selected time periods. The lethal/harmful effect level for terrestrial free-ranging wildlife (30 µg/g ww; Thompson 1996) and the observed effect level for marine mammals associated with liver lesions in bottlenose dolphins (61 µg/g ww; Rawson et al. 1993) are also shown. In cases where Min and Max concentrations are available, these are indicated by range bars (figure from Dietz et al. 2011b, 2012c)

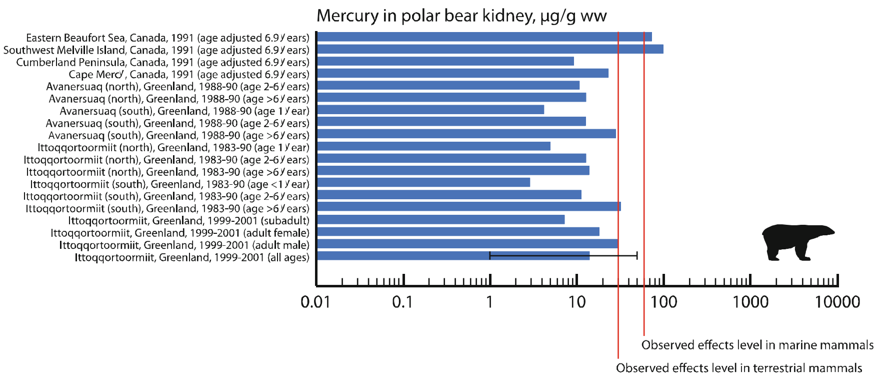

Fig. 11.5 Mercury concentrations in kidney of polar bears from selected regions of the Arctic and collected at selected time periods. The lethal/harmful effect level for terrestrial free-ranging wildlife (30 µg/g ww; Thompson 1996) and the observed threshold level for marine mammals (60 µg/g ww; Rawson et al. 1993) were exceed in a few populations (figure from Dietz et al. 2011b, 2012c)

exceeding threshold values. Toothed whales appear to be one of the most vulnerable groups, with high concentrations of mercury recorded in brain tissue and associated signs of neurochemical effects. Evidence of increasing concentrations in mercury in some biota in Arctic Canada and Greenland is therefore a concern with respect to ecosystem health.

Biomarkers of Endocrine Disruption

Many anthropogenic pollutants have been identified as endocrine disruptive chemicals (EDCs) (Colborn et al. 1993). Indeed, associations between POPs and circulating levels of thyroid hormones and vitamins have been documented in polar bears (Braathen et al. 2004; Haave et al. 2003; Skaare et al. 2001). For the Norwegian component of IPY BearHealth, the aim was to examine effects of OHCs on the homeostasis of several endocrine systems of polar bears in more detail. The study was based on a unique archived material from East Greenland and on samples collected from polar bears in the Norwegian Arctic (Svalbard) during 2007–2008. Multiple statistical modeling was applied to model to identify which specific POP compounds affected the homeostasis of different hormone systems (thyroid and steroid hormones). In polar bears, it was shown that the most important variables with a negative influence on cortisol levels were particularly BDE-99, but also CB-180, CB-201, BDE-153, and CB-170/190 (Bechshøft et al. 2012b). The most important variables with a positive influence on cortisol were CB-66/95, α-HCH, TT3, as well as heptachlor epoxide, dieldrin, BDE-47, and *p,p′*-DDD. Although statistical modeling does not necessarily fully explain biologic cause–effect relationships, relationships indicate that (1) the hypothalamic–pituitary–adrenal (HPA) axis in East Greenland polar bears is likely to be affected by OHC contaminants and (2) the association between OHCs and cortisol may be linked with the hypothalamus–pituitary–thyroid (HPT) axis (Bechshøft et al. 2012b).

As for all other Arctic animals, polar bears are exposed to and accumulate hundreds of (presently known) anthropogenic pollutants. There is virtually no knowledge about which of the compounds that polar bears accumulate are thyroid disruptive. In a study on polar bears from East Greenland, multivariate statistical modeling was applied in an attempt to identify which of the OHCs could be involved in the thyroid hormone disruption (Villanger et al. 2011a, b). The study showed that some particular OHCs were especially important in explaining variations in circulating TH levels. These were BDE-99, BDE-100, BDE-153, CB-52, CB-118, *cis*-nonachlor, *trans*-nonachlor, trichlorobenzene (TCB), and pentachlorobenzene (QCB). Both negative and positive relationships with THs were found. The most important OHCs that influenced TH levels in the significant PLS models may potentially act through similar mechanisms on the hypothalamic–pituitary–thyroid (HPT) axis, suggesting that both combined effects by dose and response addition and perhaps synergistic potentiation may be a possibility in these polar bears. The study also revealed that adult male polar bears were less susceptible to thyroid-disruptive chemicals than females and subadults (Villanger et al. 2011a, b).

Important aims of IPY BearHealth were to examine to which extent the OHCs in polar bear cubs bind to TTR and also to examine whether the reduced levels of PCBs, OH-PCBs, and PFOS in cubs from 1998 to 2008 resulted in less binding of these compounds to TTR. Blood samples were collected from nursed polar bear cubs (approximately 4 months of age), and a sample preparation method combining solid-phase extraction (SPE) and liquid–liquid extraction (LLE) was developed to extract a broad range of thyroid hormone disruptors from plasma (Simon et al. 2011). A first screening revealed that the thyroid hormone-disrupting pollutants had a high TTR-binding potency and that OH-PCBs could explain 60–85 % of the TTR-binding activity (Simon et al. 2011). In a follow-up study applying the developed method (Bytingsvik et al. 2013), we showed the measured TTR-binding activity correlated positively with plasma levels of hydroxylated PCBs (OH-PCBs). No such association was found between TTR-binding activity and the plasma levels of perfluoroalkyl substances (PFASs). The OH-PCBs in the cubs explained 60 $\pm$ 7 and 54 $\pm$ 4 % of the TTR-binding activity in 1998 and 2008, respectively, and PFASs explained ≤ 1.2 % both years. Thus, almost half the TTR-binding activity could not be explained by the contaminants we examined, and further efforts are needed to identify these thyroid-disruptive contaminants. The study also showed that the TTR-binding activity related to contaminant levels was significantly lower (45 %) in 2008 than in 1998 (Bytingsvik et al. 2013). Thus, the 50 % decrease in levels of Σ_6OH-PCBs in Svalbard polar bear cubs from 1998 (98 $\pm$ 23 ng/g ww) to 2008 (49 $\pm$ 21 ng/g ww) (Bytingsvik et al. 2012a) have indeed resulted in a reduced binding of the contaminants to TTR.

Conclusions

East Greenland polar bears are among the most contaminated species on our globe in spite of their remote Arctic habitat. This subpopulation inhabits Arctic biotopes being constantly under pressure from global warming and associated environmental changes. Anthropogenic environmental contaminants seem to be a cofactor in various organ system lesions in East Greenland polar bears. This includes effects on the immune system and sexual and reproductive organ size, reduced bone density, as well as series, thereby having potential impacts on individual health and population maintenance. On top of this, also global warming seems to affect polar bears via negative energy balances, which may have consequences for fecundity and immune resistance. Clearly, the marine food web changes in relation to sea ice changes that have been documented emphasize that Arctic wildlife including polar bears will continue to be subject to region-specific changes over time in the levels and patterns of POPs that they accumulate via their diets.

Future Challenges

A major overall challenge in the future will be to integrate the cumulative impact of multiple stressors on biota and wildlife, including Arctic warming and exposures to a cocktail of various contaminants with similar or dissimilar modes of action, across temporal and spatial gradients by integrating empirical data and laboratory studies.

One specific challenge is to link effects on the hormone and reproductive and immune systems to effects on the population level. As we have discussed, contaminant exposure is suggested to have potential health effects on various organ systems in East Greenland polar bears which may also be the case for other populations. However, contaminants are not the only environmental stressor in the Arctic. Also, global warming leading to decreased food access and negative energy balance may influence bear health via (sub)clinical impacts on immune functioning and reduced fecundity having an impact on populating size. In addition to this, global warming may also increase the infectious stress due to invasive micro-pathogen and parasitic diseases. The main challenge in the future is therefore to integrate the cumulative impact from these multiple stressors across temporal and spatial gradients by integrating empirical data and laboratory studies. The polar bear seems to be an excellent biomonitoring organism for such cumulative impact studies.

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Chapter 12
A Circumarctic Review of Contaminants in Ringed Seals

Katrin Vorkamp and Derek C.G. Muir

Introduction

This chapter reviews the information on contaminants in Arctic ringed seals available till 2013. We have limited the contaminants to persistent organic pollutants (POPs), mercury (Hg), and cadmium (Cd) because the majority of studies have focused on these and they represent the contaminants of greatest concern both for the seals and for the exposure of humans consuming harvested seals. To provide proper context for this review, we have included information on transport pathways and food web bioaccumulation of contaminants. However, readers are encouraged to seek out other sources for detailed descriptions of pathways and marine food webs that can be found, for example, in AMAP assessment reports on POPs and Hg (AMAP 2004, 2005, 2011; Murray 1998). Ringed seals have also been important bioindicators of contaminants trends and effects in the Baltic Sea (Nyman et al. 2003). However, we have limited our coverage to the Arctic Ocean populations.

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Ringed Seals in the Arctic Food Web

Ringed seal is the most abundant Arctic pinniped with a circumpolar distribution. It has been a key biomonitoring animal for examining spatial and temporal trends of POPs and Hg in the Arctic since the 1970s (Murray 1998). Because of their high abundance, ubiquitous distribution, and central position in the food web, ringed seals play an important role in the dynamics of Arctic marine ecosystems (Smith et al. 1991). The ringed seal diet consists of fish, mainly Arctic cod (*Boreogadus saida*), polar cod (*Arctocadus glacialis*), and crustaceans (amphipods, mysids, and euphausiids) (Reeves 1998). Fatty acid studies suggest that ringed seals forage within limited areas (Thiemann et al. 2007) and confirm other observations that they are relatively sedentary with males possibly occupying the same under ice habitat for up to 9 months (Reeves 1998; Smith 1987). Some regional movements, e.g., from the western Canadian Arctic islands to the East Cape of Siberia (Smith 1987) and between Greenland and Canadian waters, have been documented (Teilmann et al. 1999).

Contaminant Exposure of Ringed Seals

Most of the organic contaminants discussed in this chapter are emitted to the environment at temperate latitudes during their production, use, or disposal. Semi-volatile compounds can be rapidly transported to the Arctic with the atmosphere, i.e., in days or weeks. Depending on their volatility and the prevailing temperature, the compounds move at different velocities, including phases of deposition and revolatilization ("grasshopper effect") (Wania and Mackay 1996). Typical meteorological conditions over the oceans lead to a more effective transport in winter than in summer, with the side effect of less photolytic degradation and efficient contaminant scavenging by snow (AMAP 2004). The transport to the Arctic by ocean currents usually takes years, but is important for compounds with low Henry's Law Coefficients, such as β-hexachlorocyclohexane (β-HCH) or perfluorooctane sulfonate (PFOS). Additional input of contaminants into the Arctic environment can come from large rivers (Carroll et al. 2008).

Concentrations of POPs such as polychlorinated biphenyls (PCBs) are generally very low in Arctic air, snow, seawater, and sediments but sufficiently high to be measurable (Gustafsson et al. 2005; Hung et al. 2010; Savinov et al. 2003). As most of these compounds are very lipophilic, they favor accumulation in lipid-rich rather than aquatic media, including lipids of marine invertebrates ("bioconcentration"). This transfer from the abiotic to the biotic environment also marks the entry into food chains: Fish foraging on invertebrates take up POPs as part of their diet, and this POP transfer is repeated with each trophic level of the food chain. Thus, POP concentrations increase from prey to predator ("biomagnification") and through the food chain ("trophic magnification").

Poorly metabolized organic compounds with relatively low octanol-water partition coefficients (K_{OW}), i.e., logK_{OW} values of (2–5), but high octanol–air partition coefficients (K_{OA}), i.e., logK_{OA} values >6, such as HCH isomers and tetrachlorobenzene, can also biomagnify in air-breathing organisms such as ringed seals because of the low rate of respiratory elimination to air (Kelly et al. 2007). In addition to changes in POP patterns during transport and partitioning processes, the biological processes of absorption, metabolism, and excretion further alter their composition (Norstrom and Muir 1994).

Although Hg and Cd are naturally occurring elements, global human activity has led to increased concentrations in the Arctic based on evidence from archives such as dated lake sediment and ice cap cores (Li et al. 2003; McConnell and Edwards 2008; Munthe et al. 2011). A meta-analysis of studies on Hg in hard tissues of marine species (teeth and feathers) found that there had been a 92 % increase of Hg in Arctic marine animals that began in the late nineteenth century and accelerated in the twentieth century (Dietz et al. 2009).

The Hg cycle within the Arctic Ocean is unique globally due to a number of features such as its semi-enclosed waters and the ice cover of the Arctic Ocean, a large freshwater input compared to other oceans, the seasonality of light, and the proximity to pollution sources in midlatitude regions (Douglas et al. 2011; Macdonald and Loseto 2010). An array of processes promotes the conversion of atmospheric and ocean-transported Hg to methylmercury (MeHg), its bioaccumulative and toxic form.

The fate of Hg in Arctic ecosystems was comprehensively reviewed recently (Douglas et al. 2011; Munthe et al. 2011; Braune et al. 2015a). The AMAP assessment also included a meta-analysis of trends of Hg in ringed seals and other species (Braune et al. 2011).

Cadmium has also entered Arctic marine environments as a result of anthropogenic inputs from both atmospheric deposition and local sources such as mining (Dietz et al. 1998a). Atmospheric inputs were much greater in the 1970s than in the 1990s (Boutron et al. 1991; Li et al. 2003). In the ocean, Cd is incorporated in plankton along with other divalent cations such as phosphate (de Baar et al. 1994) and remineralizes as organic matter degrades and forms sinking particles (Macdonald and Sprague 1988).

Macdonald et al. (2000) concluded that the atmospheric contaminant Cd flux is less than 1 % of either the Cd fluxes into or out of the Arctic Ocean and only approximately 5 % of the estimated flux of Cd to sediments. This makes anthropogenic fluxes difficult to detect.

Contaminant Toxicokinetics and Biodegradation in Ringed Seals

Ringed seals take up POPs from their diet, via gastrointestinal absorption (AMAP 2004). The absorption process involves bile salt micelles which transport the contaminants away from the digested lipids and through an unstirred water layer into the epithelial cells (Kelly et al. 2004). Following the diffusion into blood, as either lipid-associated or individual molecules (Kelly et al. 2004), lipophilic contaminants are distributed to lipid-rich tissues such as liver and adipose tissue (AMAP 2004). More hydrophilic contaminants, such as PFOS, bind to proteins and accumulate in protein-rich tissue such as liver and blood (Jones et al. 2003). Tributyltin (TBT) has been found to be distributed to seal fur as well (AMAP 2004).

For lipid-associated POPs, variation in lipid content affects POP concentrations: As juvenile ringed seals grow, their lipid stores grow rapidly (Lydersen et al. 1992), leading to growth dilution of contaminants and decreasing concentrations. The opposite process occurs during periods of fasting: As lipid stores are utilized, POP concentrations increase and new equilibria are established between tissues (AMAP 2004). Increasing POP levels during fasting periods have been shown for harp seals (*Phoca groenlandica*) (Lydersen et al. 2002).

Organ- and tissue-specific concentrations are available for several chlorinated contaminants in ringed seals from West Greenland and East Greenland (Johansen et al. 2004; Vorkamp et al. 2004) (Fig. 12.1). For POPs, such as PCBs and DDT, lipid-normalized concentrations were highest in blubber. Other, less persistent organochlorines primarily occurred in the liver, based on lipid-normalized concentrations. If concentrations were not lipid-normalized, blubber concentrations would always be highest (Fig. 12.1).

The presence of PCB and DDT metabolites in ringed seal tissue (and their absence in ringed seal prey) shows that some metabolism of these persistent compounds occurs (Letcher et al. 1998). Phase I metabolism involving xenobiotic metabolizing liver enzyme systems (Cytochrome P450, CYP) reduces the compound's lipophilicity, often by inserting a hydroxy (OH) group. Phase II metabolism forms complex conjugates which can be excreted more easily. Ringed seal liver was shown to contain CYP1A and CYP3A enzymes as well as the phase II enzymes UDP glucuronosyl transferase (UDPGT) and glutathione S-transferase (GST) (Wolkers et al. 1998). These enzymes can catalyze the reactions to OH–PCBs, methylsulfonyl ($MeSO_2$)–PCBs, and $MeSO_2$-*p,p′*-DDE, which have been detected in ringed seals (Letcher et al. 1998).

Mixed results were reported with regard to the activity of CYP2B in ringed seal liver (Boon et al. 1997; Wolkers et al. 1998). According to Boon et al. (1997), pinnipeds also show an inducible CYP2B activity, which catalyzes the insertion of an OH-group at *meta*-*para*-unsubstituted positions in PCB molecules, as present in, e.g., CB-52, CB-101, and CB-149. These PCB molecules in particular are further metabolized via phase II reactions to $MeSO_2$ metabolites (Boon et al. 1997). In the main OH-PCBs in ringed seals, the OH-group is in *para*-position, either from arene oxide formation or from direct insertion of an OH-group in *para*-position (Routti et al. 2008a).

CYP1A catalyzes the reaction in planar PCB congeners with vicinal H atoms in *ortho*- and *meta*-positions, while OH-groups cannot be inserted into molecules without vicinal H atoms (e.g., CB-153 and CB-180) (Boon et al. 1997). A CYP1B-like protein has also been found in ringed seal liver, possibly catalyzing the metabolism of polycyclic aromatic hydrocarbons (PAHs) (Nyman et al. 2000).

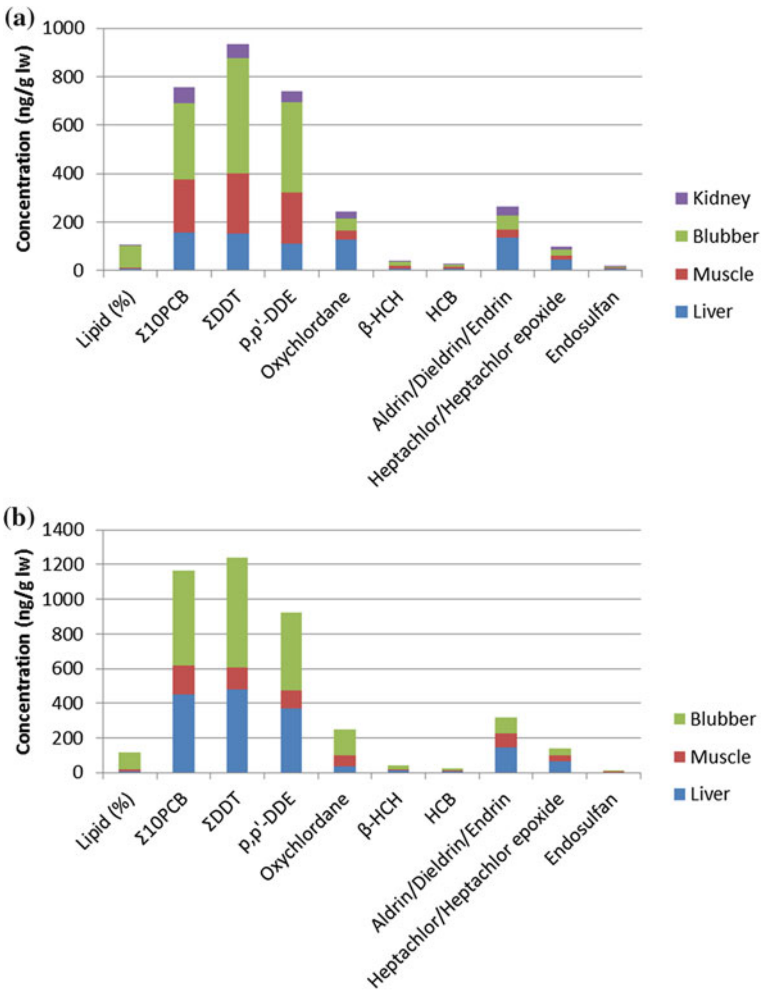

Fig. 12.1 Concentrations (ng/g lipid weight) of organochlorine compounds in liver, muscle, blubber, and kidney of ringed seals from West Greenland (**a**) and East Greenland (**b**). For ringed seals from East Greenland, no kidney data were available. Data from Johansen et al. (2004) and Vorkamp et al. (2004). Σ_{10}PCB includes PCBs 28, 31, 52, 101, 105, 118, 138, 153, 156, and 180

Besides PCBs and *p,p*′-DDE, only few compounds have been studied with regard to their metabolism in ringed seals. Chlordanes were found to be metabolized to oxychlordane and *trans*-nonachlor, possibly involving CYP3A, which also seems to be involved in the formation of the toxaphene congener CHB-44 (Routti et al. 2009b). Similarly, pentachlorophenol and 4-hydroxyheptachlorostyrene have been suggested to be a result of phase I biotransformation of hexachlorobenzene and octachlorostyrene, respectively (Routti et al. 2009b). Unlike OH–PCBs, hydroxylated metabolites of polybrominated diphenyl ethers (PBDEs) have only been detected at low levels in ringed seal blubber, if at all (Kelly et al. 2008; Letcher et al. 2009; Routti et al. 2009a). Methoxy (MeO)–PBDEs in ringed seals are most likely the result of bioaccumulation from natural sources in the marine environment, rather than of biotransformation (Kelly et al. 2008).

Comparisons between ringed seals from the Arctic and more polluted areas such as the Baltic Sea have revealed some differences in enzyme activities, based on enzyme induction at elevated exposure levels (Nyman et al. 2000). Thus, highly contaminated ringed seals biotransformed PCB and DDE more efficiently (Routti et al. 2008a, 2009a). However, the possibility of enzyme saturation was discussed as well, as higher concentrations of the metabolite oxychloradane were found for ringed seals from remote areas compared with the Baltic Sea (Routti et al. 2009a).

The formation of metabolites is not necessarily a detoxification process (AMAP 2004). The *para*-substituted OH–PCBs, which are predominant in ringed seals, are similar to thyroid hormones and thus capable of endocrine disrupting effects (Routti et al. 2008a). Disturbed thyroid hormone and vitamin A homeostasis in ringed seals have been related to PCB exposure (Nyman et al. 2003).

The main excretion route of POPs and their metabolites in ringed seals is via feces and, to some extent, urine (AMAP 2004). Female ringed seals can excrete a substantial part of their POP body burden via lactation. Hickie et al. (2005) calculated that during 40 days of lactation, female ringed seals could transfer 5–40 % of their body burden of various contaminants. The authors concluded that rapid juvenile growth, a high rate of population turnover, and their biotransformation capacity make ringed seals respond quickly to changes in contaminant loadings (Hickie et al. 2005).

Mercury and MeHg biomagnify in the Arctic marine food web (Campbell et al. 2005). In the Northwater food web study (northern Baffin Bay), the percent MeHg of total Hg increased from 7.5 % in copepods to 100 % in Arctic cod. More than 95 % of the MeHg in food items is taken up by mammals, whereas the corresponding proportion for inorganic Hg is only 15 % (Berlin 1986). The percentage MeHg in liver of ringed seals from Alaska and Ulukhaktok in the Western Canadian Arctic declined rapidly with age, reaching a relatively constant value of 5–20 % in adults even as levels of total Hg continued to increase (Brookens et al. 2007; Dehn et al. 2005).

Like other high-trophic-level marine mammals and birds, ringed seals may be partially protected against MeHg toxicity by a range of mechanisms including demethylation and binding of inorganic Hg with Se in an approximate 1:1 molar ratio (Koeman et al. 1973). In the case of ringed seals, this is illustrated for three widely separate sampling areas in the Canadian Arctic in Fig. 12.2. Se:Hg ratios in liver ranged from 1:10 to 1:1. Similar ratios were found in seals from Greenland (Dietz et al. 2000). Se:Hg ratios were higher in ringed seals from Barrow which had low total Hg concentrations (2.47 μg/g wet weight) compared with seals from Ulukhaktok which had tenfold higher levels of total Hg. This difference is thought to be due to higher Hg concentrations giving rise to a larger percentage of total Hg as inorganic Hg associated with Se in liver. The Se–Hg complex has been identified as tiemanite (SeHg) (O'Hara et al. 2003).

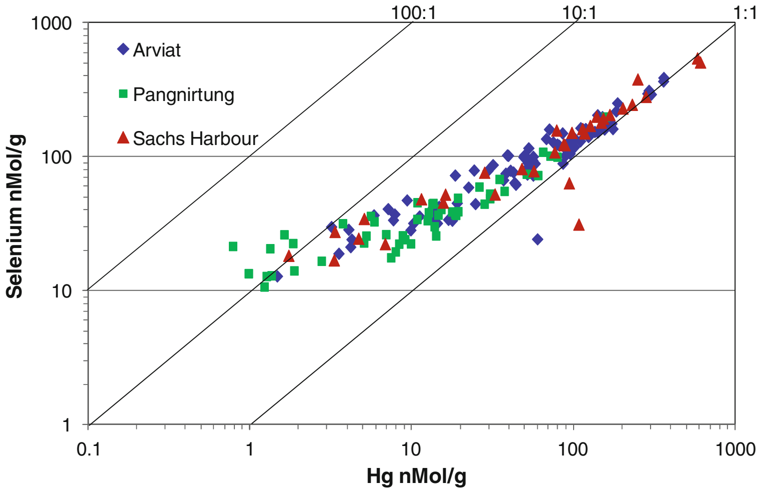

Fig. 12.2 Selenium–mercury relationships in livers for three ringed seal populations from the Canadian Arctic

Ringed seals also accumulate high levels of Cd in renal and hepatic tissues (Dehn et al. 2005; Dietz et al. 1998b). However, Cd concentrations declined with trophic level in the Northwater marine food web due to high concentrations in invertebrates especially in the amphipod *Themisto libellula* (Campbell et al. 2005). Dehn et al. (2005) also found that trophic level was negatively correlated with Cd in Alaskan ringed seals, suggesting dependence of Cd with low trophic-level prey. The high Cd concentrations in ringed seal tissues suggest the importance of dietary zooplankton prey compared with fish such as Arctic cod and Pacific herring which have very low Cd concentrations (Campbell et al. 2005; Dehn et al. 2005).

Geographical Trends of Contaminants in Ringed Seals of the Arctic

Spatial trends of contaminants in the Arctic are of interest for assessing sources and pathways. Ringed seals are particularly useful for such studies because of their circumpolar distribution and the availability of samples from traditional hunting. Previous spatial trend studies have been conducted using data for PCBs and organochlorine pesticides (Muir et al. 2000) and on Hg and Cd (Rigét et al. 2005). AMAP assessment reports on POPs in 1998, 2004, and 2009 also included spatial trend maps for selected POPs in ringed seals (AMAP 2004, 2009a; de March et al. 1998; de Wit et al. 2010). Earlier AMAP heavy metal assessments also included spatial trends of Hg and Cd in ringed seals (AMAP 2005; Dietz et al. 1998a).

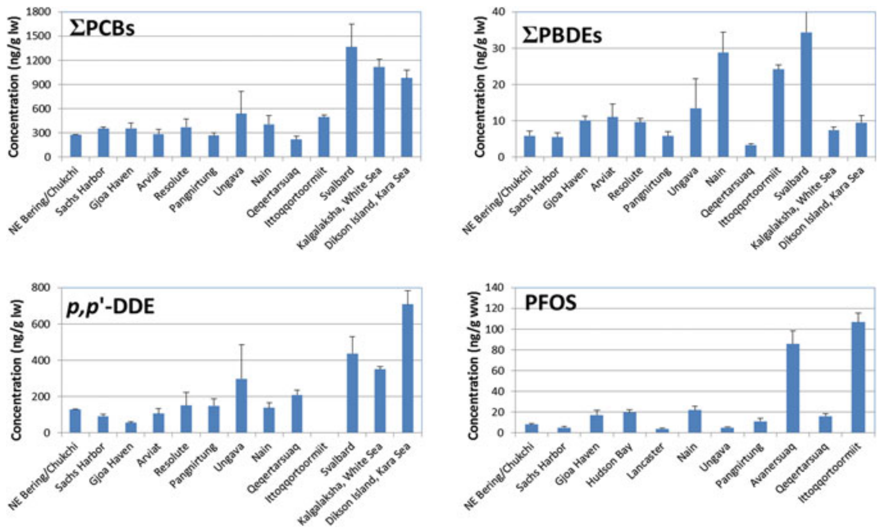

Fig. 12.3 Geographical trends of major POPs in ringed seals. Sampling locations are arranged from west (Bering/Chukchi Sea) to east (Svalbard/Kara Sea). Data from Muir et al. (2013), Quakenbush (2007), Quakenbush and Citta (2008), Quakenbush et al. (2011), Rigét et al. (2013a, b), Routti et al. (2008a, 2009a, b), Rotander et al. (2012a), Savinov et al. (2011) and Vorkamp et al. (2008, 2011) partly recalculated. ΣPBDEs from Qeqertarsuaq only show BDE-47. See text for details

Geographical trends for PCBs, *p,p′*-DDE, total (Σ)PBDEs, and PFOS in ringed seals from Alaska, Canada, Greenland, and Russia are presented in Fig. 12.3. They have been selected because they represent the major groups or individual POPs of concern in terms of exposure of ringed seals. To assemble these data, we used geometric mean or arithmetic mean concentrations and standard errors provided in the most recent published reports for each location. For PCBs, *p,p′*-DDE and PBDEs results from Canada, Greenland, and Russia were for blubber from female and/or juvenile males. This limits the effect of age as most POPs increase with age in males, but not in females because of the contaminant transfer to the offspring (Addison and Smith 1974). For results from the northeast Bering/southern Chukchi Seas (Alaska), we used combined males and females because results were presented in this format (Quakenbush 2007; Quakenbush and Citta 2008; Quakenbush et al. 2011). For ringed seals from Svalbard, recent results for PCBs, *p,p′*-DDE, and PBDEs were only available for liver (Routti et al. 2008a, 2009a, b) and, for comparison with other locations, were converted to lipid weight by dividing by fraction lipid.

The highest concentrations of PCBs and *p,p′*-DDE were observed in female ringed seals from Svalbard and the Russian Arctic (Fig. 12.3). This observation is consistent with an earlier report for ringed seals (Muir et al. 2000) that was based on statistical analysis of a combined dataset from the mid-1990s. This geographical pattern has been attributed to emissions from existing PCB containing equipment and other products and to continued use of older pesticide stocks in the Russian Arctic in the 1990s and early 2000s (AMAP 2003, 2009b; Muir et al. 2000). The dates of collection of the samples for PCB and *p,p′*-DDE varied from 2001 (Kalgalaksah and Dikson Island) to 2010 (Canadian Arctic, Greenland). Therefore, the results from the Russian Arctic, in particular, may not reflect the status of these contaminants as of 2010. On the other hand, rates of decline of PCBs and *p,p′*-DDE have been slow in Arctic biota in general (1.9 % per year for Σ_{10}PCB and for *p,p′*-DDE; Rigét et al. 2010) and therefore, the picture may not have changed significantly over the period 2001–2010.

Geographic trends for ΣPBDEs differed from PCBs with higher concentrations in two eastern Canadian Arctic locations (Nain and Ungava Bay), in East Greenland as well as in Svalbard (the latter based on liver; Routti et al. 2009a). Lowest concentrations were found in the Bering–Chukchi Sea seals (Quakenbush 2007) and in seals from the western and central Canadian Arctic archipelago (Sachs Harbor, Gjoa Haven, and Resolute). BDE-47, a predominant congener in ringed seals in the Canadian and Greenland samples, was below detection limits in the Bering–Chukchi ringed seals. Higher use of PBDEs occurred in North America relative to Europe, and use of the commercial PentaBDE product was more limited in Japan and China (de Wit et al. 2010). Therefore, the overall geographical pattern may be due to atmospheric transport and deposition from North America influencing the northwest Atlantic Ocean.

PFOS measurements appear to be available only for the Bering–Chukchi Sea, Canadian Arctic, and Greenland and therefore, the geographic comparison is more limited. Higher concentrations of PFOS are observed in ringed seals from Greenland relative to locations in Canada and Alaska. Possibly, there are multiple sources given the transport of PFOS ions in ocean waters as well as via neutral precursors in the atmosphere (Butt et al. 2010). Rotander et al. (2012a) noted that the pattern of perfluorinated carboxylates (PFCAs) in samples from North West Greenland (Avanersuaq) differed from the Canadian Arctic and East Greenland.

The geographic coverage for Hg and Cd in liver of ringed seals is more extensive than for organic contaminants with data available for two regions in Alaska, the Canadian Arctic, Greenland, Svalbard, and the Chukotka–Lavrentiya in eastern Russia (Fig. 12.4). Highest liver Hg concentrations are found in the western Canadian Arctic (Sachs Harbor and Ulukhaktok). The elevated concentrations are

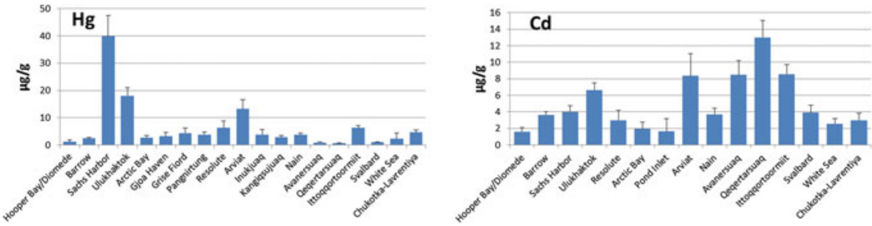

Fig. 12.4 Geographical trends of mercury (Hg) and cadmium (Cd) in liver of ringed seals. Concentrations are geometric means or arithmetic means ± standard error from reports, most of them published in the 2000s. Data from AMAP (2005), Chételat and Braune (2012), Dehn et al. (2005), Quakenbush et al. (2011), Muir et al. (unpublished data) and Rigét et al. (2005; 2012)

not observed further west at Barrow (Dehn et al. 2005), nor in Gjoa Haven to the east and are therefore rather unique to this region. The elevated Hg was first noted by Wagemann et al. (1996). Rigét et al. (2005) also studied this geographical trend using samples from the late 1990s. Stomach content analysis of ringed seals from Alaska and from Ulukhaktok showed that all seals had cod (Gadidae) as a main component of the diet implying that diet cannot explain the differences (Dehn et al. 2007). However, carbon isotope signatures in ringed seals from Ulukhaktok were significantly depleted (more negative) compared to ringed seals from Alaska suggesting different sources of carbon for the southeastern Beaufort Sea compared to the Barrow and Bering–Chukchi area.

Cadmium concentrations in ringed seals are quite variable with higher levels in the western Canadian Arctic as well as in Hudson Bay (Arviat) and in West and East Greenland populations (Fig. 12.4). Rigét et al. (2005) previously noted that Cd concentrations in ringed seals were highest in the eastern Canadian Arctic and West Greenland based on data from the late 1990s. Relatively high Cd concentrations in ringed seal tissues have been attributed to the ingestion of zooplankton (Macdonald and Sprague 1988; Dehn et al. 2005), which is a significant diet component although not as important as fish according to Dehn et al. (2007). Rigét et al. (2005) concluded that local/regional geology influencing the prevailing levels of Cd in the marine environment was the most likely explanation for differences.

Temporal Trends of Contaminants in Ringed Seals of the Arctic

Ringed seal is a key species for monitoring the variation of contaminant concentrations in the Arctic, primarily in Canada and Greenland. Regular sampling was introduced in the mid-1990s to early 2000s, and some time series have been extended backward by including archived samples from environmental specimen banks. Table 12.1 summarizes publications of the last 10 years which provide details on contaminant time series in ringed seals from Arctic Canada and Greenland. All biota-based time series from the Arctic, including those on ringed seals, were reviewed and statistically analyzed by Rigét et al. (2010). While "legacy" organochlorine compounds generally decreased in ringed seals and other species, there are few interesting observations of differences between compounds and locations.

Figure 12.5 shows PCB concentrations in ringed seals from Ulukhaktok (Beaufort Sea) and Arviat (Hudson Bay) as well as in central East and central West Greenland (Addison et al. 2014; Muir et al. 2013; Vorkamp et al. 2008, 2011). Vorkamp et al. (2011) have noted that the trend analyses can lead to different results depending on the offset of the time series, as the most pronounced decrease in PCB concentrations could be expected prior to the beginning of regular monitoring. Indeed, the time series from Ulukhaktok dating back to 1972 showed a steep

Table 12.1 Summary of original publications since 2005 providing details on contaminant time series in ringed seals from the Arctic

Country	Location	Compounds	Time period	Reference
Greenland	Qeqertarsuaq, Ittoqqortoormiit	Toxaphene	1986–2012	Vorkamp et al. (2015)
Canada	Beaufort Sea[a], Lancaster Sound[b], East Baffin, Hudson Bay[c]	Hg	1999–2009	Braune et al. (2015a)
Canada	Ulukhaktok	PCBs, DDTs, HCB, chlordane-related pesticides	1972–2010	Addison et al. (2014)
Canada	Beaufort Sea[d], Lancaster Sound[e], East Baffin, Hudson Bay[f]	PCBs, DDTs, HCB, chlordane-related pesticides, HCHs, toxaphene, PBDEs, HBCD, PFASs	1972–2010[g]	Muir et al. (2013)
Greenland	Qeqertarsuaq, Ittoqqortoormiit	PFASs	1982–2010	Rigét et al. (2013a)
Greenland	Qeqertarsuaq	CB-52, CB-153, *p,p′*-DDE, HCB, α-HCH, β-HCH	1994–2010	Rigét et al. (2013b)
Canada	Ulukhaktok	PCBs, DDTs, HCB, chlordane-related pesticides, HCHs, toxaphene	1993–2008	Gaden et al. (2012)
Canada	Beaufort Sea[a], Lancaster Sound[b], East Baffin, Hudson Bay[c]	Hg	1999–2009[g]	Chételat and Braune (2012)
Greenland	Avanersuq, Qeqertarsuaq, Ittoqqortoormiit	Hg	1984–2010[g]	Rigét et al. (2012)
Greenland	Ittoqqortoormiit	HBCD	1986–2010	Vorkamp et al. (2012)
Greenland	Ittoqqortoormiit	PCBs, PBDEs, HBCD	1986–2008	Vorkamp et al. (2011)
Canada	Ulukhaktok	HCHs	1978–2006	Addison et al. (2009)
Canada	Ulukhaktok	Hg	1973–2007	Gaden et al. (2009)
Canada	Ulukhaktok	Hg	Middle ages to today	Outridge et al. (2009)
Greenland	Qeqertarsuaq, Ittoqqortoormiit	HCHs	1986–2006	Rigét et al. (2008)
Greenland	Qeqertarsuaq	PCBs, DDTs, HCB, chlordane-related pesticides, HCHs, toxaphene, BDE-47	1982–2006[g]	Vorkamp et al. (2008)
Canada	Lancaster Sound	PCBs, DDTs, HCHs	1975–2006	Lohmann et al. (2007)
Canada		PFASs	1992–2005	

(continued)

Table 12.1 (continued)

Country	Location	Compounds	Time period	Reference
	Lancaster Sound, Hudson Bay			Butt et al. (2007)
Greenland	Ittoqqortoormiit	PCBs, PBDEs	1986–2004	Rigét et al. (2006)
Canada	Ulukhaktok, Arctic Bay, Grise Fjord	PCBs, DDTs, HCHs, chlordane-related pesticides	1972–2001[g]	Braune et al. (2005)
Greenland	Ittoqqortoormiit	PFASs	1986–2003	Bossi et al. (2005)

HCB: Hexachlorobenzene, HBCD: Hexabromocyclododecane, PFASs: Perfluorinated alkylated substances
[a]Sachs Harbor
[b]Resolute
[c]Arviat
[d]Summarizes data from Sachs Harbor and Ulukhaktok
[e]Summarizes data from Resolute, Grise Fjord, and Arctic Bay
[f]Summarizes data from Arviat and Inukjuaq
[g]Maximum time span. Smaller time spans for some compounds or locations

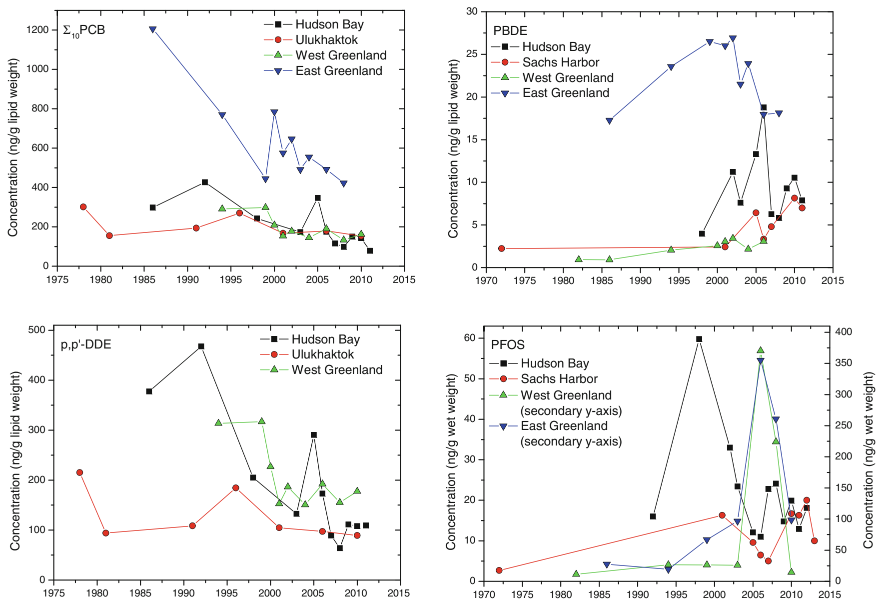

Fig. 12.5 Concentrations of Σ_{10}PCB, *p,p′*-DDE, PBDEs (geometric means in ng/g lipid weight), and PFOS (geometric means in ng/g wet weight) in female and juvenile male ringed seals from Ulukhaktok or Sachs Harbor (Beaufort Sea), Arviat (Hudson Bay), West and East Greenland. For PFOS, the results from West and East Greenland refer to the secondary y-axis (ng/g wet weight) on the right. Σ_{10}PCB includes PCBs 28, 31, 52, 101, 105, 118, 138, 153, 156, and 180. Data from Addison et al. (2014), Muir et al. (2013), Rigét et al. (2013a, b) and Vorkamp et al. (2011), partly recalculated

decline of PCBs between 1972 and 1981 and relatively stable concentration since the 1990s (Addison et al. 1986, 2014; Addison and Smith 1998; Gaden et al. 2012). This stabilization has also been apparent in the Greenland time series since about the year 2000 (Fig. 12.5). It suggests recent emissions of PCBs, which could originate from former open applications of PCBs, e.g., in sealants (Kohler et al. 2005), from the disposal of PCB containing material, e.g., e-waste (Chen et al. 2014), or from secondary sources, e.g., releases from a warming abiotic environment (Schmid et al. 2011). Changes in feeding habits and food web structures may play a role as well (McKinney et al. 2011; Braune et al. 2015b).

The concentration of ΣDDT has decreased at all Canadian locations and in West Greenland, as exemplified for *p,p′*-DDE in Fig. 12.5 (Muir et al. 2013; Vorkamp et al. 2008). No trends have recently been published from East Greenland, but decreasing trends have been reported in national reports. Recently, increasing atmospheric concentrations have been reported of *o,p′*-DDT, possibly as a consequence of its occurrence in the insecticide dicofol (Venier and Hites 2014), and it will be interesting to follow whether this will be reflected in ringed seals of the Arctic.

In contrast to the generally decreasing trends of organochlorine compounds in ringed seals of the Arctic, β-HCH increased at some, but not all Canadian locations (Muir et al. 2013). At Ulukhaktok, the annual increase was as high as 12 and 16 % in males and females, respectively, between 1978 and 2006 (Addison et al. 2009; Muir et al. 2013). The time series from Greenland did not show an increase of β-HCH, but the decreasing trend was considerably lower (−1.4 to −3.9 % per year) than that of α- or γ-HCH (Rigét et al. 2008). Interestingly, toxaphene was also found to increase at one Canadian location in the last four years and to decline nonsignificantly at others (Muir et al. 2013).

Early reports on PBDE levels in ringed seals from Ulukhaktok (Canada) showed an exponential increase of the tri- to hexabrominated PBDEs, which paralleled the production of the PentaBDE mixture (Ikonomou et al. 2002). Doubling times of 4–9 years were predicted based on the data available at the time. The recent update still showed significantly increasing concentrations for ringed seals from the Beaufort Sea (Muir et al. 2013) (Fig. 12.5). Detailed concentration curves from other Canadian locations, however, indicate peaks in the mid-2000s, but current or recent concentrations still exceeded those of the early 1970s (Muir et al. 2013). Likewise, the results from Greenland do not show a uniform trend (Fig. 12.5). The time trend from East Greenland suggests peaking concentrations around the year 2000 (Vorkamp et al. 2011). The trend of BDE-47 in seals from West Greenland is still positive, but the maximum concentration was measured in 2002 (Vorkamp et al. 2008). Overall, the time series indicate peaking concentrations between approximately 2000 and 2005. This would precede the global ban of Penta- and OctaBDE which were included in the Stockholm Convention in 2009. More data points will be needed to prove the changing trend statistically.

Changes from increasing to decreasing trends have also been reported for PFOS (Butt et al. 2007; Rigét et al. 2013a). The recent update from Canada confirmed peaking concentrations between 1998 and 2000 (Fig. 12.5), suggesting a rapid response to voluntary and regulatory measures to reduce PFOS emissions. Like Penta- and OctaBDE, PFOS was added to the Stockholm Convention in 2009, but this had been preceded by national programs and the main manufacturer's ceased production of perfluorooctanesulfonyl fluoride-based compounds in 2001 (Butt et al. 2007). In both East and West Greenland ringed seals, the PFOS concentrations peaked in the year 2006 (Rigét et al. 2013a). Disappearance half-lives increased in the order West Greenland (0.9 years) < East Greenland (2.4) < Hudson Bay (3.2) < Lancaster Sound (9.2) (Muir et al. 2013; Rigét et al. 2013a).

The trends of PFCAs are similar to those of PFOS in the Canadian ringed seals. The only statistically significant trend, however, was observed in Lancaster Sound, with an increase until 2003 and an annual decrease of 18 % between 2003 and 2011 (Muir et al. 2013). The data from Greenland appear more complex as PFOA peaked in East and West Greenland in 2006/2008, and so did the longer chained PFCAs (C_9–C_{11}) in West Greenland. Increasing concentrations were also found for ringed seals from northwest Greenland until 2006 (Rotander et al. 2012a). In East Greenland, however, these compounds continue to increase (Rigét et al. 2013a). Differences in trends of PFOS and PFCAs might be related to continuing emissions of PFCAs or their precursors (Hart et al. 2009).

Further time trends have been established for hexabromocyclododecane (HBCD) in ringed seals from East Greenland, Beaufort Sea, Lancaster Sound, and Hudson Bay, showing rapid concentration increases (Muir et al. 2013; Vorkamp et al. 2012). The Canadian data also indicated increasing concentrations of 1,2-bis (2,4,6-tribromophenoxy)-ethane (BTBPE), a replacement product of OctaBDE (Muir et al. 2013). For both East Greenland and Canadian ringed seals, decreasing concentrations of polychlorinated naphthalenes have been found (Muir et al. 2013; Rotander et al. 2012b).

Time trends of Hg in ringed seals of the Canadian Arctic were recently reviewed (Chételat and Braune 2012). No statistically significant trend could be detected, possibly due to large inter-annual variation of Hg levels in ringed seal liver. The longer time series from Ulukhaktok examined ringed seal muscle, but did not find time-dependent changes of total Hg either, for the years 1973–2007 (Gaden et al. 2009). Time series from Greenland detected a significant increase of Hg in ringed seals from central East Greenland (Rigét et al. 2012). Hg concentrations also increased in ringed seals from northwest Greenland between 1984 and 2010, but the trend was only close to being significant. In ringed seals from central West Greenland, on the other hand, Hg concentrations decreased significantly (Rigét et al. 2012). Figure 12.6 summarizes time series for Hg from two locations in Canada and from central East and West Greenland based on data from Dietz et al. (1998a), Rigét et al. (2012) and unpublished data by Muir et al. An overall increase

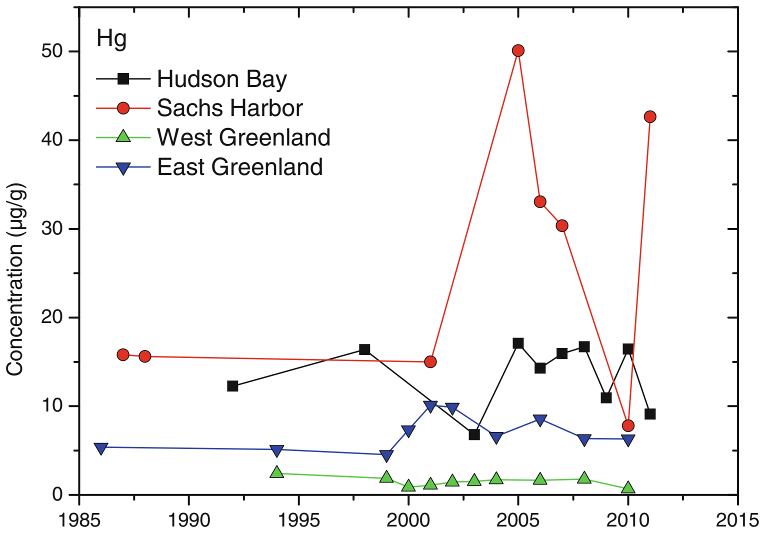

Fig. 12.6 Concentration of mercury (geometric means and arithmetic means in µg/g wet weight) in ringed seals from Sachs Harbor (Beaufort Sea) and Arviat (Hudson Bay) as well as West and East Greenland. Data from Dietz et al. (1998a), Muir et al. (unpublished data) and Rigét et al. (2012)

of Hg in ringed seals for the Arctic since medieval times was documented by Outridge et al. (2009) based on the analysis of seal teeth.

Potential impacts of climate change on contaminant trends are topics of increasing research. The seal population at Ulukhaktok showed increasing trends of PCBs and *p,p′*-DDE toward years of early sea-ice breakup, which might lead to earlier foraging in addition to a larger foraging radius (Gaden et al. 2012). Similarly, CB-153 in ringed seals from West Greenland showed a negative relationship with number of sea-ice days (Rigét et al. 2013b). Other physical parameters such as the Atlantic Oscillation Index (AOI) were also associated with organochlorine and Hg concentrations (Rigét et al. 2012, 2013b). Higher Hg concentrations in ringed seals from Ulukhaktok were found following years with short and long ice-free seasons (Gaden et al. 2009). The authors discuss potential relationships with Arctic cod populations, which might be biased toward older—and thus more contaminated—fish in years of both few and many ice-free days. Climate and contaminant associations are complex as they include direct impacts on contaminant transport and availability as well as indirect effects on food web structures and ecological processes.

Health Effects of Contaminants on Ringed Seals in the Arctic

It is recognized that ringed seals in the Arctic are exposed to heavy metals and organic contaminants at levels which can cause adverse health effects (Fisk et al. 2005; Sonne-Hansen et al. 2002). However, information on effects in Arctic ringed seal is sparse. At lower latitudes, multiple effects of organic contaminants in particular have been observed for ringed seals from the Baltic Sea (Vos et al. 2000). Similarly, harbor seals from the Wadden Sea were severely affected by organic contaminants (Laane et al. 2013). Effects on ringed seals included reproductive impairment, impaired immune function, pathological lesions, skeletal deformities, and biochemical changes (Basu et al. 2006; Vos et al. 2000). Reproductive disorders were particularly severe, with 70 % of female ringed seals subjected to autopsy suffering from uterine stenosis and occlusions (Vos et al. 2000). According to Routti et al. (2008b), uterine occlusions still occur in about a third of adult female ringed seals in the Baltic Sea, rendering them sterile for life.

Field studies showed a cause–effect relationship between organochlorine compounds in ringed seals and reproductive toxicity (Vos et al. 2000). PCBs were ascribed a stronger effect than DDT compounds as high DDT exposure had apparently also been present prior to reproductive failure. However, $MeSO_2$-DDE was found to be a potent adrenal toxicant, possibly involved in the cause of Cushing's disease in ringed seals from the Baltic Sea (Vos et al. 2000).

Ringed seals from the Arctic have been included in some studies to contrast the findings for the Baltic Sea animals, as discussed above for the toxicokinetic processes (Nyman et al. 2000; Routti et al. 2008a, 2009a). Studying potential associations between POP exposure and bone- and thyroid-related parameters, Routti et al. (2008b) found differences in hepatic vitamin D3, circulating $1,25(OH)_2D$—a metabolite of vitamin D3—and thyroid hormone levels between ringed seals from the Baltic Sea and from Svalbard. For the animals from Svalbard, no correlation was observed between any of these parameters and the POP levels.

A chemical cocktail resembling concentrations and profiles in Arctic ringed seals was used for in vitro tests of reproductive impairment in a pig cell system (Campagna et al. 2001). The study showed effects on the maturation of oocytes and their subsequent development, with potential consequences for fertilization and embryo development. As these findings were in line with laboratory tests, the authors expect them to also occur in vivo.

As many organochlorines are neurotoxicants, neurological effects have been studied in vitro, in terms of inhibition of the muscarinic cholinergic receptor in cellular membranes isolated from the cerebrum of ringed seals from Arctic Canada (Basu et al. 2006). Slight effects were found for toxaphene, at the highest concentration of 320 μM, but for none of the other organochlorines studied. $MeHg^+$ and Hg^{2+}, however, showed significant effects. The authors noted that behavioral and physiological consequences of such alterations were unknown (Basu et al. 2006).

No effects were found on the skeleton system of ringed seals from North West Greenland, despite high Cd exposure (Sonne-Hansen et al. 2002). The authors discussed effective detoxification mechanisms, possibly favored by a ringed seal diet which is rich in vitamin D, calcium, phosphorous, zinc, and proteins. However, metallothionein (MT), an intracellular protein with a high metal-binding capacity and a common biomarker of heavy metal exposure, only bound 2–15 % of metals in the liver and kidney of ringed seals, which is considerably less than found for other marine mammals (Sonne et al. 2009). The authors discussed that MT-upregulating might be insufficient in ringed seals or alternative detoxification mechanisms might exist, involving, e.g., selenium or glutathione (Sonne et al. 2009).

In summary, the limited number of biological effect studies on ringed seals in the Arctic makes any risk assessment challenging. Concentrations of organochlorines and heavy metals exceed threshold values associated with adverse effects (Fisk et al. 2005). Furthermore, experience from Europe shows that ringed seals are susceptible to a range of adverse effects caused by organochlorine exposure. Organochlorine concentrations are lower in the Arctic and show generally decreasing trends, but additional stressors (climate change etc.) may be significant.

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Chapter 13
Concluding Remarks

Roland Kallenborn

Initiated by the International Polar Year (IPY 2007–2009), a large number of projects in environmental Polar research were performed. Within this project cluster, several projects on the fate and distribution pathways of environmental pollutants were performed and successfully concluded. The here-presented book consists of both project descriptions from research initiatives setup as IPY projects and projects associated with IPY initiatives utilizing joint logistical support, hardware, and technology. Some of these projects are still continuing as long-term scientific commitments and represent thus strong legacy aspects of IPY.

A representative and comprehensive selection of research activities is represented here, however, with a strong focus on the Arctic environment. The documented research field covers many scientific disciplines, from social sciences, education, and anthropology to toxicology, environmental chemistry, and photochemistry. Nevertheless, the eleven studies described here in separate chapters are only to be considered as a small but representative selection of all environmental pollutant-related projects initiated via IPY 2007–2009.

Large international project initiatives directly funded via national IPY funding programs, such as

OASIS: “*The Ocean–Atmosphere–Sea Ice–Snowpack*” *project.*
COPOL: “*Contaminants in Polar regions*”
GEOTRACES: “*Tracers of Climate Change in the Arctic*”
POLARCAT: “*Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, of Climate, Chemistry, Aerosols, and Transport,*”

could not be adequately represented in the here-provided overview.

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R. Kallenborn (ed.), *Implications and Consequences of Anthropogenic Pollution in Polar Environments*, From Pole to Pole, DOI 10.1007/978-3-642-12315-3_13

However, the intention with the here-provided overview in our book "Implications and Consequences of Anthropogenic Pollution in Polar Environments" was to provide a scientific insight into the rich and scientifically complex, interdisciplinary research on Arctic pollutants initiated by the International Polar Year. These aspects are illustrated completely in the here-provided overview. Furthermore, many of the projects represented in our studies are considered as long-term scientific commitments, as such they are vivid examples for the legacy of IPY now and in the years and decades to come. These long-term aspects of many of the here-presented research activities are illustrating impressively the scientific, societal, and cultural value of this important initiative as an outstanding heritage hopefully visible and actively used by the coming generations of Polar scientists committed to research on the anthropogenic footprint in the most remote regions of our planet, the Polar regions.

Erratum to: A Circumarctic Review of Contaminants in Ringed Seals

Katrin Vorkamp and Derek C.G. Muir

Erratum to:
Chapter 12 in: R. Kallenborn (ed.), *Implications and Consequences of Anthropogenic Pollution in Polar Environments*, From Pole to Pole, DOI 10.1007/978-3-642-12315-3_12

The book was inadvertently published without incorporating the corrections in Chap. 12. The erratum book and the chapter has been updated.

The updated original online version for this chapter can be found at
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