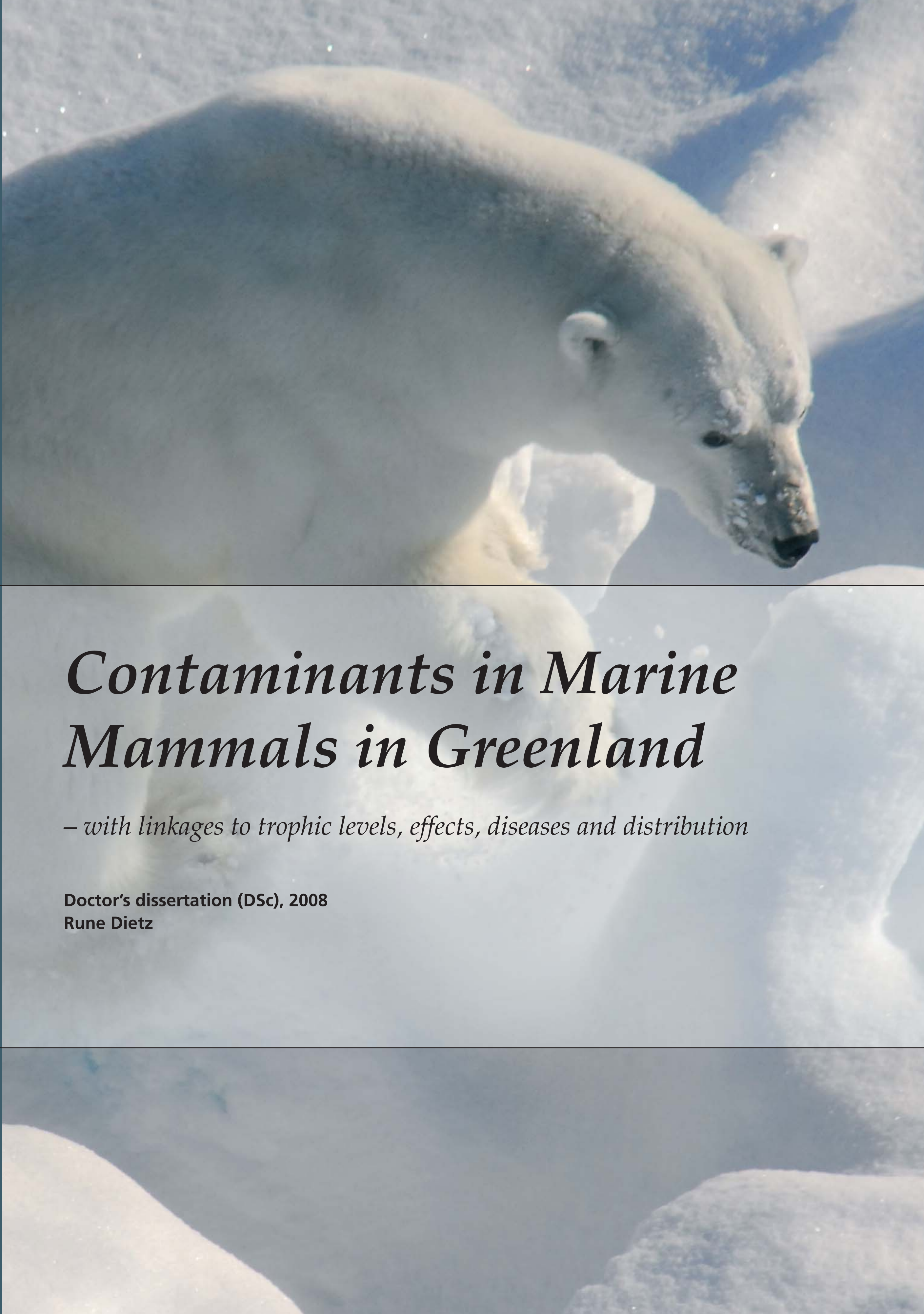

Contaminants in Marine Mammals in Greenland

– with linkages to trophic levels, effects, diseases and distribution

Doctor's dissertation (DSc), 2008
Rune Dietz

Denne afhandling er af Det Naturvidenskabelige Fakultet ved Københavns Universitet antaget til offentligt at forsvares for den naturvidenskabelige doktorgrad.

København den 5. februar 2008.
Nils O. Andersen, Dekan

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Contents


Photo: R. Dietz

Summary/ sammenfatning

Summary

The present dissertation provides a review of key determining parameters (age, sex, season, food and climate), trends (geographic and temporal), bioaccumulation, human exposure and effects of contaminants in top predators in the Greenland marine ecosystem. Furthermore, the dissertation links the contaminant issue to marine mammal distribution and stock separations monitored mainly by satellite telemetry. The review and conclusions are based on 30 key publications as well as selected supporting literature.

Setting the stage

The Arctic has previously been regarded a pristine environment. It is a region with limited industry, almost no agriculture and only a few local areas where some organohalogenated compounds (OHCs) have been used for pest control. However, during the 1970s and 80s it became evident that contaminants such as heavy metals and OHCs were present in significant concentrations in the higher trophic levels of marine ecosystems and in Inuit populations that use them for food. Since then, a substantial effort has been addressed to resolve the contaminant questions relating to origins, transport, geographical and temporal trends as well as toxicity and biological effects. The Arctic has become an important place to study contaminants, well suited for the study of chemical persistence, bioaccumulating and biomagnificating properties, long-range transport and adverse effects that are important criteria identified under international agreements and Conventions aimed at regulating OHCs or persistent organic pollutants (POPs).

The focus area

Some of the highest human exposure levels to persistent toxic contaminants are found in the Arctic. This is due to the long-range transport of contaminants to the region, long marine food chains that include slow-growing species, and the fact that marine mammal predators at the top of these food-chains constitute an important part of the Inuit consumer's food intake. In addition, the focus on the Greenland ecosystem is of major importance as the Greenland Inuit population were found to have the highest exposures of any people in the Arctic when it comes to Hg, PCB, DDE, oxychlordane and toxaphene. Diets including marine mammals were identified as the major source of the contaminant exposure to the Greenlanders and other Arctic populations. Therefore, the marine environment, and in particular species at the higher trophic levels, such as certain marine mammals, including polar bears, became a focus of my work over the past years, and therefore also the subject of this dissertation. Data are also presented for lower trophic levels that were part of the screening of the entire ecosystem and provide information that explains where the exposures are the highest.

Photo 1. Marine mammals, including polar bears are the focus of this dissertation due to their hight loads of contaminants and hence their chance for displaying effect due to their high trophic position and their high regional exposure.
Photo: R. Dietz.

Structure of the dissertation

This dissertation addresses three main topics: 1) Marine contaminant loads, 2) Contaminants related pathological effects and diseases, and 3) Marine mammal migration and stock separations.

Conclusions

The main conclusions are:

(i) Basic parameters such as age and sex of the animal, tissue type, season of collection, affect contaminant loads. It was documented that older animals tend to have higher concentrations of Hg and Cd and for some OHC groups adult males tend to have the higher concentrations in the Greenland marine ecosystem. Mercury concentrations are highest in liver, Cd is highest in kidney and OHC are highest in adipose tissue or liver. Seasonal differences may in some cases be substantial and should be taken into account in geographical and temporal trend comparisons.

(ii) Ecosystems, differences in trophic level, bioaccumulation and climatic differences will have an affect on contaminant loads. Due to the longer food chains and hence higher trophic position of most marine top predators, Hg, Cd and OHC loads are higher than those found in the terrestrial ecosystem. There is clear evidence of bioaccumulation of Hg, OHCs, and to certain extent Cd throughout the Arctic marine food chain. Differences in trophic level of food, which can also be associated with climatic change or variability, is important information that needs to be taken into account in geographical and temporal trend comparisons, and predictions of future trends.

(iii) Geographical patterns can be detected in contaminant loads within Greenland and other Arctic marine mammal populations. Northwest Greenland and the central Canadian Arctic have the highest concentrations of Hg; Central West Greenland and Northwest Greenland have the highest concentrations of Cd; while East Greenland together with Svalbard and still further east the Kara Sea have the highest loads of most lipophilic OHCs. This information provides an indication of where possible effects of contaminants due to high levels are most likely to occur, and where the lowest exposed animals for use as reference groups may be found.

(iv) Temporal trends in contaminant loads are detectable in key species in the Greenland ecosystem. Long-term studies in appropriate media reveal increases of Hg with a substantial anthropogenic contribution. These increases appear to be continuing in Northwest Greenland and the Central Canadian Arctic. Mercury levels east of Greenland and levels of "legacy OHCs", such as PCBs, DDTs, HCHs, HCB, chlordanes, dieldrin, and coplanar PCBs are showing declines. Time series on toxaphene, PCDDs and PCDFs are more uncertain, but may be decreasing. Increases in concentrations of a number of "new" OHCs such as the PBDEs and the PFCs took place prior to the turn of the millennium in the entire Arctic. PFCs continue to increase in Greenland, but there is some evidence that in recent years, PFCs and PBDEs may have decreased again in some areas.

(v) The most highly exposed groups in the Arctic ecosystem, the top-level carnivores, are affected by contaminants. Mercury levels are high enough to cause effects in some top predators. Neuropsychological dysfunction in humans and the first histopathological and neuro-chemical receptor biomarker investigations indicate effects of Hg, but these are subtle effects and more investigations are needed. Selenium, being present in surplus in the Arctic marine ecosystem, is likely to reduce the effect of Hg. Although Cd concentration in several marine species is above threshold levels for effects, Cd has not yet been shown to cause effects in Arctic wildlife. Examples of effects from high exposure to OHC include reduced size of reproductive organs, tissue alterations found in liver and kidney, reduction of bone mineral density, and impairment of the immune system. However, no linkage could be documented between contaminant exposure and pseudohermaphroditism, immunological response and skull pathology in East Greenland polar bears. Skull asymmetry showed linkages to contaminants in only some of the investigations. In well defined mass mortality events, such as the two PDV outbreaks in recent years, it has not been possible to make a clear linkage between contaminants, immune suppression and the number of deaths caused by the disease. A large number of confounding factors can play a significant role for such disease events.

(vi) The Inuit population can minimize their contaminant intake and risk of health problems by reducing their intake of internal organs (Hg, Cd, PBDEs and PFCs), adipose tissue (OHCs), and preferential consumption of lower trophic species. Intake of young animals will result in lower Cd and Hg and in some cases OHC exposure. For OHCs adult females will be less polluted compared to adult males. At the same time, these foods are sources of important nutrients and changes in diet can bring other health risks.

(vii) Marine mammal distribution is of major importance in planning contaminant studies and in interpreting results of such studies. In some regions contaminant samples and samples for investigation of effect parameters can only be obtained during tagging operations. Satellite tagging together with contaminant analysis in samples from the same animals has the potential for linking contaminant levels with dispersal, behaviour and possible effects on the tagged animals. In cases where tagging has proven difficult to conduct, genetics and contaminants analyses can be used to elucidate population relationships.

Sammenfatning

Nærværende afhandling giver en sammenfatning over nøgleparametre (alder, køn, årstid, fødevalg og klima), trends (geografisk og tids), bioakkumulering, human eksponering og effekter af kontaminanter i toppredatorer i det grønlandske marine økosystem. Ydermere knytter afhandlingen kontaminant-emnet sammen med havpattedyrs fordeling og bestandsadskillelse, fortrinsvis undersøgt ved brug af satellittelemetri. Gennemgangen og konklusionerne er baseret på 30 videnskabelige afhandlinger og yderligere udvalgt dokumenterende litteratur.

Baggrund

Arktis har tidligere været betragtet som et "jomfrueligt" og uforurenet område, da det ligger fjernt fra industrielle kilder, stort set er uden landbrugsproduktion, og da pesticider ikke er nødvendige i dette område. Det blev imidlertid dokumenteret i 1970erne og 80erne at tungmetaller og organohalogen forbindelser (OHCs) forekom i betragtelige koncentrationer på de højere trofiske niveauer af det marine økosystem og i Inuit befolkningen. Siden da har der været lagt en betydelig forskningsindsats i forureningsspørgsmålet for at belyse emner så som kilder til emission, transportmekanismer, geografiske og tidsmæssige trends samt giftigheden og biologiske effekter. Arktis har vist sig at være et vigtigt område i studiet af kontaminanter, da kriterier så som kemisk persistens, bioakkumulering og biomagnificering, fjerntransport og skadelige effekter, som er nøgleparametre i regulerings- og konventionsarbejdet, er særdeles velegnede at studere i de arktiske områder.

Baggrunden for fokuseringen på de grønlandske havpattedyr

Nogle af de højeste humane eksponeringer finder sted i Arktis. Dette skyldes fjerntransporten af en lang række kontaminanter, de lange marine fødekæder, nedsatte vækstprocesser og de trofisk højtliggende havpattdyrs fødemæssige betydning for den arktiske Inuit befolkning. Ydermere har netop det grønlandske økosystem været vigtigt at undersøge, da Inuit befolkningen i Grønland har de højeste indhold af stoffer som Hg, PCB, DDE, oxychlordane and toxaphene sammenholdt med andre områder af Arktis. Havpattedyr har vist sig at være hovedkilden til disse høje niveauer i grønlændere og andre arktiske befolkningsgrupper. Derfor er det marine miljø og specielt havpattedyrene blevet centrale i mit arbejde og i nærværende afhandling. Vi har dog undersøgt visse lavere trofiske niveauer for at dokumente forskelle og opkoncentreringen via fødekæderne.

Afhandlingens disponering

Denne afhandling omfatter tre hovedemner: 1) Marine kontaminant niveauer, 2) Kontaminanters relaterede effekter og sygdomme og 3) Havpattedyrs vandringer og bestandsforhold.

Konklusioner

De væsentligste konklusioner er:

(i) Basale parametre så som alder, køn, vævstyper og årstiden vil påvirke kontaminantniveauerne. Det blev således dokumenteret at ældre dyr synes at have højere niveauer af Hg og Cd, hvilket også er tilfældet for visse OHC grupper i voksne hanner i det grønlandske marine økosystem. Kviksølv koncentrationer er generelt højest i lever, Cd er højest i nyrer, mens OHC ligger højest i fedtvæv eller leveren. Årstidsbetingede forskelle kan i visse tilfælde være betragtlige, og disse variationer bør der således tages højde for i geografiske og tidmæssige sammenligninger.

(ii) Økosystemer, forskelle i trofisk niveau, bioakkumulering og klimatiske ændringer vil have en betydning for kontaminant-niveauerne. På grund af de længere fødekæder og dermed højere forekommende trofiske niveauer vil de fleste marine toppredatorer have højere Hg, Cd og OHC niveauer i det marine end i det terrestriske økosystem. Der forekommer en tydelig bioakkumulering af Hg, OHCs og i et vist omfang af Cd i de arktiske marine fødekæder. Forskelle i fødevalg og klimatiske forhold bør ligeledes indgå i undersøgelser af geografiske forskelle, tidsserier og fremskrivning af kontaminternes udvikling.

(iii) Geografiske forskelle forekommer i kontaminant-belastningen mellem grønlandske og andre arktiske pattedyrpopulationer. Nordvestgrønland og det nordlige del af det centrale arktiske Kanada har de højeste Hg niveauer, Central- og Nordgrønland har de højeste Cd niveauer, mens Østgrønland, Svalbard og Kara Havet har de højeste niveauer af de fleste lipofile OHCer. De geografiske og fødekædemæssige mønstre er retningsgivende for hvor man bør undersøge mulige effekter og hvor referenceområder bør udvælges.

(iv) Det er muligt at påvise forskelle i kontaminant-niveauer over tid for nøglearter i det grønlandske økosystem. Historiske tidsserier har således påvist stigninger i Hg-niveauerne, hvoraf en betydelig andel er menneskeskabt. Stigninger synes at fortsætte i Norvestgrønland og i den centrale del af arktisk Kanada. Kviksølv øst for Grønland og PCB, DDT, HCH, HCB, klordan, dieldrin, and coplanare PCBer er faldende. Tidsserier af toxaphen, PCDDer and PCDFer mere usikre, men niveauerne falder formentlig ligeledes. En række af de "nye" OHCer så som PBDEer og PFCer synes at stige op til årtusindskiftet i hele Arktis. PFC gruppen fortsætter med at stige i Grønland, mens stigningen over de senere år kan være aftaget og/eller vendt i andre områder.

(v) Tesen om at de højest eksponerede dyregrupper i det arktiske økosystem er påvirket af kontaminant-niveauerne er ligeledes blevet belyst. Kviksølv-niveauerne var på et nivau hvor effekter kunne forventes i visse toppredatorer. Neurofysiologiske effekter på mennesker og de første histopatologiske and neuro-kemiske receptor biomarkør undersøgelser antyder effekter af Hg, men yderligere undersøgelser bør foretages. Selen, som generelt forekommer i betydeligt omfang i de marine fødekæder, kan sandsynligvis nedbringe Hg-effekterne. Selv om Cd-koncentrationerne i mange marine arter er over effektniveauerne, er der endnu ikke påvist skader på det arktiske dyreliv. Der er imidlertid påvist en række effekter fra eksponeringen til høje niveauer af OHCer omfattende: Reduceret størrelse af kønsorganer, forandringer i lever- og nyrevæv, et fald i mineraltætheden i knogler samt beskadigelse af immunsystemet. Imidlertid kunne der ikke påvises effekter som pseudohermaphroditisme, effekter på immunologiske organer og patologiske effekter i kranier fra de østgrønlandske isbjørne. Kranie asymmetri viste kun en kobling med kontaminanter i nogle undersøgelser. Det var heller ikke muligt at foretage en direkte kobling mellem udbruddet af det største massedødfald blandt havpattedyr – sælpesten – og kontaminantniveauer. Dette skyldes at en lang række andre forhold har betragtelig betydning for omfanget af dødfaldene fra denne virus.

(vi) Inuit befolkningen kan nedbringe deres indtag af kontaminanter og dermed risiko for sundhedsproblemer ved at reducere indtaget af lever og nyre (Hg, Cd, PBDEer og PFCer), spæk og fedtvæv (OHCer) og ved i højere grad at spise dyr fra et lavere trofisk niveau. Ved at spise yngre dyr kan man desuden nedbringe indtaget af Cd, Hg og visse OHCer. Desuden vil OHC-niveauerne generelt være lavere i voksne hunner end i voksne hanner.

(vii) Migrationer og bestandsforhold er vigtige at kende, når man undersøger havpattedyr for kontaminanter. I visse områder er det kun muligt at få prøver til kontaminant- og effektundersøgelser når dyrene håndteres under mærkninger. Koblingen mellem satellitsporede dyr og kontaminantanalyser fra de samme dyr kan bibringe oplysninger om kontaminant-eksponeringen, forekomsten, adfærd og mulige efffekter på de mærkede dyr. I tilfælde hvor mærkning volder problemer kan analyser af genetik og kontaminanter bidrage til at belyse populationsrelationer.

List of papers

The present dissertation is based on the following 30 publications. They will be cited in the text by their numbers (**Paper 1,…30**). In addition they have been categorised according to one or more of the three thematic topics used in this dissertation: 1) Marine contaminant loads, 2) Contaminant effects and diseases and/or 3) Marine mammal distribution and stock separations. The topic is indicated at the end of the reference in brackets **[1, 2 or 3]** in the publication overview below and in the reference list for papers that I have coauthored.

The 30 articles are available as pdf files on the included CD together with an electronic version of this dissertation.

Paper #), Reference and [Topic]:

1) Dietz, R., C.T. Hansen, P. Have & M.-P. Heide-Jørgensen 1989a. Clue to seal epizootic? – Nature 338: 627. **[2]**
2) Dietz, R., M.-P. Heide-Jørgensen & T. Härkönen 1989b. Mass Deaths of Harbor Seals (*Phoca vitulina*) in Europe. – Ambio 18 (5): 258-264. **[2]**
3) Nielsen, C.O. & R. Dietz 1989. Heavy metals in Greenland seabirds. – Meddelelser om Grønland, Bioscience 29: 26 pp. **[1]**
4) Dietz, R., C.O. Nielsen & M.M. Hansen & C.T. Hansen 1990. Organic mercury in Greenland birds and mammals. – Science of the Total Environment 95: 41-51. **[1]**
5) Hansen, C.T., C.O. Nielsen, R. Dietz & M.M. Hansen 1990. Zinc, Cadmium, Mercury and Selenium in Minke Whales, Belugas, and Narwhals from West Greenland. – Polar Biology 10: 529-539. **[1]**
6) Dietz, R., M.-P. Heide-Jørgensen, J. Teilmann, N. Valentin & T. Härkönen 1991. Age determination in European Harbour seals *Phoca vitulina* L. – Sarsia 76: 17-21. **[1]**
7) Dietz, R., M.-P. Heide-Jørgensen E.W. Born & C.M. Glahder 1994. Occurrence of narwhals (*Monodon monoceros*) and white whales (*Delphinapterus leucas*) in East Greenland. – Meddelelser om Grønland, Bioscience 39: 69-86. **[3]**
8) Dietz, R., E.W. Born, C.T. Agger & C.O. Nielsen 1995a. Zinc, cadmium, mercury, and selenium in polar bears (*Ursus maritimus*) from Central East Greenland. – Polar Biology 15: 175-185. **[1]**
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12) Dietz, R., J. Pacyna, D.J. Thomas, G. Asmund, V. Gordeev, P. Johansen, V. Kimstach, L. Lockhart, S.L. Pfirman, F. Riget, G. Shaw, R. Wagemann & M. White 1998a. Chapter 7: Heavy metals. In: AMAP Assessment Report: Arctic Pollution Issues. – Arctic Monitoring and Assessment Programme. Oslo, Norway: 373-524. **[1]**
13) Dietz, R., P. Paludan-Müller, C.T. Agger & C.O. Nielsen 1998b. Cadmium, mercury, zinc and selenium in ringed seals (*Phoca hispida*) from Greenland and Svalbard. – NAMMCO Scientific Contributions 1: 242-273. **[1]**
14) Dietz, R., J. Nørgaard & J.C. Hansen 1998c. Have Arctic Marine Mammals Adapted to High Cadmium Levels? – Marine Pollution Bulletin 36(6): 490-492. **[1, 2]**
15) Dietz, R., F. Riget & E.W. Born 2000a. An assessment of selenium to mercury in Greenland marine animals. – Science of the Total Environment 245: 15–24. **[1]**

16) Dietz, R. F. Riget & E.W. Born 2000b. Geographical differences of zinc, cadmium, mercury and selenium in polar bears (*Ursus maritimus*) from Greenland. – Science of the Total Environment 245: 25-48. **[1]**

17) Dietz, R., F. Riget, M. Cleemann, A. Aarkrog, P. Johansen & J.C. Hansen 2000c. Comparison of contaminants from different trophic levels and ecosystems. – Science of the Total Environment 245: 221–232. **[1]**

18) Dietz, R., M.-P. Heide-Jørgensen, P. Richard & M. Acquarone 2001. Summer and Fall Movements of Narwhals (*Monodon monoceros*) from Northeastern Baffin Island towards Northern Davis Strait. – Arctic 54(3): 246-263. **[3]**

19) Dietz, R., F. Riget, K. Hobson, M.-P. Heide-Jørgensen, P. Møller, M. Cleemann, J. de Boer & M. Glacius 2004a. Regional and inter annual patterns of heavy metals, organochlorines and stable isotopes in narwhals (*Monodon monoceros*) from West Greenland. – Science of the Total Environment 331(1-3): 83-105. **[1, 3]**

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21) Sonne, C., R. Dietz, E.W. Born, F. Riget, M. Kirkegaard, L. Hyldstrup, R.J. Letcher & D.C.G. Muir 2004b. Is Bone Mineral Composition Disrupted by Organochlorines in East Greenland Polar Bears (*Ursus maritimus*)? – Environmental Health Perspectives 112(17): 1711-1716. **[1, 2]**

22) Bossi, R., F. Riget & R. Dietz 2005b. Temporal and Spatial Trends of Perfluorinated Compounds in Ringed Seal (*Phoca hispida*) from Greenland. – Environmental Science & Technology 39: 7416-7422. **[1]**

23) Riget, F., J. Vikelsøe & R. Dietz 2005. Levels and temporal trends of PCDD, PCDFs and non-ortho PCBs in ringed seals from East Greenland. – Marine Pollution Bulletin: 1523-1529. **[1]**

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26) Härkönen, T., R. Dietz, P. Reijnders, J. Teilmann, P. Thompson, K. Harding, A. Hall, S. Brasseur, U. Siebert, S.J. Goodman, P.D. Jepson & T.D. Rasmussen 2006. Review of the 1988 and 2002 Phocine Distemper virus epidemics in European habour seals. – Diseases of Aquatic Organisms 68: 115-130. **[1, 2]**

27) Sonne C., R. Dietz, H.J.S. Larsen, K.E. Loft, M. Kirkegaard, R.J. Letcher, S. Shahmiri & P. Møller 2006b. Impairment of Cellular Immunity in West Greenland Sledge Dogs (*Canis familiaris*) Dietary Exposed to Polluted Minke Whale (*Balaenoptera acutorostrata*) Blubber. – Environmental Science & Technology 40: 2056-2062. **[1, 2]**

28) Dietz, R., F. Riget, C. Sonne, D.C.G. Muir, S. Backus, E.W. Born M. Kirkegaard & R.J. Letcher 2007. Age and Seasonal Variability of polybrominated diphenyl ethers in free-ranging East Greenland Polar Bears (*Ursus maritimus*). – Environmental Pollution 146 (1): 177-184. **[1]**

29) Dietz, R., F. Riget, M.T. Olsen, D. Boertmann, C. Sonne, M. Kirkegaard, G. Asmund, K. Falk, J. Fjeldsaa, C. Egevang, F. Wille & S. Møller 2006b. Time trend of Mercury in Feathers of West Greenland Birds of Prey During 1859-2003. – Environmental Science & Technology 40: 5911-5916. **[1]**

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Photo: R. Dietz

1

Introduction

Setting the stage

The Arctic is not a pristine environment

The Arctic has previously been regarded a pristine environment, with limited industry, almost no agriculture, and only very localised use of some persistent organic compounds for pest control. As early as the 1970s it became evident, that contaminants such as certain heavy metals and persistent organic pollutants (POPs), in particular organohalogenated compounds (OHCs), were present in high concentrations in higher trophic level species and in the Inuit population (e.g. Addison & Brodie 1973, Addison & Smith 1974, Koeman et al. 1973, 1975, Smith & Armstrong 1975, 1978, Helle et al. 1976a, 1976b, Addison & Brodie 1977, Olsson 1978, Johansen et al. 1980, Reijnders 1980, Born et al. 1981, Helle 1981, Eaton & Farant 1982). Since then, a substantial effort has been directed at resolving the questions relating to sources, transport, geographical and temporal trends and effects of "old" or "legacy" and "new" and "emerging" contaminants.

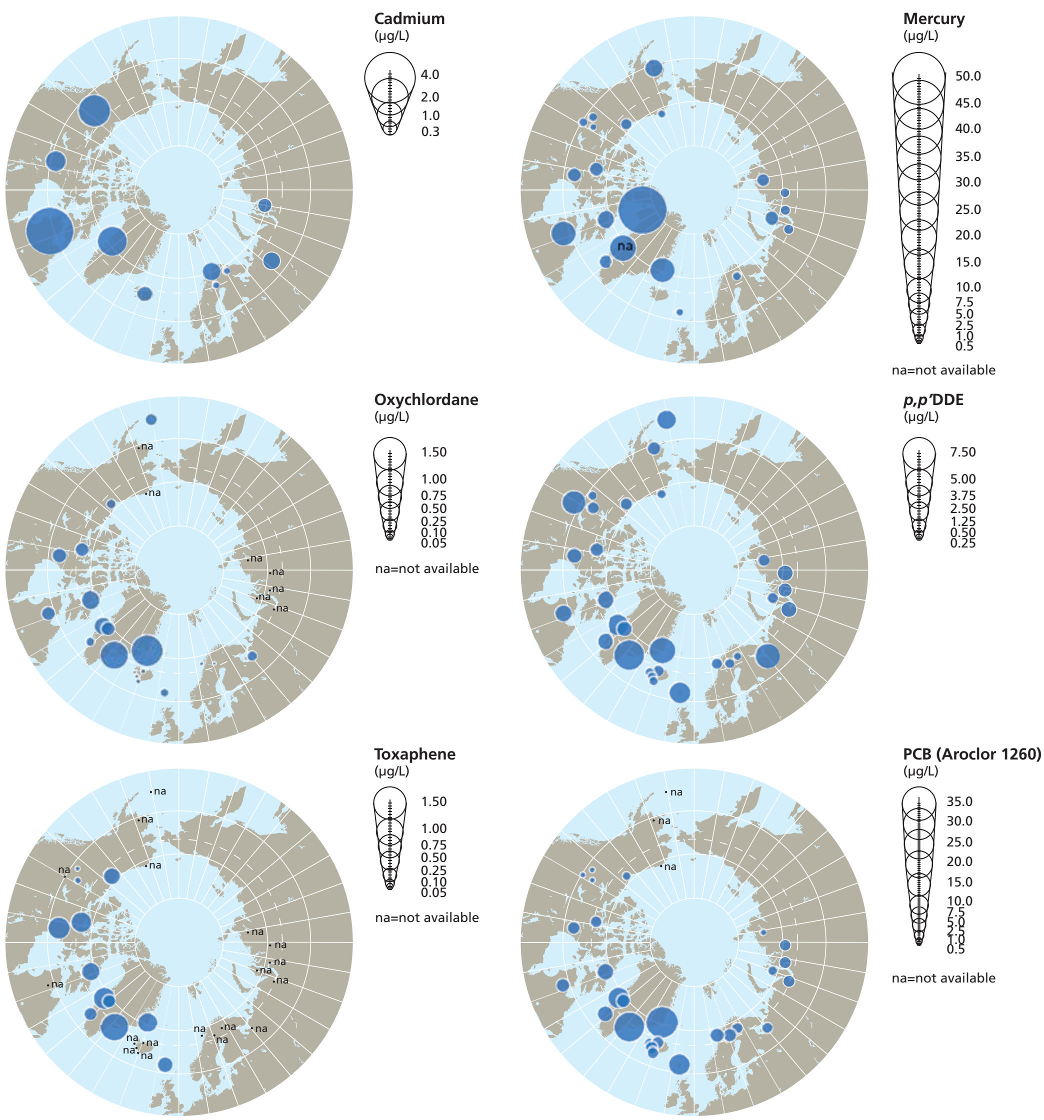

Fig. 1. Contaminants concentrations in Arctic human blood (Hansen et al. 1998, Ostdam & Tremblay 2003).

Why study contaminants in the Greenland marine ecosystem?
There are a number of reasons why it is important to study contaminants the Greenland/Arctic marine ecosystem. One major reason is that this area has some of the highest human exposure levels (for certain heavy metals and OHCs) in the world. This is due to the long marine food chains, in which higher trophic level marine mammals form an important part of the Inuit food intake (Hansen et al. 1998, Ostdam & Tremblay 2003). In addition, the focus on Greenland ecosystems is of major importance as the Greenlandic Inuit population has the highest levels within the Arctic when it comes to levels of Hg, Oxychlordane, DDE, Toxaphene and PCB in their blood (see Fig. 1 and Hansen et al. 1998, Ostdam & Tremblay 2003). Finally, the criteria used in global agreements and Conventions to define chemicals that have the characteristics of "persistent organic pollutants" include presence in locations "distant from sources" and "monitoring data showing that long-range environmental transport of the chemical … may have occurred". The Arctic has therefore become an important indicator region for assessment of persistence and bioaccumulation. The Arctic environment is well suited as a region in which to evaluate OHCs. Cold conditions favour persistence of OHCs relative to temperate or tropical environments (de Wit et al. 2004).

Levels of some contaminants in some Arctic human populations, and in particular populations in parts of Greenland are high enough that they may potentially affect children's mental development and resistance to infections (Dewailly & Weihe 2003). In addition, many OHCs (e.g. PCBs, DDTs, HCHs, Chlordanes and Toxaphenes) have been associated with disruption of hormones that are important for growth and sexual development (de March et al. 1998, de Wit et al. 2004). Most of the OHCs, and a substantial part of the Hg that is found in the Arctic environment and its ecoystems, reaches the Arctic as a result of long-range transport (by air or water). Hence the Arctic is an excellent monitoring region for quantifying the magnitude of these transports and providing information on trends in emissions, since interference due to contamination from local sources is minimal. The fact that exposures are high also makes the Arctic important as an "early-warning" region for health effects likely to appear at southern latitudes at a later stage if pollution continues to increase.

I have had the privilege to be involved in this work over the past quarter of a century.

Contaminants included in the dissertation

The contaminants dealt with in the present dissertation include the heavy metals Hg and Cd as well as many OHCs. Se is dealt with in relation to its protective properties against Hg. Lead and radioactivity are not included as Pb is not bioaccumulating in the marine ecosystem and levels of radioactivity are low in the Greenland marine ecosystem (**Paper 10, 12, 17**).

Heavy metals

Mercury properties and use
Mercury exists in nature as elemental mercury (Hg^0), and as inorganic and organic mercury compounds (O'Driscoll et al. 2005). Due to its chemical inertness, volatility and low solubility in water, gaseous Hg^0 has a relatively long atmospheric residence time (1–3 years), resulting in high long-range atmospheric transport potential and hence a global distribution (e.g. Roos-Barraclough et al. 2002, Roos-Barraclough & Shotyk 2003). An estimated 200–300 tonnes of Hg per year from various human activities at mid-latitudes is transported to the Arctic by atmospheric processes, ocean currents, and rivers (**Paper 12**, Nilsson & Huntington 2002, Skov et al. 2004). It has been discovered that elemental Hg is deposited from the atmosphere in the Arctic each spring as a result of "mercury depletion events", showing that the Arctic acts as a sink for globally emitted Hg (Schroeder et al. 1998, Berg et al. 2001, Lindberg et al. 2001). Recent conversion to cleaner-burning power plants and use of fuels other than coal significantly reduced the emissions of Hg during the 1980s in Europe and North America (Nilsson & Huntington 2002). It has been

suggested that recent Hg increases in Arctic biota in West Greenland and the Central Canadian may be linked to Asian coal burning, as Hg emissions from China in particular, together with other Asian coal burning countries have been increasing, these countries now producing half of the world's anthropogenic Hg emissions to the atmosphere (Nilsson & Huntington 2002, Braune et al. 2005a, **Paper 25**).

Cadmium

Cadmium is emitted to the atmosphere predominantly as elemental Cd and cadmium oxide from coal combustion, nonferrous metal production and refuses incineration. The residence time of Cd in air however is relatively short (days to weeks), which may be the reason why Cd has not drawn the same attention as Hg as a global pollutant (**Paper 12**, WHO 2000).

OHCs

A number of criteria serve to define what is meant by "OHCs" identified and listed under the Stockholm Convention on Persistent Organic Pollutants. These criteria are that the chemicals must be persistent, bioaccumulating, have the potential for long-range environmental transport and have adverse effects (see additional information in de Wit at al. 2004). These criteria are likewise of major importance for the focus of the present dissertation.

The OHCs dealt with in this dissertation can be categorized into industrial products and by-products including polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB) and other chlorinated benzenes, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs); persistent chlorinated pesticides such as dichlorodiphenyltrichloroethane (DDT), chlordane, heptachlor, dieldrin and polychlorobornanes and camphenes (toxaphene); less persistent chlorinated pesticides such as hexachlorocyclohexanes (HCH); and "new" chemicals with OHCs characteristics such as polybrominated diphenyl ethers (PBDEs), perfluoroalkylsulfonates and perfluorochemicals (PFCs).

Industrial products

Chlorinated industrial chemicals and by-products

Persistent organochlorine contaminants, such as HCB and PCDD/F, are mainly produced as unwanted by-products of chemical processes and waste incineration. Others, such as PCBs and PBDEs, have been manufactured and used in large quantities because of their stability and flame-retardant properties. Further brief details on the various OHC groups are given below and when no other references are provided they are based on information from de March et al. (1998).

PCBs

PCBs were introduced in 1929. They are chemically stable and heat resistant, and were used worldwide as transformer and capacitor oils, hydraulic and heat exchange fluids, and lubricating and cutting oils. Open use is currently banned in all circumpolar countries, but there are still large amounts in permitted use in large capacitors and transformers. Current uses and disposal practices in the developing world are not well documented.

HCB

HCB is produced as a by-product in the production of a large number of chlorinated compounds, particularly lower chlorinated benzenes, and in the production of several pesticides. It had limited use in the 1960s as a fungicide.

PCDD/Fs

PCDD/Fs enter the environment as by-products of industrial processes. The most significant sources are low-temperature, incomplete incineration of chlorine-containing materials such as plastics. Other major sources include thermal processes, such as motor vehicle fuel combustion in countries where leaded fuel containing chlorine scavengers is still used, and metallurgical industries. Pulp and paper mills using chlorine in the bleaching process have been important sources to the aquatic environment of 2, 3, 7, 8-tetrachlorodibenzo-p-dioxin (2, 3, 7, 8-TCDD) and 2, 3, 7, 8-tetrachlorodibenzofuran (2,3,7,8-TCDF).

Chlorinated pesticides

Persistent pesticides

DDT

DDT was introduced in 1945 as an insecticide. Its use has been restricted in Canada, the USA, and Western Europe for nearly two decades; however, it is used in pest (in particular malaria mosquito) control programs in southern Asia, Africa, and Central and South America and may be used in China and Russia.

CHL

In the past, chlordane (CHL) was released into the environment primarily from its application as an insecticide and for seed dressings and coatings. In the USA, it was used extensively prior to 1983, and from 1983 to 1988 it was registered for termite control. It was cancelled for this use in 1988.

Dieldrin

Dieldrin was mainly used as a soil insecticide. It is no longer manufactured in Canada and the USA, and its use is now restricted for termite control. Manufacture in Europe, especially for export to developing countries, continued until the late 1980s. It is also a degradation product of aldrin, also no longer in use in circumpolar countries.

Toxaphene

The complex mixtures of polychlorobornanes and camphenes known as toxaphene were widely used in the USA on cotton crops. Use peaked between 1972 and 1975. Manufacture was banned in the USA in 1982 and uses ceased in 1986. Similar products have been, and may continue to be used in Mexico, Central America, Eastern Europe, and countries of the former Soviet Union.

HCH

Lindane (γ-HCH), the most biologically active insecticidal isomer, is the only form of HCH currently used in its pure form in North America, Japan and Europe, where it is used mainly in seed treatment. Other isomers have been banned for use in the USA and most other circumpolar countries since the late 1970s. Technical HCH is still used in China as an insecticide on hardwood logs and lumber, seeds, vegetables and fruits, and on existing buildings and structures.

"New" chemicals with OHC characteristics

Some of the "new" chemicals have been in use for a number of years. New refers mainly to their identification and quantification in environmental media, partly as methods for analysis have developed and improved.

PBDEs

PBDEs are used as flame retardants in polymeric materials. Some products that are flame-retarded are textiles, plastics, electrical equipment, building materials, and linings of vehicles. The increasing use of flame retardants in modern societies has led to increases of PBDEs in the environment.

PFCs

Carboxylated and sulfonyl-based perfluorochemicals (PFCs) including perfluoroalkanoic acids (PFAs) have been produced and used for over 40 years in a variety of consumer products and industrial materials. In 2000 the US Environmental Protection Agency (EPA) banned perfluorooctane sulfonate (PFOS) from the US market, and shortly thereafter the major manufacturer of PFCs, the 3M Company, announced a phase-out of the production of carboxylated and sulfonylbased perfluorochemicals from December 2000. Perfluorooctanoic acid (PFOA) and longer chain perfluorinated carboxylic acids (PFCAs) continue to be manufactured as emulsifiers and additives in the polymerization process, as the industry has not yet found a suitable replacement for these compounds (**Paper 22**, Dietz et al. 2008).

Historic overview

Why provide a historic overview?

In order to understand why and how different projects were conducted by Department of Arctic Environment (DAE) and collaborators, it is important to understand the changing circumstances and functions of our institute over the years. Our scientific involvement in contaminant work in the Greenland environment has evolved over the years as a function of working conditions, focus areas and funding possibilities, but also the state of knowledge, ongoing collaboration processes as well as the political and economic situation in Denmark and collaborating countries. During this period not all projects have been conducted or ideas for work followed-up, often due to funding limitations. In other cases higher priority projects have overruled further investigations of less important issues. This historic overview deals with some of the more important conditions and relations linked to DAE and Greenland contaminant-related issues over the past 25 years.

The earliest samplings and investigations
Prior to the mid-1980s, various studies of heavy metals and OHC pollution in Greenland marine mammals had been made (Clausen & Berg 1975, Johansen et al. 1980, Born et. 1981). However, these studies were conducted on an opportunistic basis and were not part of any regular monitoring programme. Samples collected further back in time have, however, subsequently proven very valuable for extended time trend analyses (e.g. **Paper 25, 29**).

Monitoring of mines
During the same period, monitoring of heavy metal contaminant levels in a number of marine species was started in relation to monitoring of mining activities at a number of sites in Greenland (Anon. 1988).

Heavy Metals in the Greenland Marine Environment
During the mid-1980s, we conducted the first large scale survey involving analysis of contaminants in samples from around Greenland during the programme: Heavy Metals in the Greenland Marine Environment (HMGME) supported by the Danish National Science Foundation and the Commission for Scientific Investigations in Greenland. As Greenlanders are primarily dependent on marine organisms as food resources (Kapel & Petersen 1982), focus of this and later investigations were dedicated to the marine ecosystem. The collections of the HMGME project included biological samples from the entire Greenland West Coast up to Avanersuaq (ca 78°N) and the East Coast north to Kong Oscars Fjord (ca 73°N) (with a few samples from Daneborg, ca. 74°30'N within the National Park). Focus in this study was given to geographical and trophic patterns in the Greenland marine biota, as a three-year study period was not likely

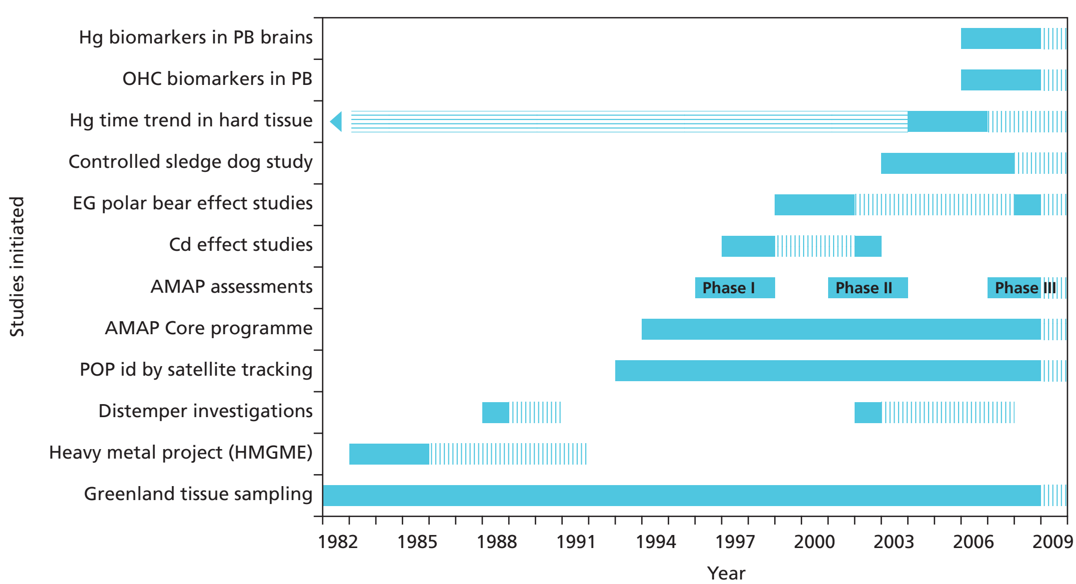

Fig. 2. Historic expansion of working fields over the last 25 years dealt with in the present dissertation.

to provide meaningful results on time trends. However, the samples obtained in this study have been important to our later time trend investigations. Although this was a large-scale investigation, it was only conducted on a national level. Also although OHCs were not part of the HMGME program, the necessary tissue samples (blubber and other lipid rich tissue) were obtained for later analysis (Fig. 2).

Photo 2. As Greenlanders are primarily dependent on high trophic marine organisms as food resources the primary focus of this dissertation is on the marine ecosystem. Photo: R. Dietz.

The Finnish initiative

In 1989, the protection of the circumpolar Arctic region and its inhabitants from adverse effect of human activities was addressed internationally. Finland convened an intergovernmental conference in Rovaniemi with participants from Canada, Denmark/Greenland, Iceland, Norway, Sweden, the Soviet Union and the United States (the eight member states of what would eventually become the Arctic Council). This meeting decided to produce the first "State of the Arctic Environment Report" addressing issues relating to the main pollutants. This first report (Anon. 1991) was presented at the first Arctic Ministerial Conference in Rovaniemi in 1991. This conference was considered a breakthrough in the international co-operation for the protection of the Arctic with the adoption of the Arctic Environmental Protection Strategy (AEPS 1991). The AEPS was followed up by Arctic Ministerial Conference until 1997, where The Arctic Council was formed.

The AMAP work

To implement the AEPS, four programs were initiated: Arctic Monitoring and Assessment Programme (AMAP), Conservation of the Arctic Flora and Fauna (CAFF), Emergency Prevention, Preparedness and Response (EPPR), Protection of the Arctic Marine Environment (PAME). Further programmes on Sustainable Development and Utilization (SDU) and Action Plan for Remediation (ACAP) were added later following the establishment of the Arctic Council. DAE became primarily involved with the AMAP process, including the monitoring programs and the AMAP Phase I and Phase II Assessments. The results of the Phase I were published in a popular science report (Nilsson 1997) and in a scientific background report, with chapters covering the priorty issues of concern, including OHC, heavy metals and human health (e.g. de March et al. 1998, **Paper 12**, Hansen et al. 1998). After five more years of investigations, the Phase II results were prepared, and again published in a popular science report authored by Nilsson and Huntington (2002) followed by a series of scientific assessment reports covering, among other issues, human health, OHCs and heavy metals (AMAP 2003, de Wit et al. 2004, Marcy et al. 2005). Along with the International Assessments, National Assessment Reports were produced addressing the Greenland and Faroese situation and scenarios (Dietz et al. 1997a, b, c, Johansen et al. 2003, Riget et al. 1997, 2003)

OHC focus in the Greenland ecosystem

OHC programs on a regular basis was first started at NERI under the AMAP programme, as most contaminant work with a few exceptions (e.g. Stern et al. 1994) previously was focused on heavy metals due to the historic linkage of the institutional work with resource related monitoring (primarily linked to mining activities). However, the adoption of the AMAP program and related research programmes contributed new valuable knowledge from the Greenland area (e.g. Cleemann et al. 2000a, b, c, d, **Paper 19, 20**, Johansen et al. 2004a, b, Krone et al. 2004, Sørensen et al. 2004, Bossi et al. 2005a, **Paper 22**, Glasius et al. 2005, **Paper 23**, Vorkamp et al. 2005, **Paper 28**).

New priority areas

Within the AMAP process, considerable scientific progress and new insights were achieved. Most of all, the consensus conclusions and recommendations have provided priorities for the continued and future programs and collaboration. Bringing scientists together from different countries and disciplines and following a well structured plan provided a tremendous lift to the general knowledge. The co-ordination was carried out within the framework of the AMAP Working Groups and Secretariat. Based on the information obtained during AMAP Phase I on geographical patterns, levels of concern and human exposures, and the associated Phase I recommendations, the Arctic countries continued to conduct recommended essential monitoring and added a number of additional new tasks including effect studies. The Arctic Assessment Report (AMAP 1998), the State of the Arctic Environment Report (Nilsson 1997), the Reports to Ministers (e.g. AMAP 2000) and administrative work by the AMAP Secretariat strongly supported the continued funding for these activities and the consecutive efforts to negotiate agreements and protocols for mitigation of Arctic (and global) environmental contamination (see Appendices).

Throughout the AMAP Phases I and II, monitoring of contaminant concentrations has been carried out with varying degrees of effort in the different Arctic countries; this has varied over time in response to changing political support and funding. In Greenland, we have gradually focussed the so-called AMAP Core monitoring program to a limited numbers of species with a geographical spread. Temporal trend monitoring was strengthened by introducing programmes for yearly sampling between the initial five-year interval programme (1999–2004) at selected locations; since 2004, the interval was defined at every second year. In addition, effect studies were initiated, investigating East Greenland polar bears (and later also sledge dogs) based on their contaminant levels in relation to documented OHC and Hg levels of concern. As Greenland provided a unique opportunity to obtain samples from the traditional hunt, a gross and histopathological assessment of multiple organ systems was initiated in 1998 (**Paper 21, 24, 27**, Section "Contaminant related pathological effects and diseases" and additional references herein).

Thesis of the dissertation

The following 7 thesis points will be addressed and documented in this dissertation:

(i) Basic parameters such as age and sex of the animal, tissue type, and season of collection, are likely to affect contaminant concentrations in biota.

(ii) Ecosystem structure, differences in trophic level, biomagnification characteristics, and climatic differences will have an affect on contaminant concentrations in biota.

(iii) Geographical patterns can be detected in contaminant concentrations within Greenlandic and other Arctic marine mammal populations, reflecting regional loading of the systems.

(iv) Temporal trends in contaminant levels can be detected for key species in the Greenland ecosystem, reflecting global trends in emissions and pathways.

(v) The most highly exposed groups in the Arctic system, i.e. top predators, may be affected by contaminants, however even well defined and examined epidemiologic disease outbreaks such as PDV can be hard to link to contaminant levels due to confounding parameters.

(vi) The Inuit population can reduce their contaminant intake by following food recommendations and thereby reduce their risk of being affected by contaminants.

(vii) Studies of marine mammal population structure are highly relevant to both the conduct of contaminant monitoring and to the interpretation of information on contaminant patterns and trends in the Arctic.

Structure of the dissertation

This dissertation consists of a summary, a general introduction, a results and discussion section and conclusions, including perspectives for the future contaminant work. The 30 papers that form the basis of this thesis address three main topics.

a) Marine contaminant loads
b) Contaminants effects and diseases
c) Marine mammal distribution and stock separations

The first part deals with the contaminant loads of marine mammals and some of their prey, and their dynamics relative to age, sex, trophic levels, geography and development over time. The second part deals with the potential effects of contaminants, and the third part deals with a number of supporting disciplines including animal migrations and stock separations discussed in relation to contaminant issues. A simplified overview, tying the three parts together with subsections and topics consideres beyond the scope of the dissertation is given in Fig. 3. This dissertation aims to interrelate these three topics and provides examples of how they support each other in terms of knowledge, but also through collaboration between scientists, institutions and different funding agencies and programmes. Additional articles reporting emerging evidence primarily from areas adjacent to Greenland (i.e. Canada and Svalbard) has been used in the present dissertation to set our results in perspective rather than to provide an exhaustive review. Information on the physical and chemical characteristics, the sources of pollution, pathways and how contaminants enters the food chain were considered to be beyond the scope of this thesis.

Many of these issues, such as age, trends and/or bioaccumulation are the subject of many papers. However, the primary articles do not necessarily deal the bioaccumulation issue or with human exposure, as such complex discussions have not been repeated in each of the species or animal group specific articles. The bioaccumulation issue and human exposure is typically dealt with in overview articles or the AMAP assessments by compiling data from many food items from one or more regions. Overview articles may hence leave less room for explanatory and normalising variables (e.g. **Paper 10, 12**, Johansen et al. 2004a, b).

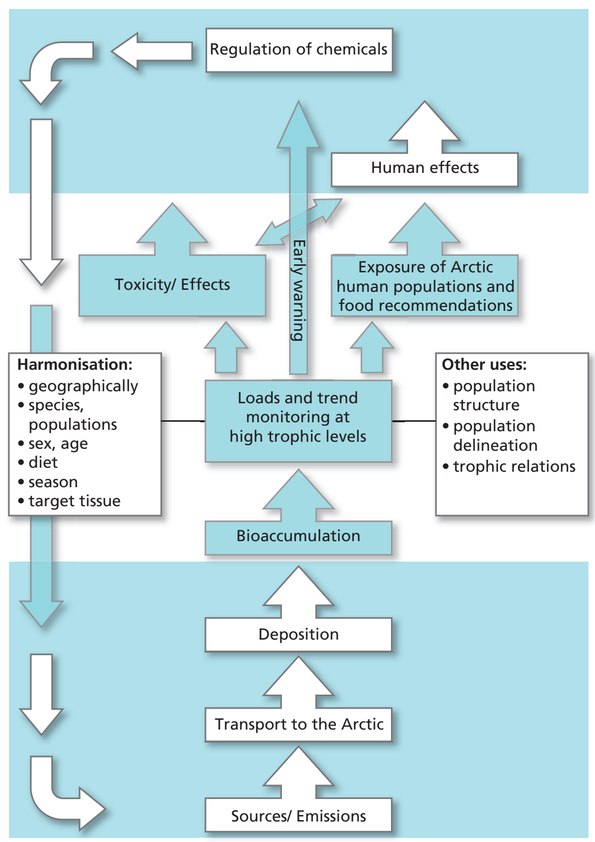

Fig. 3. Overview on the main topics and their mutual interactions dealt with in the present dissertation. Topics in the blue-shaded boxes (bottom and top) are outside the scope of the dissertation.


Photo: R. Dietz

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Results and discussion

Importance of the marine ecosystem
As the majority of mainland Greenland is ice-covered and all settlements are situated along the coast, the marine environment has always provided the most important sources of food in Greenland (e.g. Kapel & Petersen 1982, Huntington et al. 1998, Pars et al. 2001, Deutch 2003). When considering the contribution to human dietary exposure to contaminants, foods derived from marine species are almost the only group of importance, whereas terrestrial food is almost negligible (Astrup et al. 2000, Deutch 2003, Johansen et al. 2004a, b). As marine mammals contribute significantly to the Greenlandic diet, and as it was recognised early-on that contaminants were bio-accumulating and bio-magnifying in these species, marine mammals received the highest priority in our investigations (see "Marine contaminant loads"). However, our work has also included investigations studying crustaceans, fish and seabirds to verify the conclusions regarding these trophic differences and to contribute to the documentation of bio-accumulation in the marine environment. Information on the physical and chemical characteristics, the sources of pollution, pathways and toxicological characteristics have not been included here because they are considered to be beyond the scope of this thesis. Further information on these issues concerning the Arctic can be found in e.g. Dietz et al. (1998; **Paper 12**), de March et al. (1998), de Wit et al. (2004) and Marcy et al. (2005).

Marine contaminant loads

Importance of key parameters for normalization and exposure
When describing contaminant loads there are a number of considerations and factors that need to be taken into account. One of the important questions to answer is how basic biological parameters (e.g. age/length, sex, season or feeding habits/stable isotopes) are linked to contaminant levels. Such information is important for normalizing data prior to conducting geographical and temporal trend analysis, for evaluating bioaccumulation rates and for determining particularly exposed groups and seasons. All of this information is also important as a basis for providing advice in relation human consumption so that contaminant intakes can be minimized. Other parameters such as the chemical form of the contaminant – its speciation – (e.g. organic versus inorganic Hg) and congener composition are important to understand in relation to intake, partitioning between tissues and organs, and toxicity. Finally, understanding the relationship between contaminants and key parameters linked to climate change including temperature, precipitation and transport processes are also important in explaining and predicting changes in contaminant exposure in future scenarios. As stated in the Canadian Phase II National Assessment report: *"It is strongly suggested that statistical analysis and interpretations of all metal and OC data in biota incorporate relevant biological data. New monitoring programs should include a suite of biological measurements such as stable isotopes, age, sex, etc."* (Fisk et al. 2003). Only little attention has been payed to investigating how representative the subsamples from various tissue compartments are for the animals (e.g. Nielsen & Dietz 1990).

Age and sex related differences

Ways of dealing with age and sex differences
In order to study the relationship between contaminant levels and age it is important to have a reliable method of age determination. Most recent comparisons are carried out taking into account differences in age accumulation rates among regions or periods, using age normalised means either calculated by selecting only specific age groups or calculated to represent a particular age based on the relationship for a broad span of ages. Where a sex difference can be detected, such comparisons are also often conducted for a specific gender. In some programs both gender are analysed, in other programs the gender best represented in the samples are chosen. When relating contaminants to effects, the sex and age groups that are most exposed or vulnerable may be chosen. To detect gender differences a considerable sample size is often needed. Therefore, some authors do not separate their data into different gender groups and thus less information is available on this parameter. In most in-

vestigations, age and sex patterns are investigated or dealt with to achieve thorough descriptions and reliable comparisons. Parameters such as age and sex are also important when studying effects of contaminants, in addition to a wide range of other variables such as population dynamics and dispersal.

Age determination

Age may seem to be a trivial parameter, but given the extensive international QA (Quality Assurance) programs that are devoted to contaminant analyses, relatively little attention has been given to the importance of age as a covariate and the QA of age determination methods. There is general agreement on which techniques are appropriate to use on species like seals and polar bears, where decalcification followed by thin sectioning, staining and readings of the cementum growth layer groups is the most commonly used technique (e.g. **Paper 5, 6, 8, 13**, Grue & Jensen 1979, Calvert & Ramsay 1998). In walrus, ages are obtained from reading growth layer groups (GLGs) in the cementum of thin sections (not decalcified) of molariform teeth (e.g. Born et al. 1981). Estimates of age in beluga whale are generally obtained from reading GLGs in the dentine using polarized light (Heide-Jørgensen et al. 1994, Stewart 1994). Recently, measurements of radiocarbon 14C using the signature from atomic bomb tests in the 1950s and 1960s has been used to verify that one GLG is laid down annually in beluga teeth (Stewart et al. 2006). Narwhals have caused more problems as neither layers in the lower jaw, dentinal nor cementum layers in the embedded tusk have been ideal (e.g. **Paper 5, 19**). Recently, aspartic acid racemization of the eye lenses has been suggested as a way to solve the problem of age determination in narwhals (Garde et al. 2007). Age determination in baleen whales has also been problematic, as no way to verity readings from captive animals with known age history is possible. Use of body length as a proxy for age is a common practise for whales with age determination problems, and as a time and resource saving alternative at lower trophic level species. Ear plugs and aspartic acid racemization of the eye lenses seems to be the best alternatives to length measurements in baleen whales (e.g. Aguilar & Borrell 1994, George et al. 1999). For some birds, plumage is used for a rough age categorization (**Paper 3**). In fish, otoliths layers can be counted, and in molluscs seasonal growth lines are visible in the shell (e.g. Brousseau 1979, Härkönen 1986).

Mercury

Invertebrates and fish

Mercury concentrations are higher in large than in small decapods (**Paper 10, 12**). Mercury concentrations in soft tissue of blue mussels were positively correlated with shell length (Riget et al. 1996, 2000). However, Atwell et al. (1998) could not detect an accumulation with age in clams (*Mya truncata*) from Lancaster Sound, despite an age range of 42 years. Riget et al. (**Paper 11**) did a thorough comparison of a larger number of fish species in relation to fish length and Hg concentrations. Of 27 comparisons among muscle, liver and kidney, 24 showed a positive relationship and only 3 showed a negative trend with size. Of the positive trends, 8 were significant, which was not the case for any of the negative trends. Stange et al. (1996) reported a positive Hg-size relationship for fish from the North Atlantic.

Birds

In general, birds are not suitable for a detailed evaluation of age and sex relationships as absolute ages are difficult to obtain and most species are therefore normally only grouped into yearlings and adults. Nielsen & Dietz (**Paper 3**) investigated five seabird species from Greenland. A difference between age groups was only detected in one species, probably due to low number of samples and questionable age determination. A three-fold higher Hg concentration was found in 2.2+ year-groups compared to 1-year groups of black guillemots (**Paper 3**). No accumulation with age was found in livers of glaucous (*Larus hyperboreus*) and Iceland (*Larus glaucoides*) gulls from Greenland in the AMAP Phase I data (Riget et al. 2000). Levels in older kittiwakes (*Rissa tridactyla*), Brünnich's guillemots (*Uria lomvia*), and black guillemots (*Cepphus grylle*) from Lancaster Sound, Canada, and black guillemots from West Greenland compiled in the first AMAP assessment, were generally higher in older birds compared to younger (**Paper 12**).

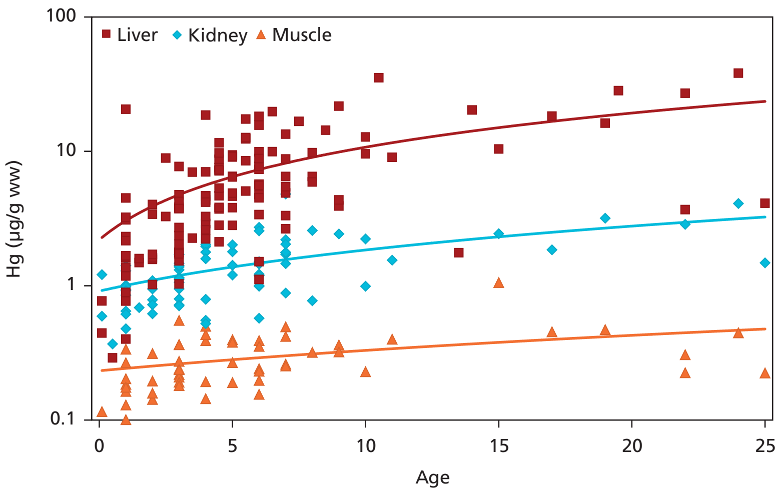

Fig. 4. Mercury concentration (µg/g ww) versus age in ringed seal (n = 87) tissues from Ittoqqortoormiit (modified from **Paper 13**). Lines represents exponential curves.

higher concentrations of Hg (and Se) in liver compared to males (**Paper 19**). Paludan-Müller et al. (1993) likewise found that in harbour porpoises (*Phocoena phocoena*) from Greenland waters, Hg increased with age until 4 years of age in muscle, skin (and kidney), whereas Hg appeared to increase in liver throughout the entire lifetime. Julshamm et al. (1987) found that the increase in Hg concentration levelled-off in both muscle and liver with increasing size of pilot whales (*Globicephala melas*), as also documented for narwhals and harbour porpoises. Similarly, increases in Hg concentration with age have been documented for belugas, narwhals, harbour porpoises, white-beaked dolphins (*Lagenorhynchus albirostris*) and pilot whales from Canadian waters (Gaskin et al. 1972, 1979, Wagemann et al. 1983, Muir et al. 1988, Wagemann et al. 1990, 1996).

Seals

There is a general consensus that Hg increases with age in marine mammals. An age-related accumulation of Hg was documented in Greenland ringed seals (Fig. 4; **Paper 13**). Hg showed a continuing accumulation throughout life, with 2.9-, 6.9- and 3.0-fold increase in muscle, liver and kidney, respectively, for >15 year old seals compared to 1-year-old seals. Similar relationships have been documented for seals in the Canadian Arctic (e.g. Smith & Armstrong 1975, 1978, Wagemann et al. 1996). Mercury rarely differed between genders in the seals (**Paper 13**).

Whales

Hansen et al. (**Paper 5**) found that Hg concentration was positively correlated with age or body length in muscle, liver and kidney of narwhals, with liver and kidney in belugas, and with liver in minke whales. Few differences in Hg concentrations between the two sexes were detected in the examined tissues of the three species (**Paper 5**). In a later study, Dietz et al. (**Paper 19**) investigated a larger sample of narwhals from Northwest Greenland and found that Hg (and Se) concentrations in muscle, liver and kidney increased in the first 3–4 years (measured a GLGs) of life, after which no further dependence on age was observed. Females had significantly

Polar bear

In polar bears, an increase of muscle, liver and kidney Hg with age has been documented (Norstrom et al. 1986, Braune et al. 1991, **Paper 8, 16**).

Cadmium

Invertebrates and fish

Cadmium concentrations in crustaceans increase with length/weight (age). In the amphipod *Parathemisto libellula* and the deep sea prawn (*Pandalus borealus*) the Cd concentrations were approximately twice as high in large compared to small animals (**Paper 12**). Cadmium increased with size/age in blue mussels from Greenland (Riget et al. 1996). However, there was no solid evidence that the concentration of Cd increases with size of fish. Of 28 liver and kidney (muscle often below detection limits) comparisons among several Greenland fish species, half showed a positive trend and the other half showed a negative trend with size (**Paper 11**). Of the positive trends, only one was significant, and none of the negative trends were significant. Bohn & Fallis (1978) found a tendency of increasing Cd concentrations with size for shorthorn sculpin from Canadian waters. Hellou et al. (1992) found Cd concentrations in Atlantic cod (*Gadus morhua*) from the northwest Atlantic to be negatively correlated with size.

Birds

For some Greenland bird species, Cd concentrations increased with age (**Paper 3**). This was particularly pronounced in common eider (*Somateria molissima*) and king eider (*Somateria spectabilis*) muscle, liver and kidney, where 6 of 9 comparisons (3 tissues, two species and two regions for common eider) were significantly increasing. Tendencies were found in Iceland gull, Brünnich's guillemot and black guillemot as well, of which only the relationship for muscle tissue in one population of black guillemot was significant (**Paper 3**). In the Greenland 1994 AMAP results, Riget et al. (1997, 2000a) found that Cd concentrations in liver of glaucous gull and Iceland gull clearly increased with age. Furness & Hutton (1979) analysed ringed Great skua (*Catharacta skua*) with exact ages from Scotland and found significant correlation between bird age and Cd in the kidney. However, Norheim (1987) found no differences between juvenile and adult birds from Svalbard, including glaucous gull, Brünnich's guillemot, little auk (*Alle alle*) and common eider.

Seals

Cadmium is virtually absent from the mammalian body at birth and is not transported transplacentally (WHO 1992a, 1992b, **Paper 12, 13**). Increase of Cd concentration with age in seals has been documented by several authors (e.g. Sergeant & Armstrong 1973, Wagemann et al. 1996). Dietz et al. (**Paper 13**) also found a highly significant correlation of Cd with age for muscle, liver and kidney until 5–10 years of age, with 2.7-, 2.3- and 2-fold increases relative to seals of 1 year of age (Fig. 5). While muscle concentrations continued to increase in seals >15 years of age (3.0-fold higher than 1 year old seals), liver and kidney decreased to 53 % and 73 % of concentrations of the 5–10 year-old seals (**Paper 13**). Cadmium has seldom been found to vary with the sex of seals (**Paper 13**).

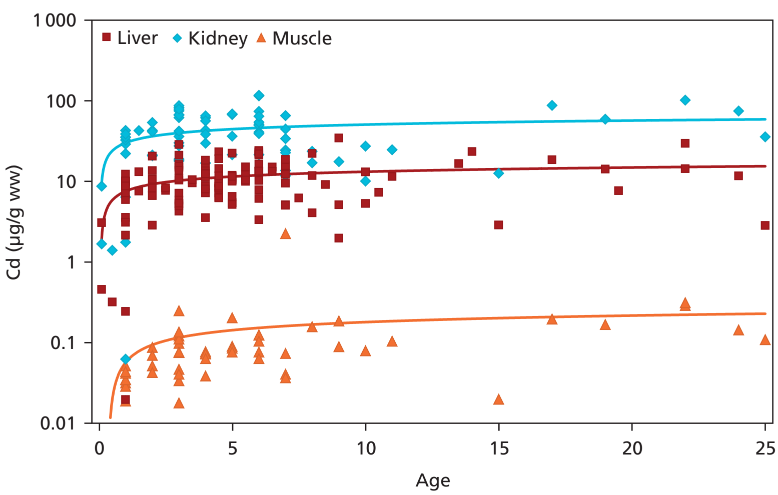

Fig. 5. Cadmium concentration (µg/g ww) versus age in ringed seal (n = 87) tissues from Ittoqqortoormiit (modified **from Paper 13**). Lines represents exponential curves.

Whales

In a Greenland study of minke whale, beluga and narwhal, 4 of 12 comparisons showed a significant increase of Cd with age (**Paper 5**). If length was used instead of age for narwhals, an additional 2 comparisons became significant. However, among the oldest investigated age groups of belugas and narwhals, Cd showed a decrease in all the comparisons among muscle, liver and kidney. Paludan-Müller et al. (1993) found that Cd increased until four years of age in Greenland harbour porpoises. In animals older than four years, muscle and liver concentrations reached a constant level, whereas kidney levels decreased. Wagemann et al. (1983, 1990, 1996) and Muir et al. (1988) noted an increase in Cd concentrations with age in whales (beluga, narwhal, Pilot whale and White-beaked dolphin) from Canadian waters.

Polar bear
An increase of Cd concentrations in polar bear tissues with age has been documented by several authors, while no decrease in older bears has been reported (Norstrom et al. 1986, Braune et al. 1991, **Paper 8, 16**).

Part conclusion on sex differences and age related accumulation of Hg and Cd
Older animals tend to have higher concentrations of Hg and Cd than younger animals in the Greenland marine ecosystem. In some cases the increase levels off in older animals and for Cd in liver and kidney a decrease may be seen in older animals. Differences among sexes are seldom recorded. For human consumption, preferences for young animals will result in lower Cd and Hg intake.

Effect of age and sex on OHC levels in marine biota

Fish
In general, little information is available on age accumulation of OHCs in fish and the information is somewhat contradictory. Muir et al. (2000) stated that length and age were not significant covariates in a study of Arctic char (*Salvelinus alpinus*) from two locations in Labrador and three locations in Nunavik. However, Fisk et al. (2003) used the size of turbots (*Scophtalmus maximus*) as an explanation for the 5- to 10-fold differences in PCB and DDT concentrations from two studies in the Davis Strait conducted in 1992, 1997 and 1999 (Berg et al. 1997, Fisk et al. 2002). Riget et al. (2004) also used the length to adjust concentrations in Greenland sculpin time trend comparisons to avoid bias in the comparisons.

Seabirds
OHCs have rarely been documented to vary significantly with age in seabirds (Fisk et al. 2003, Riget et al. 2004, Braune et al. 2005a, b). Also, there exists no consistent information on differences between sexes of seabirds (Olafsdóttir et al. 1998, Buckman et al. 2004). Several seabird studies in addition use eggs to eliminate age and sex as covariables.

Seals
The general pattern for most OHCs is that levels are higher in adults compared to juvenile seals and higher in males than in females. The lower levels in females are believed to be attributable to lipid transfer during gestation and lactation. The extent to which females accumulate OHC may depend on contaminant exposure, as high exposure may affect how often they successfully produce and wean offspring and therefore how much is eliminated through lipid transfer (de Wit et al. 2004). A tendency for concentrations to increase with age was observed in ringed seals, but was not statistically significant. Differences in concentrations between females and males were only significant for HCB and HCH within certain age classes and sampling areas (Cleemann et al. 2000b). Concentrations of ΣPCB, ΣDDT and ΣCHL were found to increase with age in both male and female ringed seals of the Nunavut region (Fisk et al. 2002). OHC concentrations were higher in male than female ringed seals, and the rate of age accumulation differed between the two genders (Weis & Muir 1997, Fisk et al. 2002). Such differences were not found for ΣCBz and ΣHCHs in seals. A similar variability in age related trends with different contaminants, species and sex was also concluded by Muir et al. (1999a).

Whales
OHC concentrations in blubber of narwhals were dependent on age and sex (**Paper 19**). Females showed decreasing OC concentration in the first 8–10 years of age, while males increased during their first few years of life, after which the concentrations became stable (**Paper 19**). Although not explicitly stated, it is assumed that Fisk et al. (2003) found an accumulation of most OHCs with age in beluga from Pangnirtung in the eastern Canadian Arctic, since ΣHCH, ΣDDT, Σtoxaphene ΣPCB, endosulfan, 1,2,3,4-chlorobenzenes, HCB, ΣCBz, dieldrin and a large number of congeners were all age-adjusted for temporal trend comparisons.

Polar bears
In East Greenland, adult male polar bears had higher levels of ΣPCBs, ΣCBzs, ΣDDTs, Mirex and dieldrin and lower levels of ΣCHLs compared to adult females and subadults (Fig. 6; **Paper 20**). However, only concentrations of

ΣCBzs were significantly higher in adult males than in the two other groups. In temporal trend analyses, Dietz et al. (**Paper 20**) separated the comparisons in the case of ΣCHLs and Dieldrin due to significant differences among sexes and age groups (subadult, adult males and adult females). Norstrom et al. (1998) concluded that concentrations of ΣPCB were significantly higher in male than female polar bears from the Canadian Arctic. On the other hand, chlordanes in male Canadian polar bears were 30% lower than in females (Norstrom et al. 1998), which can be explained by the male ability to metabolize chlordanes during seasonal fasts (Polischuk et al. 2002). The corresponding differences in Greenland were 10% higher ΣPCB and 23% lower ΣCHL in males compared to females (**Paper 20**). However, a substantial sex difference in the relative concentrations of ΣCHL and the other OHCs are observed over the entire year in the Greenland bears (**Paper 20**). Therefore, differences in seasonal patterns and sampling seasons among regions may also be an explanation for the observed differences. Concentrations in females were lower than in males for most of the year, but in April for ΣPCB, in March-July for ΣHCH, and in March for ΣCHL and Dieldrin the levels in females were higher than in males (**Paper 20**). The most probable explanation for the lower concentrations in females are the transfer of OC compounds transplacentally to the foetus, and transfer in milk to the cubs during the weaning period, which may last up to two years (e.g. Polischuk et al. 1995, Bernhoft et al. 1997, Norstrom et al. 1998, Polischuk et al. 2002, Skaare et al. 2002).

Part conclusion on sex differences and age related accumulation of OHCs

Although not consistent among all species, OHC groups or studies, older males tend to have higher concentrations of OHCs than females and young in the Greenland marine mammals. In mammals, fat soluble contaminants can be transferred to offspring through gestation and lactation, giving mature females ways to excrete these compounds and thereby reducing their body burden. Consumption of older males may therefore result in higher OHC intake.

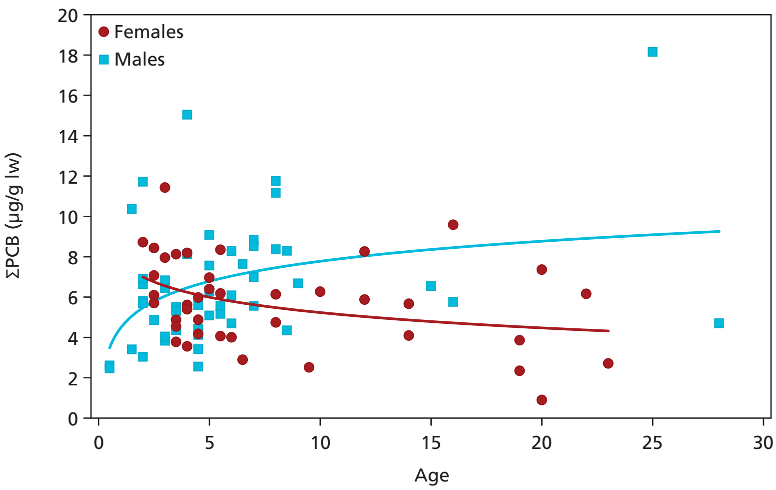

Fig. 6. ΣPCB concentration (ng/g lw) versus age in polar bear (n = 92) adipose from Ittoqqortoormiit (data from **Paper 20**). Lines represents exponential curves.

Normalization for seasonal changes in contaminant loads

The seasonal variation in animal distribution and food intake are likely to cause differences in contaminant loads. These differences may give rise to different exposures, but such differences may also introduce confounding factors when conducting temporal and geographical comparisons if samples are not obtained in the same season. Information on seasonal variability is seldom available to correct for such differences. Fasting periods and migration patterns are the major explanation for seasonal differences. In one study, fasted harp seal (*Phoca groenlandica*) blood OC levels showed a significant time-dependant increase, even though no differences were determined in the blubber (Lydersen et al. 2002). In the second part of this study, OHC concentrations in blood and blubber from seals collected in prime condition before the breeding season were compared with animals collected in poor condition during moulting. Blood and blubber levels of most

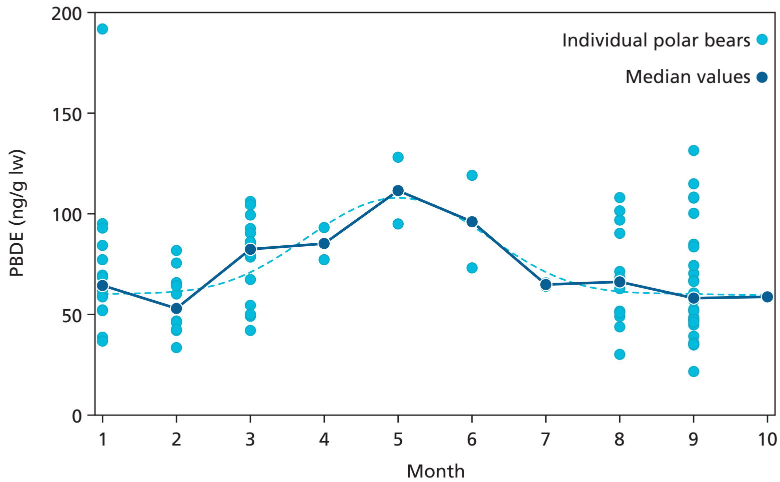

Fig. 7. ΣPBDE concentration (lw) versus month, calculated on lipid weight (top) basis in adipose tissue from East Greenland polar bears during 1999–2001. The data were not normalised by sex or age, as no significant relations were found. Light blue circles represent individual polar bears. Dark blue circles represent median values of the month connected by a solid line. Broken lines represent the trigonometric regression curve fit (**Paper 28**).

OHCs were significantly higher in the thin seals compared with the levels found in the fat seals. Dietz et al. (**Paper 20, 28**) likewise found a substantial seasonal variation in the various OHCs analyzed among different age and sex groups of East Greenland polar bears. For PBDE it was suggested that geographical data comparisons should be corrected for seasonal variability (Fig. 7; **Paper 28**). This could be relevant for several other combinations of OHCs, age/sex groups and other species as well (**Paper 20**).

Part conclusion on seasonal differences
Limited information is available on seasonal changes in contaminant levels. Geographical data comparisons and temporal trend monitoring may be affected by seasonal variability in contaminant loads. Such variability can be reduced by collecting specimens at the same time of the year or by correcting the data for such seasonal variability.

Normalization for differences in food

Stable isotope analysis of tissues provides a tool for evaluating trophic position and food source and is increasingly being included in monitoring programs to facilitate the interpretation of contaminant levels (Hobson & Welch 1992, Hobson 1999, Braune et al. 2002, Muir et al. 1995, **Paper 19**, Hobson et al. 2004, Hobson 2005, Riget et al. 2007b). The use of stable isotopes has expanded over the last decade but only recently have suggestions been made on how this information can be used to normalize data. Riget et al. (2007b) used $\delta^{15}N$ to normalize Hg concentrations in Greenland ringed seals for differences in food intake. Including tissue $\delta^{15}N$ values as a covariate in some cases had a dramatic effect on the results. In ringed seal from Central West Greenland the annual changes in $\delta^{15}N$-adjusted Hg was estimated to 5.0% between 1994 and 2004 and 2.2% between 1999 and 2004 compared to 1.3% and 12.4%, respectively, for the non-adjusted Hg. One should be cautious when using stable isotopes for normalisation of time series, as a time signal from food-related changes resulting from, e.g. climatic change, could be hidden by the normalisation procedure.

Part conclusion on food normalization
Stable isotopes can be used to explain differences in contaminant loads. Stable isotopes may also be used to normalise data for time trend analysis, although such normalisation may mask real changes due to, e.g., climate change.

Effect of climate change

The Arctic faces threats from climate change that will inevitably have an effect on contaminants loads in the Arctic ecosystem (Macdonald 2005). Macdonald et al. (2003) reviewed the available literature on contaminants and climate change. This review concluded that the routes and mechanisms by which heavy metals and OHCs are delivered to the Arctic are strongly influenced by climate variability and global climate change. These pathways involve a number of factors, such as temperature, precipitation, winds, ocean currents, and snow and ice cover in complex interactive systems. Studies have indicated the potential for substantial changes in atmospheric and oceanographic pathways that carry contaminants to, within, and from the Arctic. Pathways within food webs, growth processes and the effects on biota may also be modified by changes in climate. These effects mean that climate-related variability in recent decades may be responsible, in part at least for some of the trends observed in contaminant levels. Macdonald (2005) concluded that climate change induced changes in contaminants loads will mainly pose a risk to top Arctic predators as these species are most exposed to contaminants, and are most likely to become stressed by other parameters related to climate change. One of best documented example of climate stress is on the Hudson Bay polar bear population, which are deprived of their ability to hunt seals during spring due to changes in the presence of ice in spring and autumn (e.g. Stirling 2002). The burning of stored fat through metabolism results in release of archived fat-soluble contaminants and, potentially, an increase of contaminant burden in the remaining fat reservoir (e.g. Lydersen et al. 2002). Longer periods of starvation due to change in ice conditions or change in prey populations could lead to higher doses of OCs sequestered in fat – usually at a time when the animal can least afford it. The overall effect of changes in polar bear feeding, from their stable diet of ringed seal to species at other trophic levels, where these are available, will probably vary by region and remains fairly speculative. Faster growth of the lower food chain organisms may reduce their burdens of heavy metals, as indicated by lower heavy metal concentrations in biota in warmer Arctic waters around Svalbard (see section on geographic trends). Svalbard is strongly influenced by the relatively warm Gulf Stream, leading to faster growth of the lower food chain organisms, which ultimately results in lower body burdens of metals. Species such as polar cod, ringed seal and polar bear are all lower in Cd in this region (**Paper 12, 13, 16**). Slow growing poikilothermic organisms accumulate metals over a longer period of time before entering the food chain (Muir et al. 1996, **Paper 12**). However, increases in precipitation and melting of the permafrost may result in more heavy metals being released from soils and carried to the marine environment by rivers, which would increase the amount of these contaminants available for uptake by biota (Macdonald et al. 2003).

Part conclusion on season, feeding patterns and changing climate

The dynamics behind the pathways and accumulation of contaminants are complex and may be driven by many processes. Beside age related processes, seasonal differences, differences in trophic level of food and differences associated with climatic variability and change are important information that if possible should be taken into account in geographical and temporal trend comparisons.

Geographical trends

The primary reason for conducting geographical trend analyses on biota is to understand the sources and pathways of contaminants. Also such information can be used to identify the areas where highest human exposure is likely to occur as well as areas where effects on biota and man may first occur. Effects studies in areas with low exposure may still be important, however, serving as a reference for comparisons with areas of high exposure. Samples from biota can be compared with the trend data on human exposure. These patterns should partly follow the same trends, but differences in hunting and feeding traditions and habits may lead to different patterns as human diets differ among cultures and over time. Therefore temporal trend in humans are not as reliable for anthropogenic development as the Arctic biota.

Heavy metals

Mercury

The AMAP assessment in 1998 clearly documented that Hg levels were higher in the central Canadian Arctic compared to other Arctic regions. This was shown for at number of species and tissues, of which polar bear liver (e.g. Lentfer & Galster 1987, Norstrom et al.

1986, Braune et al. 1991, **Paper 8, 12, 16**) and hair (Eaton & Farant 1982, Renzoni & Norstrom 1990, Born et al. 1991, **Paper 12**) showed the clearest pattern (Fig. 8).

The same trend was documented in other species including ringed seal and beluga whales (review in **Paper 12**). In the Phase II AMAP assessment, similar geographical trends were confirmed based on Hg analysis in ringed seals normalised to 5-year means from 18 areas in the late 1990s (Ford et al. 2005). However, high sub-regional variability was also detected. Based on many of the same data, Riget et al. (2005) verified this trend with the highest concentrations around 120° W longitude for both subadult (0–5 years) and adults (>6 years) ringed seals. Such geographical trends could not be documented in seawater, algae, invertebrates and fish, while birds tended to carry higher Hg concentrations at higher latitudes (**Paper 12**, Ford et al. 2005, Marcy et al. 2005). Among the causes that have been proposed to explain the geographical variations is the sedimentary geology across the Canadian Arctic (Wagemann et al. 1996, Muir et al. 1999a). In Greenland, no obvious linkage between sediment concentrations and concentrations in marine biota was found (**Paper 10**). The correlation between the local geological structures and levels in marine biota should indicate that natural sources were the primary cause of the geographical trends in Arctic. Such a close relationship does not agree well with the time trends indicating that anthropogenic sources are the most important contribution to post-industrial increases in Hg in the Arctic (e.g. Outridge et al. 1997, 2000, 2002, 2005, **Paper 25**; and section on temporal trends). In Greenland, no clear geographical pattern of Hg was found within the entire ecosystem (**Paper 10, 12**). Young ringed seals showed higher concentrations in Northwest and East Greenland compared to seals from Southwest Greenland (**Paper 12, 13**). Such a gradient could not be detected in ringed seals from the Canadian Arctic (Ford et al. 2005). Neither could an increasing south-to-north trend in Hg be detected for lower trophic level species (**Paper 10, 12**, Ford et al. 2005). Feeding behaviour is, however, likely to be an important factor influencing the spatial patterns, as suggested by several authors (e.g. Muir et al. 1995, **Paper 12**).

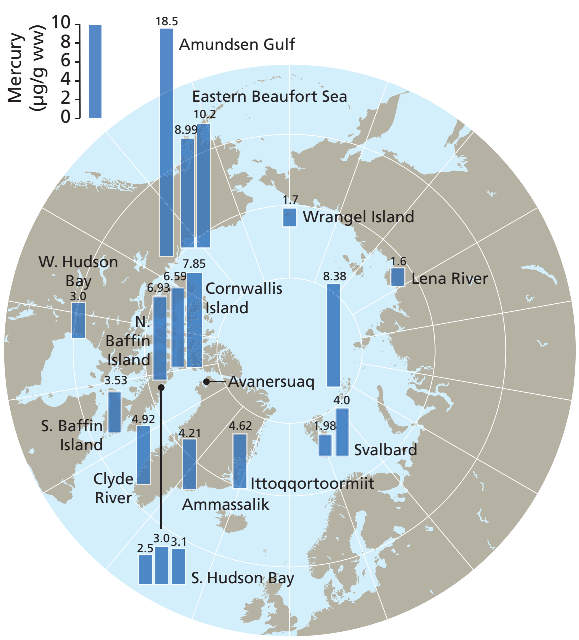

Fig. 8. Mercury levels are higher in biota from Canadian areas than in other Arctic regions. Here exemplified by Hg in polar bear hair (µg/g dw.) (Sources: Eaton & Farant 1982, Renzoni & Norstrom 1990, Born et al. 1991, **Paper 12**).

Cadmium

Concentrations of Cd in marine mammal tissues of ringed seal, beluga and polar bear increase from West to East in the Canadian High Arctic (Norstrom et al. 1986, Braune et al. 1991, Wagemann et al. 1996, **Paper 12**). The same trend could be extended to include West Greenland, for ringed seals and polar bears, but not for belugas (**Paper 12**). Recent investigations on ringed seals from the Phase II of AMAP have confirmed this pattern (Ford et al. 2005, Riget et al. 2005). Wagemann et al. (1996) also explained the geographical Cd trend within Canada in terms of geological differences between the western and the eastern Canadian Arctic. In Greenland, no significant differences in the Cd levels of bottom sediment were found for the different geological structures (Loring & Asmund 1996). On the other hand, Cd levels are generally highest in ringed seals and polar bears from Northwest Greenland compared to areas further south (Fig. 9, **Paper 13, 16**). Up to five-fold difference in Cd concentrations occur across the Arctic and depend on the areas,

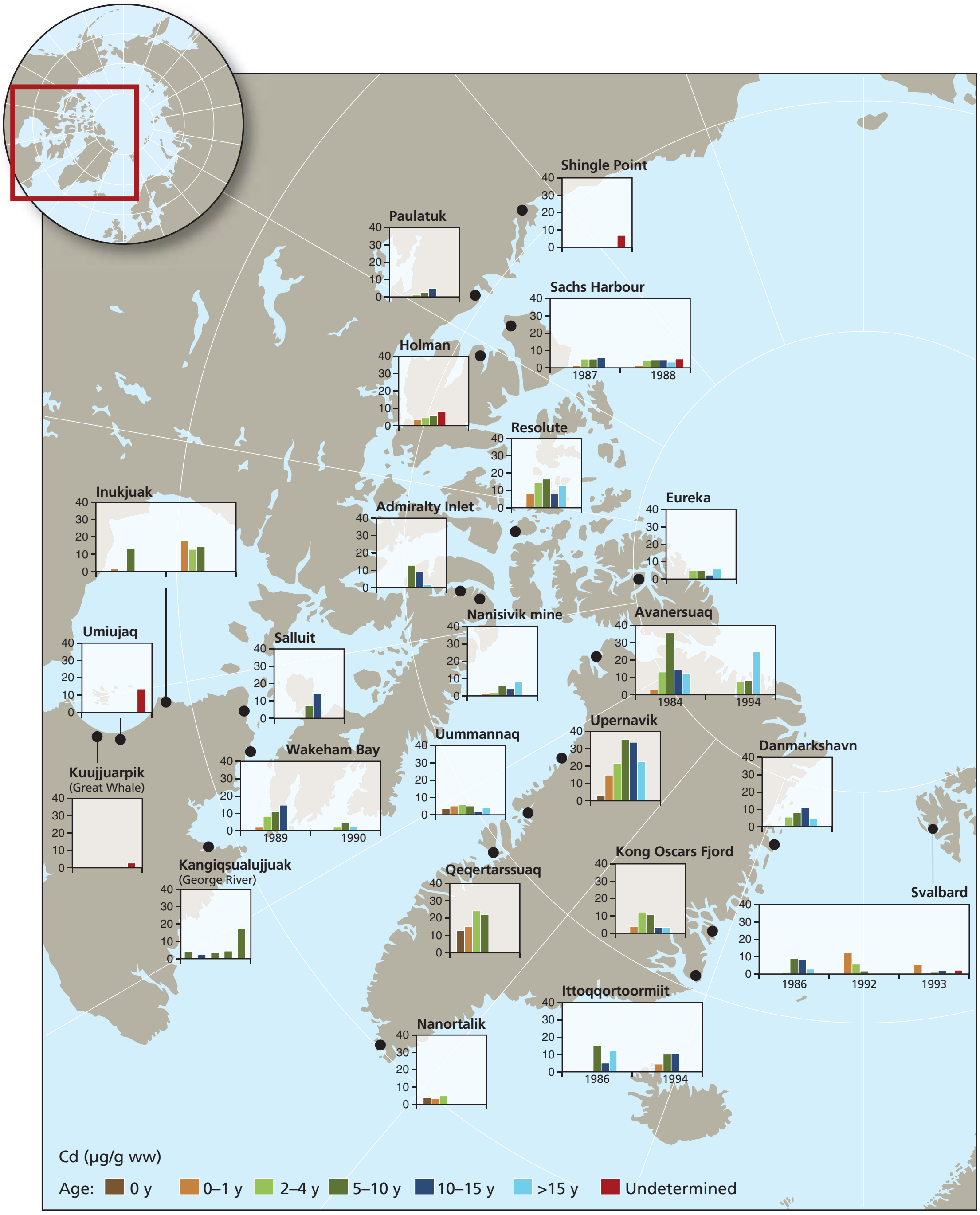

Fig. 9. Geographical trend in Cd levels (µg/g ww) of ringed seal livers showing the highest concentrations in Northwest Greenland and the lowest concentrations in Western Canada (**Paper 12**).

species and tissues examined. The Cd levels in ringed seals and polar bears in Central East Greenland are somewhat lower than in Northwest Greenland (Avanersuaq), and even lower at Svalbard. On the east cost of Greenland (Ittoqqortoormiit, Danmarkshavn and Kong Oscars Fjord), only minor differences can be detected in ringed seal Cd concentrations between areas (**Paper 13**). No discernible north-south trend is observed in Cd concentration of ringed seals as for the Hg (**Paper 13**).

Cadmium concentrations in ringed seals from the Arctic are significantly higher than those reported from the Gulf of Finland and the Gulf of Bothnia (Helle 1981, Perttilä 1986, Frank et al. 1992, **Paper 12**). Specifically, concentrations are approximately 15-fold higher

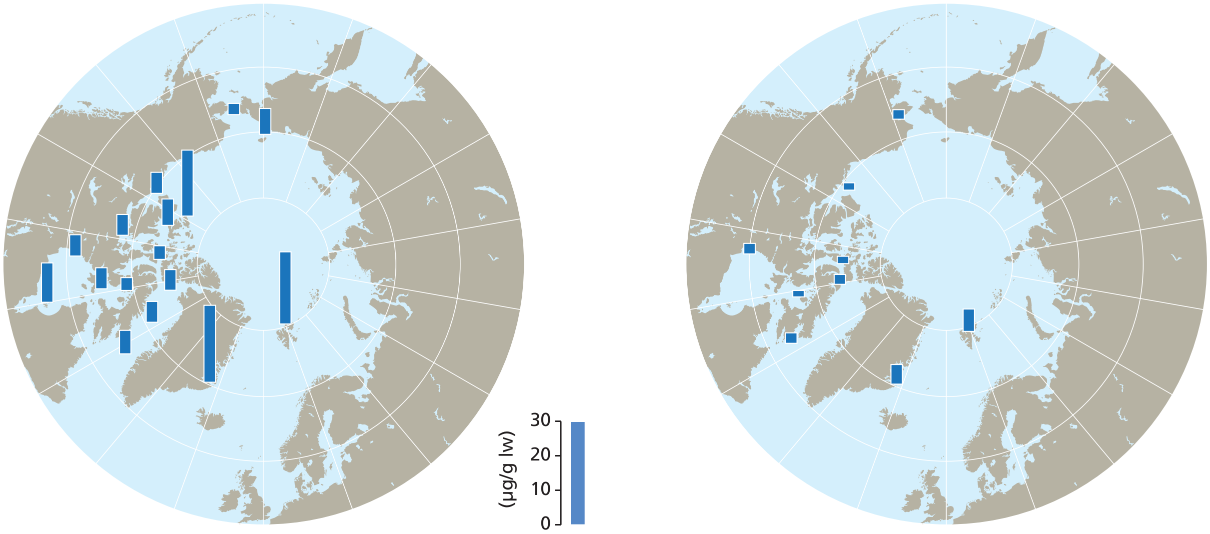

Fig. 10. Geographical trend in ΣPCB (19 congeners) levels (µg/g lw), adjusted to expected levels in 11-year-old male polar bear adipose tissue during the period around 1990 (left) and 2000 (right) (Modified from Norstrom et al. 1998, de March et al. 1998, Verreault et al. 2005).

in muscle, 16- to 75-fold higher in liver, and 24- to 42-fold higher in kidney (**Paper 12**). Johansen et al. (1980) likewise concluded that Cd levels were highest in Arctic seals. Concentrations of Cd in harbour porpoises from Greenland waters carried 10-fold higher Cd levels than did those from European waters (Paludan-Müller et al. 1993). These differences may be partly explained by differences in available food items. Species such as the pelagic amphipod *Parathemisto libellula*, and other crustaceans, as well as arctic cod (*Arctogadus glacialis*) may be important Cd sources in the Arctic (**Paper 12**). The higher levels in Arctic marine mammals may also be a consequence of slower growth rates in the Arctic (see Effect of climate change section).

OHCs

A clear geographical trend is seen for several OHCs in different Arctic species. One of the first and best documented patterns was shown for polar bear adipose tissue collected from different management zones in 1990 and analysed by the same laboratory (see e.g. ΣPCB in Fig. 10; Norstrom et al. 1998, de March et al. 1998). A comparable investigation was repeated ten years later, where the same geographical pattern was found, although at considerably lower concentrations due to decreases in ΣPCB (Fig. 10; Verreault et al. 2005).

ΣHCH were higher in Canadian and Alaskan polar bears compared to bears from East Greenland and Svalbard (Norstrom et al. 1998), which was also the case in a recent comparison carried out by Verreault et al. (2005), where ΣHCH concentrations in Alaskan bears were significantly and six-fold higher than age-adjusted mean ΣHCH levels in Svalbard bears. This pattern was taken as an indication of an ongoing contribution of HCHs from China, southeastern Asia, and North America (de March et al. 1998).

A similar pattern was detected in ringed seal, which is the major food source of polar bears and an important food resource for the Inuit population in Canada and Greenland. Ringed seals were lowest in ΣPCB and ΣDDT in Alaska, intermediate in Northern Arctic Canada and Western Greenland and highest in southern Hudson Bay, East Greenland, Svalbard and the Yenisey Gulf (Luckas et al. 1990, Daelemans et al. 1993, Schantz et al. 1993, Skaare 1996, Cleemann et al. 2000c, Krahn et al. 1997, Nakata et al. 1998, de March et al. 1998, Muir et al. 1999a, 2000, Fisk et al. 2002, de Wit et al. 2004). In contrast, ΣHCH levels were higher in Canadian and Alaskan ringed seals compared to seals from the European Arctic, in agreement with previous compilations of circumpolar data for ringed seals (Muir et al. 2000). Toxaphene showed a different pattern, with differences among congeners. Hence, Toxaphene Parlar 26 was highest in ringed seals from Hudson Strait and Ungava Bay, followed by seals from Alaska, the White Sea, Svalbard and West Greenland. Levels of Parlar 50 were highest in seals from Svalbard and Hudson Strait, and lower in seals from Ungava Bay and western Greenland (Wolkers et al. 1998, Muir et al. 1999b, 2003, Hoekstra 2002 compiled in de Wit et al. 2004). In the conclusion of the Phase II AMAP report on OHCs, de Wit et al. (2004) concluded that the same general patterns as documented above for polar bears and ringed seals are true for all marine mammals and birds. The geographic pattern found for the "new" OHCs (PBDEs and PFCs), indicates highest levels in the European Arctic, in several cases with the highest concentrations being found in East Greenland (de Wit et al. 2004, Smithwick et al. 2005b, Muir et al. 2006).

Photo 3. Geographical trends of the Greenland wildlife and Inuit population have shown a high exposure to a large number of contaminants. Photo: R. Dietz.

Part conclusion on geographical trends

A clear geographical trend can be detected within the Arctic. Northwest Greenland and the central Canadian Arctic have the highest concentrations of Hg, Central West Greenland and Northwest Greenland have the highest concentrations of Cd, while East Greenland (together with Svalbard and Kara Sea) has the highest loads of lipophilic OHCs. This information is very important for the effects-related disciplines, in indicating where to look for possible effects of contaminants due to high levels and where humans are highest in exposure.

Temporal trends

Knowledge of temporal trends in contaminant levels in Greenlandic biota has increased considerably over recent years. Riget et al. (2004, 2007a) and Riget (2006) reviewed and updated the Hg, Cd, and OHCs time series in soft tissues of Greenlandic marine, terrestrial and freshwater species that are considered "essential" under the AMAP monitoring program. In addition, extended time series of Hg in Greenland polar bear hair and feathers of birds of prey have been added to twenty year old West Greenland investigations of human hair and seabird feathers (Appelquist 1985, Hansen et al. 1989, **Paper 26, 29**). Recently data from peregrine falcon (*Falco peregrinus*) eggs, ringed seals blubber and polar bears of selected OHCs have appeared from the Greenland area (Sørensen et al. 2004, Vorkamp et al. 2005, **Paper 22, 23**, Rigét et al. 2006, Dietz et al. 2008).

Mercury

Most of our work has focused on examining soft tissue, to provide information relevant for food chain and human dietary intake studies. However, recently the challenge of extending time series further back than was possible using archived frozen samples led to investigations looking into hard tissue sample collections. As the temporal trend pattern of Hg is different for East and West Greenland, these two areas are considered separately.

Mercury in East Greenland polar bear hair before 1973

Due to the sample composition and the fact that the time trends in East Greenland changed around 1973, this year was used to split the time series in polar bear hair. A 3.1% per year increase in East Greenland was found for the period between 1892 and 1973 (**Paper 25**; Fig. 11). In addition, it was possible to analyse samples from Northwest Greenland from 1300 AD, these showing the lowest Hg concentrations (0.518 µg Hg/g dw). This concentration

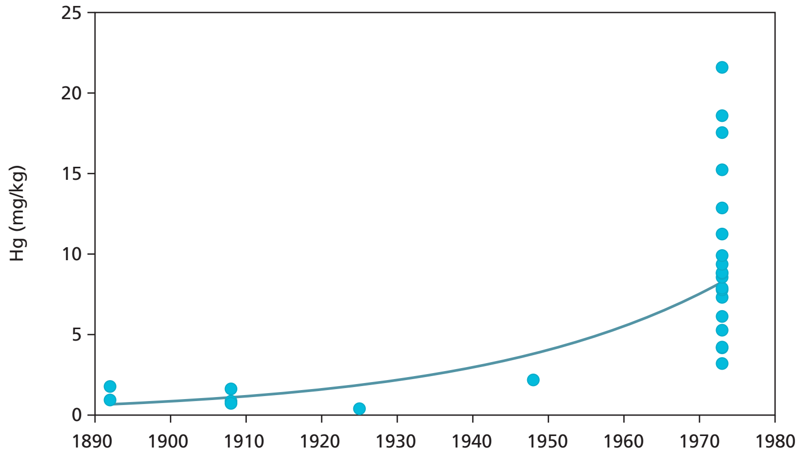

Fig. 11. Mercury in East Greenland polar bear hair (n=27) between 1892 and 1973 showing a significant ($p<0.0001$) increasing trend of ca. 3.1%/year (**Paper 25**).

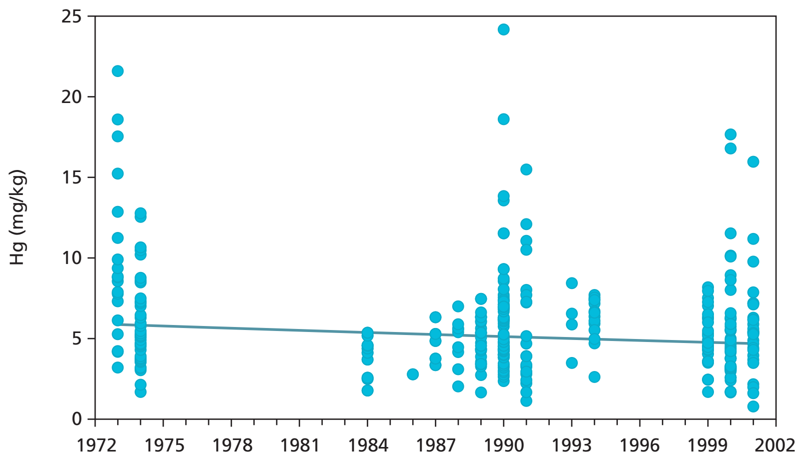

Fig. 12. Mercury in East Greenland polar bear hair (n=322) collected between 1973 and 2001 showing a significant ($p=0.009$) decreasing trend of 0.8%/year (**Paper 25**).

was regarded as a baseline value for the period before human activities increased emissions and releases of this metal world wide. The 10-year means from the Greenland East Coast after 1965 were 7.4- to 13.9-fold higher than the baseline data from 1300 AD (**Paper 25**). The highest mean level was detected in the East Greenland sample from the period 1965–1974, but as no data were available from 1950 to 1965, the peak may have appeared earlier than this. The Hg concentration in bear hair from the period 1965–1974 in East Greenland was almost 14-fold higher than the baseline data from 1300 AD. These results are in the same order of magnitude as those from studies in Norway on human deciduous (milk, primary) teeth, which showed a 13-fold increase in levels from the 12th Century to the 1970s (Eide et al. 1993). A clear increase of Hg has likewise been found from 1835 to 1969 in the fifth primary feather sampled from common guillemot (*Uria aalge*) and Brünnich's guillemot (*Uria lomvia*) from the Baltic and Kattegat areas, whereas levels were lower and the trend less pronounced in samples from the Faroe Island and Greenland for the same period (Appelquist 1985). Marine sediments from East Greenland and post industrial peat core profiles from the Faroe Islands and Norway all showed a clear enrichment in Hg relative to pre-industrial samples (Asmund & Nielsen 2000, Shotyk et al. 2003).

Mercury in East Greenland polar bear hair after 1973

A time series based on samples of hair from 322 East Greenland polar bears showed a significant 0.8% decrease between 1973 and 2001 (Fig. 12; **Paper 25**).

Recent Hg time trends east of Greenland

These results are in agreement with investigations of human deciduous teeth from Norway, which likewise suggest that Hg concentrations have declined substantially during the past 20 years (Eide et al. 1993, Tvinnereim et al. 1997). Also Hg peaked in feathers of Swedish and Norwegian birds of prey around 1966, followed by a decline. However, these species are believed to be affected from earlier Hg treatment of seed dressings, as well as chlorine-alkali, paper and pulp industries around the Baltic (Westermark et al. 1975, Odsjö 1975, Johnels et al. 1979, Lindberg & Odsjö 1983, Appelquist 1985). Recent time-series for Atlantic

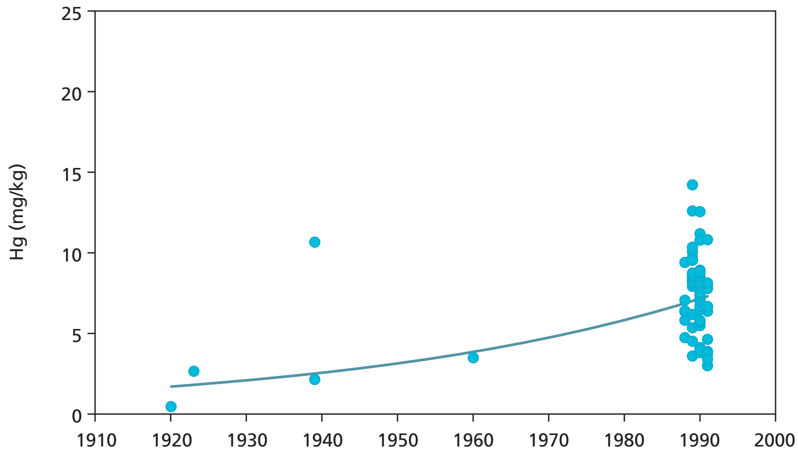

Fig. 13. Mercury in Northwest Greenland polar bear hair (n = 67) collected between 1920 and 1991 showing a significant ($p < 0.0001$) increasing trend of. 2.1% / year (**Paper 25**).

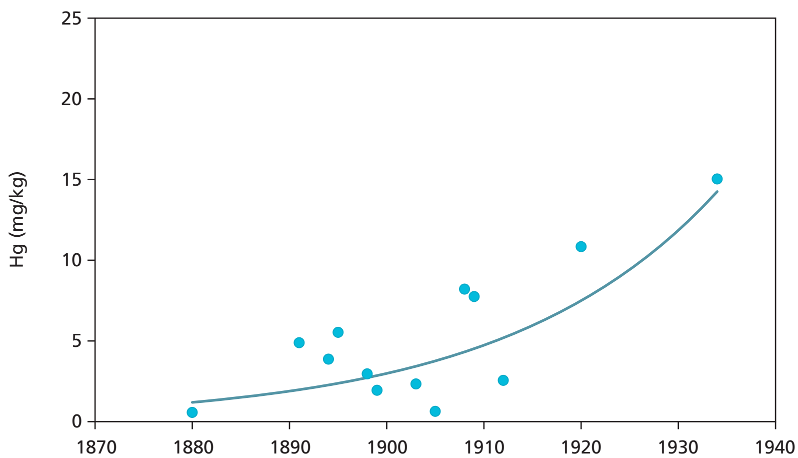

Fig. 14. Mercury concentrations in the fifth primary (n = 13) of West Greenland immature gyrfalcons collected between 1880 and 1935 showing a significant ($p < 0.020$) increasing trend of 4.5% / year (**Paper 29**).

cod (*Gadus morhua*) and dab (*Limanda limanda*) from Iceland and blue mussels (*Mytilus edulis*) from northern Norway likewise showed a significant decreasing trend (ICES 2002). The dated peat bogs from Greenland in the period from 1945 to 1995 showed a 6.2% decrease per year (Shotyk et al. 2003), and Boutron et al. (1998) reported a Hg decline of approximately 4.5% per year in Greenland ice cap from the late-1950s to the late-1980s.

Hg trends in West Greenland

In Northwest Greenland, polar bear hair samples showed a significant positive 2.1% per year increase in Hg concentrations for the period 1920 to 1991 (Fig. 13; **Paper 25**). Hair samples covering a gap for period after 1991 have been collected in spring 2006, and the Hg results will soon be available. The mean Hg concentration in bear hair from the period 1985–1994 in Northwest Greenland was 14.4-fold higher than the baseline data from 1300 AD from the same region. These results indicate that ca 93% of present day Hg in polar bears is a result of mercury from anthropogenic sources.

Hg in West Greenland birds of prey

In another investigation, Hg in primaries of West Greenland gyrfalcons, peregrine falcons and white-tailed sea eagles covering the period 1850–2004 were documented (**Paper 29**). Seven out of 8 comparisons (3 species and 2–3 age groups) were increasing, of which 4 were significant (e.g. Fig. 14). The linear regressions on unbroken time series from the period 1880 to 1935 showed increases in the range of 1.1–4.5% per year, and for the period 1880 to 1960 the increase was between 0.4–0.9% per year (**Paper 29**).

Similarly, a study of Hg in American (*Falco peregrinus anatum*) and Arctic (*F. p. tundrius*) peregrine falcon eggs from Alaska found a recent increase of Hg (Ambrose et al. 2000).

Other long term Hg investigations west of Greenland

The low 1300 A.D. Hg baseline level was supported by Canadian polar bear hair samples from the same period and recent Canadian bear hair analysis indicate an increase in the same order of magnitude as that found for the Northwest Greenland polar bears (Wheatley & Wheatley 1988). The hair of modern-day West Greenlanders also contains significantly more Hg than found in samples from 15th Century Inuit mummies, but here the difference was only 2.5-fold (Hansen et al. 1989). Similarly, Wheatley and Wheatley (1988) reported that modern Hg levels in human hair from the Canadian Arctic were several times higher than in pre-industrial samples. The teeth of Beaufort Sea beluga from Mackenzie Delta from 1993 contained significantly (4.1- to 17 fold) higher concentrations of Hg than found in archaeological samples dated to the period 1450 1650 AD (Outridge et al. 1997, 2000, 2002). A comparison of samples collected around Somerset Island during 1894–1998 showed increases of between 4.1- and 7.7-fold, indicating that a substantial part of the Hg increase has taken place during the second half of the last century (Outridge et al. 2005).

Recent Hg trend in soft tissue

Only a few time series for Hg in soft tissue covering the last 20 years exist with a significant number of sampling years. Recent changes in diet and in the foods consumed by the Inuit population are of major importance to the present and future health scenario for these groups of humans.

Hg time trends in polar bear muscle, liver and kidney were analysed for subadult (2–6 years old) and adult (≥6 years old) groups of polar bears from central East Greenland by Riget et al. (2004). In contrast to the results for polar bear discussed above, no general increases or decreases in Hg concentrations in these tissues were apparent from the time series that included up to 8 sampling years. However, rather few samples (range 1–10) were analysed from each year. On the other hand, time series for three age groups of ringed seals in Northwest Greenland, all showed a significantly ($p < 0.01$) increasing trend of Hg (and decrease in Cd) from 1984 to 1998 (Riget et al. 2004). Whether the changes reflect anthropogenic input, changes in seal feeding behaviour or other environmental factors is unknown. As both crustaceans and fish are important food items for ringed seals, the opposite trends in Hg and Cd could indicate a ringed seal feeding change from crustacean (high in Cd and low in Hg) oriented food towards fish (high in Hg and low in Cd) (Siegstad et al. 1998, **Paper 10, 12**). This theory was supported by increases in $\delta^{15}N$ levels (Riget 2006, Riget et al. 2007a). The ringed seals ≤ 4 years from Northwest Greenland were reanalyzed together with stable isotope data from all the seals and including an additional year (2004). This resulted in a non-significant ($p = 0.210$) annual increasing trend in Hg of 7.8% per year (Riget et al. 2007a). As the Hg concentration was found to be significantly positively correlated with $\delta^{15}N$, the Hg concentrations were normalized to a common $\delta^{15}N$ of 16.4‰ assuming a common slope for all years. The temporal trend estimate for the $\delta^{15}N$ normalized Hg revealed an annual increase of 8.5%, which was higher than the rate estimated using the non-adjusted concentrations; although the regression relationship improved, this trend was still non-significant ($p = 0.165$). No time trend could be detected in ringed seals from Central East Greenland, although some year-to-year differences were detected (Riget et al. 2004).

Riget (2006) has recently updated and compiled older time series. Twenty-one time-series from 3 regions in Greenland were examined and provided 14 examples of positive trends of which only 2 were significant (Riget 2006). The lack of significances is probably an effect of too few years of data (with series typically covering only 3–7 years). Of 7 time series from Avanersuaq, Northwest Greenland, 6 showed an increasing trend, but none were significant (Riget 2006). Fewer species (7 of 21) showed decreases, but none of these were significant. Among these was walrus, from Avanersuaq. Walrus have a preference for bivalves, and even though blue mussel is not among the food items of walruses, it is interesting to note that all 4 blue mussel size classes from the more southern Qeqertarsuaq showed decreases (Riget 2006).

Results on ringed seals from Canada do not provide a clear picture. Of 8 regions sampled between 1972 and 2001, increases, decreases and fluctuating trends were detected (Braune at al. 2005a, b). The most complete data set was available from Holman Island, N.W.T., but even here no clear pattern could be detected. Recent data from Fisk et al. (2003) for 1972, 1974 and 1977, from the original work of Smith & Armstrong (1978) as well as from Wagemann et al. (1996) and Muir et al. (2002) was compiled. Age-adjusted Hg concentrations in the seals varied markedly over the 30-year period, but not in any consistent temporal pattern. Significantly higher concentrations were found in 1974 and 1977 compared to 1993 and 1996, while levels in 2001 were also higher than in 1993 (Fisk et al. 2003).

Eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*), and black-legged kittiwakes (*Rissa tridactyla*) were collected from Prince Leopold Island in Lancaster Sound, Canada, between 1975 and 1998. Total Hg concentrations almost doubled between 1975 and 1998 in eggs of thick-billed murres, while the increase in northern fulmars was ca 50 % (Braune et al. 2001). Recent data from 2003 indicate a continuation of this trend (Braune et al. 2005a, b). Stable isotope analyses ($\delta^{15}N$) indicated that the temporal trends observed were not a result of shifts in trophic level feeding behaviour. Eleven of 14 sediment samples from West Greenland showed a clear increase all throughout the profiles, representing approximately the last 100 years (Asmund & Nielsen 2000). Likewise the Hg content in seven sediment cores from the Arctic Ocean in 1994 showed an increase towards the surface in the upper 10 cm of the sediment (Gobeil et al. 1999).

Trends in Cd

Few long time series in biota have been produced for Cd. Less focus has been given to Cd, as most of the Cd that occurs in air is associated with particulate matter and therefore the dispersal is often less than 30 km from the source (**Paper 12**). Of 19 marine Cd time series from the East and West Greenland regions, 15 showed a negative trend (Riget 2006). However, only 1 of these was highly significant, namely Cd in livers of older ringed seals from Avanersuaq. Of the 4 positive trends, none were significant (Riget 2006). As previously mentioned, the opposite trends in Hg and Cd in ringed seals in Avanersuaq could partly be explained by changes in feeding patterns from crustaceans towards more fish-dominated feeding (**Paper 10**, Riget & Dietz 2000, Riget 2006, Riget et al. 2007a, section "Recent Hg trend in soft tissue"). The Cd time trends have previously been discussed by Riget & Dietz (2000) and Riget et al. (2004) on a smaller data set, and are therefore not cited in further detail.

Part conclusion for time trend of heavy metals

Investigations of biota hard tissue and other media that allow study of long-term changes have revealed long-term increases of Hg with a substantial anthropogenic contribution. East of Greenland, this increase levelled-off somewhere around the 1960s–1970s and now shows a significant decline. In West Greenland, Hg has increased throughout the 20th century, but information from recent decades is sparse. Some West Greenland time series, as well as series from the Central Canadian Arctic indicate a continuing increase of Hg. Recent increases in Hg accompanied by decreases in Cd in ringed seals are most likely attributable to shifts in diet. Most of the Greenland soft tissue time series currently still include too few years of sampling to provide a clear picture of time trends and year-to-year variation.

OHCs

Due to different histories regarding the introduction, use, regulation, and bans of various OHCs, and differences in transport pathways, different temporal trends can be found for different OHCs, and for various species and regions of the Artic. The accumulated information is dealt with separately for the different OHC groups below.

PCB

ΣPCB (defined here as the sum of CB congeners 99, 149, 118, 146, 153, 138, 183, 180, 170/190 and 194) was compared in polar bears from Ittoqqortoormiit from 1990 and 10 years later (1999–2001). A significant difference was detected with ΣPCB levels for the period around 2000 being 78% lower than those found in 1990, equivalent to a yearly decrease of 14.0% (Fig. 16; **Paper 20**). Riget (2006) recently updated the ΣPCB (10 congeners) time trend data of Riget & Dietz (2000) and Riget et al. (2004). Of 8 marine time series including ringed seals, shorthorn sculpin, glaucous gull and black guillemot eggs from 3 regions, 7 showed decreasing trends of between 0.2% and 8.2% per year, based on 2 to 8 years of data. However, only the comparisons of younger ringed seals from Qeqertarsuaq (–8.2%) and Ittoqqortoormiit (–4.4%) were significant (Riget 2006). Arctic char (*Salvelinus alpinus*), the only freshwater species studied also showed a significant decrease in ΣPCB of 11.6% per year (Riget 2006). The trends in polychlorinated biphenyl (PCB) congeners 28, 31, 52, 101, 105, 118, 138, 153, 156, and 180 determined in blubber of young (≤4 years old) ringed seals from central East Greenland collected in 1986, 1994 and during the period 2000 to 2004 were recently published and compared with PBDEs (Riget et al. 2006). ΣPCB decreased significantly over the period from 1986 to 2004 with an estimated annual rate of decrease of 4.3% per year (Fig. 15). In a comparison of walruses from 1978 and 1988, no significant differences could be detected for ΣPCB (ca. 100 congeners) over this 10 year time period (Muir et al. 2000). Sørensen et al. (2004) analysed 22 PCB congeners (CBs 28, 31, 44, 49, 52, 99, 101, 105, 110, 118, 128, 138, 149, 151, 153, 156, 170, 180, 187, 188, 194, 209) in 37 eggs from peregrine falcons (*Falco peregrinus*) from Southwest Greenland from the period 1986 to 2003. Although the majority of the PCB congeners showed decreasing trends, none of the regressions were significant.

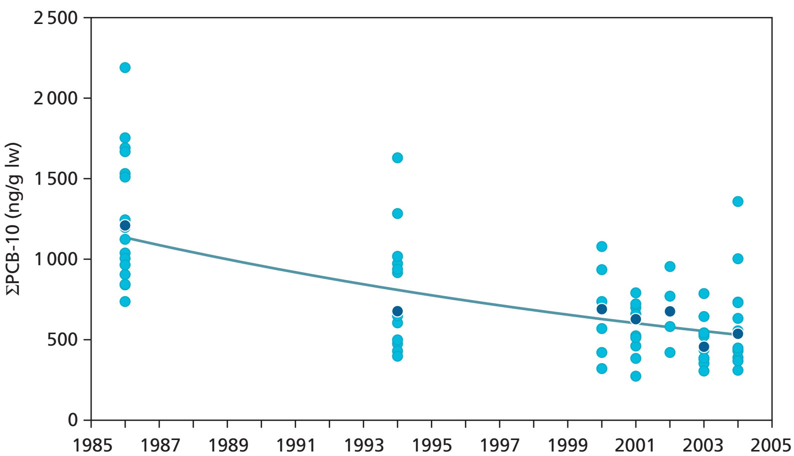

Fig. 15. Temporal trend in ΣPCB-10 concentration in blubber of young (≤4 years) ringed seals from Ittoqqortoormiit, East Greenland between 1986–2004. The dark blue circles represent the median concentrations. The solid line represents an exponential curve determined by log-linear regression analysis (Riget et al. 2006).

In Canadian polar bears, ΣPCB decreased fairly steadily throughout the 1990s, with a biological half-life of approximately 18 years (Fisk et al. 2003). This half-life was considerable longer than that found for the East Greenland polar bears, where the half-life was only 4.6 years. A large variation was found in the half-life for individual CB congeners in the Canadian bears. The half-life of CB-153 in the Canadian study was 19 years, similar to that of ΣPCBs, whilst the half-life for CB–180 was shorter (13 years) and CB-99 was longer (>50 years) (Fisk et al. 2003). The corresponding values for the East Greenland samples showed less variability and suggested much shorter half-lives (4.5 to 5.7 years; **Paper 20**) indicating a much faster reduction in the contaminant loads in East Greenland polar bears. ΣPCB concentrations in Greenland polar bears showed a reduction of 77.9% during the 10 year period from 1990 to 1999–2001, whereas less than a 50% reduction in ΣPCB levels was observed in Hudson Bay over a three times longer period (1968–1999). Temporal trends of OCs have also been studied in polar bears from Svalbard. Henriksen et al. (2001) studied the trend of CB-153 concentrations in polar bear blood annually between 1990 and 1998. Decreases of ca. 40% occurred in the early 1990s, and concentrations stabilised thereafter. De Wit et al. (2004) have estimated the annual percentage decline of PCB concentrations in polar bears to be 2.7% for Hudson Bay polar bears and 6.1% for Svalbard bears, for the period 1989–1999. These estimates are also lower than observed for the East Greenland bears, where the yearly decrease of 14% resulted in an 80% decline over the same 10 year period. The PCB levels in Svalbard and East Greenland polar bears were significantly higher than in bears from Hudson Bay. The differences in concentration levels and rate of decline is believed to be due to the proximity of Greenland and Svalbard to European sources, and air mass movements bringing higher loads of OCs to Greenland and Svalbard compared to Hudson Bay. Hence, PCB levels at Svalbard and in East Greenland may have reached an equilibrium state with globally distributed PCBs later than in Hudson Bay, due to the continuing influence and proximity of the above mentioned sources. Prior to the 1990s, the picture of temporal trends was less obvious at Svalbard. Differences in the OHC levels measured between 1967 and 1993–1994 ranged from a decrease (CB-187) to unchanged concentrations in both sexes (CBs 105, 118 and 209), to an increase in females (CBs 99 and 128), to increases in both sexes (CBs 138, 153, 156, 157, 170, 180, 194 and 206) (Derocher et al. 2003). The maximum change observed was a nine-fold increase in concentrations of CB-157 in adult females. Changes from 1967 to 1993–1994 in contaminant patterns were explained by Derocher et al. (2003) as a combination of selective metabolism and accumulation of organochlorines in polar bears and temporal changes in the contaminant mixture being transported to the Arctic.

Photo 4. Ringed seal has a circumpolar distribution and is an important food item for the Greenland Inuit population. This species has been selected as an essential AMAP monitoring organism and therefore provide information on geographic and temporal trends. Photo: R. Dietz.

Temporal trends of PCBs, in ringed seals in the Canadian Arctic have been studied in three communities from the early 1970s to the late 1990s or 2000/2001. Braune at al. (2005b) presented a brief overview of the more detailed new information presented in Addison et al. (2005) and Muir & Kwan (2003) together with the earlier studies at these sites (Addison et al. 1986, Muir et al. 1988, Weis & Muir 1997, Addison & Smith 1998, Letcher et al. 1998, Addison et al. 2000, Wiberg et al. 2000, Muir et al. 2001). ΣPCB in seals has declined significantly at all three sites over the past three decades: at Ikpiarjuk by a factor of 2.4, at Ausuittuq by a factor of 1.5, and at Holman by a factor of 5.5, based on arithmetic means. At Holman, the bulk of the decline occurred between 1972 and 1981, with no further decline between 1981 and 1991, but another significant decline between 1991 and 2001. PCBs were phased out in North America and northern Europe from about the mid-1970s onwards. Further details on temporal trends of PCBs in the Arctic ecosystems including species such as narwhals and belugas are available from de Wit et al. (2004).

Some of the best time series in Canada are those based on seabird eggs. Concentrations of ΣPCB decreased significantly in eggs of thick-billed murre, northern fulmars and black-legged kittiwake monitored from Prince Leopold Island between 1975 and 2003 (Braune et al. 2005a,b). The significant declines in concentrations of ΣPCB have also been observed in seabirds from other areas including the Baltic Sea (e.g. Olsson & Reutergårdh 1986, Andersson et al. 1988, Bignert et al. 1995) and the Barents Sea (Barrett et al. 1996).

DDT

ΣDDT (p,p´-DDE, p,p´-DDD and p,p´-DDT) was compared in polar bears from Ittoqqortoormiit from 1990 and 10 year later (1999–2001). A significant difference was detected, with the ΣDDT decreasing by around 2000 to 66.3% (p,p'-DDE declined to 71.1%) of the concentration found in 1990 (Fig. 16; **Paper 20**). This change is equivalent to a yearly decrease of 4.0% per year.

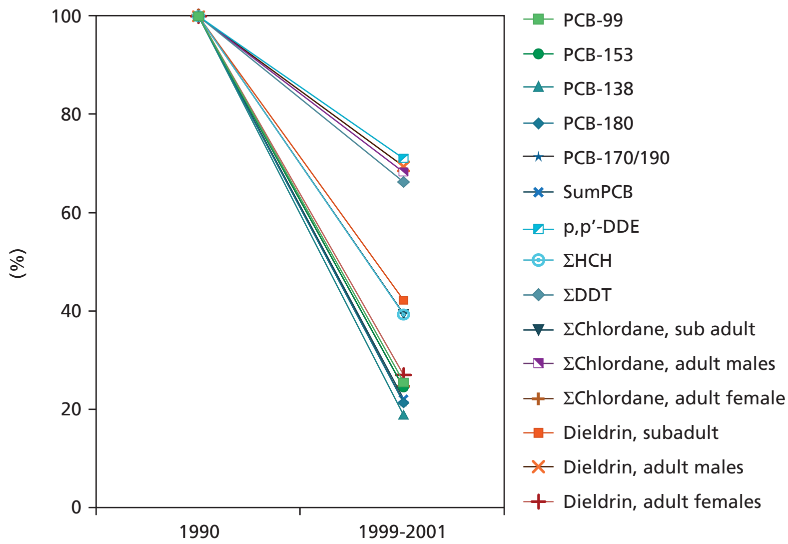

Fig. 16. Percentage of OC left of organochlorine concentrations in polar bears sampled in 1990 (data from Norstrom et al. 1998) and 1999–2001 (data from **Paper 20**) in the Ittoqqortoormiit (East Greenland) area.

Riget (2006) recently updated the Greenland time trend data presented in Riget & Dietz (2000) and Riget et al. (2004). Of 8 marine ΣDDT time series including ringed seals, shorthorn sculpin, glaucous gull and black guillemot eggs from 3 regions, all showed a decreasing trend of between 1.4% and 10.1% per year (2–8 years of data from 1986–2004). However, only the trends in young (≤4 years old) ringed seals from Qeqertarsuaq (–10.1%) and Ittoqqortoormiit (–5.4%) were significant (Riget 2006). Arctic char showed a decrease of 12.5% per year, but this was not significant (Riget 2006). In the Riget et al. (2004) investigation, concentrations of DDTs in seals and sculpin from Ittoqqortoormiit, central East Greenland showed no clear temporal trend from 1994 to 1999/2000 most likely due to the limited number of years of data. Muir et al. (2000) found no significant differences in ΣDDT in walruses from 1978 to 1988. ΣDDT in peregrine falcon eggs was stable or weakly decreasing, but not significantly so, between 1986 and 2003 (Sørensen et al. 2004).

The longest temporal record (1968 to 2002) of the major OHC groups in polar bears in the Arctic concerns the population near Churchill in western Hudson Bay. For adult females there was a generally consistent (and significant) decrease of approximately 80% in ΣDDT over the 34 year study period (Fisk et al. 2003, de Wit et al. 2004, Braune et al. 2005). The strongest decrease of ΣDDT concentrations occurred during the period from 1968 to the 1990s, after which the level remained constant until 2002 (Fisk et al. 2003, de Wit et al. 2004). Local sources, such as the spraying with DDT for insect control in the local communities and at the large military base at Churchill in the 1950s and 1960s, resulted in 2- to 3-fold higher ΣDDT levels in fat of polar bears from Hudson Bay than in bears from other areas of the Canadian Arctic in 1984. This is one of the few examples of significant local sources in the Arctic. After the DDT ban and the closure of the military base, the levels declined in subsequent years. In East Greenland, the decrease of 28.9 to 33.7% in concentrations of p,p'-DDE and ΣDDT from 1990 to 2000 was also statistically significant, even though this decrease was the lowest observed with half-lifes estimated at between 17.1 and 20.6 years (**Paper 20**).

ΣDDT declined significantly in female ringed seals from three investigated areas in the Canadian High Arctic between the early/mid-1970s and the late-1990s/2000. ΣDDT exhibited the largest decline in Canada of any "legacy" OHC ranging from 2.5- to 3.3-fold at Ikpiarjuk and Holman over 25–30 years. Significant increases of the p,p-DDE/ΣDDT ratio were found, reflecting the shift from fresh DDT to degraded older sources (Braune at al. 2005a, b).

As found for ΣPCB, ΣDDT decreased significantly in eggs of thick-billed murre, northern fulmars and black-legged kittiwake sampled from Prince Leopold Island between 1975 and 2003 (Braune et al. 2005a, b). The significant declines in ΣDDT have also been observed in seabirds from the Barents Sea and the Baltic Sea (e.g. Olsson & Reutergårdh 1986, Andersson et al. 1988, Bignert et al. 1995, Barrett et al. 1996).

HCH
The ΣHCH (sum of α-HCH, β-HCH and γ-HCH) decreased significantly, to 39.3% of the 1990 concentration over the 10 year period to 2000 in adipose tissue from East Greenland polar bears (Fig. 16; **Paper 20**). This change is equivalent to a yearly decrease of 9.0% per year, or a half life of 7.4 years.

Riget (2006), updating Riget & Dietz (2000) and Riget et al. (2004), showed a decrease in all 8 marine ΣHCH time series including ringed seals, shorthorn sculpin, glaucous gull and black guillemot eggs. Of the 6 tested trends (two had only 2 year of data), 5 were significantly decreasing, at –8.0% to –14.5% per year during 1986–2004 (Riget 2006). In a time-wise comparison for an earlier period (1978 to 1988) Muir et al. (2000) found a significant increase of ΣHCH in walruses from Avanersuaq. α-HCH and β-HCH decreased by 7.9% and 6.8% per year in peregrine falcon egg between 1986 and 2003, although not significantly (Sørensen et al. 2004).

The downward tendency of ΣHCH concentrations in Hudson Bay polar bears during the 1990s was not significant (Norstrom 2001), but it became significant when data from 1984 and 1989 were included in the analysis. The half-life calculated for α-HCH in polar bears from Hudson Bay during the 1990s was 10 years, which was slightly longer than that calculated for ΣHCH in East Greenland (7.4 years). In the Canadian sample, a decrease in α-HCH, and a consequent increase in β-HCH, over the last 30 years was found. Hence, a significantly higher proportion (50%) of present day ΣHCH in polar bears from Hudson Bay is β-HCH compared to 1984 (25%) and 1968 (17%), whereas the opposite was the case for α-HCH (Fisk et al. 2003 and references therein, de Wit et al. 2004). ΣHCH concentrations showed no significant changes in concentrations in Canadian ringed seals from the 1970s to 2001. However, as for the polar bears, β-HCH as a fraction of ΣHCH increased (de Wit et al. 2004, Braune et al. 2005a, b). ΣHCH concentrations also declined in plasma of polar bears from Svalbard between 1991 and 1999 (Lie & Skaare, unpublished data cited in de Wit et al. 2004). Concentrations were similar between 1991 and 1993, but declined by about 3-fold between 1993 and 1996. Hence, the overall decrease of ΣHCH at Svalbard from 1991 to 1996 is similar to the 2.5-fold decrease observed in East Greenland between 1990 and 1999 to 2001. ΣHCH was the only OHC for which a significant increase in concentrations was seen in the study of Canadian seabirds from Prince Leopold Island from 1975 to 2003, particularly for β-HCH in thick-billed murres and fulmars, but no explanation for this finding was given (Braune et al. 2005b).

HCBs
Riget (2006) showed a decrease in 5 out of 8 marine HCB time series including ringed seals, shorthorn sculpin, glaucous gull and black guillemot eggs with trends between –3.9% and –11.4% over the period between 1986 to 2004 (Riget 2006). Of the 3 negative trends tested, one was close to showing a significant trend and two were significantly decreasing, these being juvenile ringed seals from Qeqertarsuaq (–4.0%) and Ittoqqortoormiit (–3.9%) and shorthorn sculpin from Qeqertarsuaq (–4.7%), respectively. A similar but insignificant decrease of 5.1% per year was seen in peregrine falcon eggs for the period 1986–2003 (Sørensen et al. 2004).

ΣCBz in polar bears from the Hudson Bay appeared to increase between 1968 and 1984, followed by a consistent downward trend after that time. Most of the decline in ΣCBz was due to HCB, which had a half-life in bear adipose tissue of approximately 9 years during the 1990s Braune et al. 2005b). Braune et al. (2005b) also found that HCB concentrations declined in beluga adipose tissue from Southeast Baffin Island between 1982 and 1992, but again HCB became higher in 1996.

Chlordanes
The ΣCHL (Oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor and heptachlor epoxide) concentration in 2000 was between 24.4% and 68.3% of the 1990 level in adult female, subadult and adult male polar bears from Ittoqqortoormiit (Fig. 16; **Paper 20**). These significant decreases were equivalent to a yearly decrease between 13.1% and 3.7%, respectively.

Riget (2006) made the first attempt to evaluate marine ΣCHL time series among ringed seals, shorthorn sculpin and black guillemot eggs from Greenland. Of the 5 trends investigated, 3 showed declines (of between 1.6% and 3.6% per year) of which only juvenile ringed seals monitored between 1986 and 2004 (7 sampling years) were significant. Muir et al. (2000) found no difference in concentration of ΣCHL in walruses from Avanersuaq between 1978 and 1988. *Trans*-chlordane decreased by 5.3% per year and the corresponding figures for oxy-chlordane and *cis*-chlordane were –9.6% and –10.8% per year in peregrine falcon eggs between 1986 and 2003, but none of these were significant (Sørensen et al. 2004).

Riget (2006) made a separate evaluation of the marine *trans*-nonachlor (a major constituent of chlordane) as this component was analysed in more species and years. Of the 8 tested trends, 6 showed yearly declines of –3.4% to –12.5%, of which only the trend in juvenile ringed seals from Ittoqqortoormiit monitored between 1986 and 2004 (8 sampling years) was significant. Muir et al. (2000) found significant increases both for male and female walruses from Avanersuaq between 1978 and 1988. *Trans*-nonachlor decreased by 4.1% and *cis*-nonachlor decreased by 5.6% per year in peregrine falcon eggs between 1986 and 2003, although not significantly (Sørensen et al. 2004).

Information on time trends of chlordanes is scarce in the literature, but Muir & Norstrom (2000) reported a significant increase in ΣCHL concentrations in polar bears from Davis Strait between 1984 and 1989. Oxychlordane, the principal metabolite of *cis*- and *trans*-chlordane, and second only to *trans*-nonachlor as the most abundant chlordane related residue in the southeast Baffin beluga blubber, did not change significantly from 1982 to 1996, but declined by 38% from 1996 to 2002 (Braune et al. 2005b).

Dieldrin

For adult male and female and subadult polar bears from East Greenland, dieldrin levels decreased significantly by between 27.0 ad 69.5% from 1990 to 1999–2001 (Fig. 16; **Paper 20**,). These changes were equivalent to a yearly decrease of 3.6 to 12.2%. Similar, Muir & Norstrom (2000) reported a significant decrease in dieldrin concentrations in polar bears from Barrow Strait and Queen Maud Gulf in the central Canadian archipelago during 1984–1989, but no apparent changes were detected in bears from northern Baffin Bay in the same period. During the 1990s, no temporal trend was detected in bears from the Hudson Bay (de Wit et al. 2004). A 2-fold decline in dieldrin was observed over a 20-year period from 1982 to 2002 in blubber of age-adjusted male beluga from Pangnirtung in the eastern Canadian Arctic (Stern & Ikonomou 2003). Dieldrin decreased significantly in black-legged kittiwake eggs, one of three species monitored from Prince Leopold Island between 1975 and 2003 (Braune et al. 2005a, b).

Toxaphene

Toxaphene was not included in the polar bear OHC survey by Dietz et al. (**Paper 20**). However, Riget (2006) evaluated the marine toxaphene time series in Greenland ringed seals, shorthorn sculpin, glaucous gull and black guillemot eggs. Of the 5 trends tested, only 3 showed declines, of between –4.8% and –8.1% per year, and two increases, of 1.0% and 1.3% per year. Only the juvenile ringed seals from Ittoqqortoormiit monitored between 1986 and 2004 with 7 years of data showed a significant decreasing (8.1% per year) trend (Riget 2006). In walrus from Avanersuaq, NWG, Muir et al. (2000) found only significant increases for adult females between 1978 and 1988. Toxaphene trends varied from 1.0% (CHB-41) to –6.6% (CHB-40) among 6 congeners in peregrine falcon eggs from South Greenland between 1986 and 2003, but none were significant (Sørensen et al. 2004).

Toxaphene is not included in the Canadian seabird time trend study (Braune et al. 2005b). No clear trends were evident in total toxaphene and toxaphene congeners 26 and 50 from 1982 to 1996 in Canadian belugas from Cumberland Sound, but more recent measurements suggest a 40% decline from 1996 to 2002 (Braune et al. 2005b). No trend was detected for toxaphene in Northeast Baffin Island narwhals sampled between 1982–1983 and 1992–1999 (Braune et al. 2005b).

PBDE

No investigations have yet been conducted on time trends in brominated flame retardants of polar bears, although polybrominated diphenyl ethers (PBDEs) have been investigated in polar bears in relation to age, season and geographical trends (Muir et al. 2006, **Paper 28**). Neither did Muir et al. (2000) include PBDEs in the walrus study in Northwest Greenland.

The concentrations of BDE congeners 17, 28, 47, 49, 66, 85, 99, 100, 153, 154, and 183 were determined in blubber of young (≤4 year old) ringed seals from central East Greenland collected in 1986, 1994, 1999 and during the period 2001 to 2004 (Fig. 17; Riget et al. 2006). The levels of ΣPBDE in East Greenland ringed seals are among the highest observed in ringed seal from the Arctic. No significant trends were observed for ΣPBDE or for the congeners BDE 28, 47 and, 99 during the entire period from 1986 to 2004. However, an increase may have taken place prior to 2001 after which the concentrations appear to have started to decline.

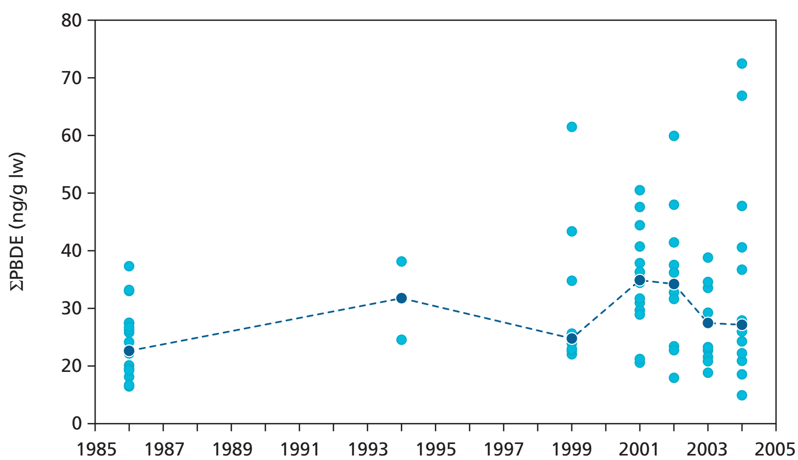

Fig. 17. Temporal trend in ΣPBDE concentration in blubber of young (≤4 years) ringed seals from Ittoqqor-toormiit, East Greenland between 1986 and 2003. The dark blue circles represent the median concentrations connected by a broken line. (Riget et al. 2006).

The lack of trends in our study is not in agreement with the only other time trend study of PBDEs in Greenland biota. The temporal trend of PBDEs in peregrine falcon eggs from South Greenland, covering approximately the same period from 1986 to 2003 was studied by Vorkamp et al. (2005). They found a significant increase of approximately 5 to 10% per year for BDE-99, -100, -153 and -209, but for ΣPBDE the significance level was just above 5%. However, the peregrine falcon in Greenland migrates to Central and South America in winter and therefore the observed increase of PBDEs may not solely reflect contamination of the Greenland environment (Vorkamp et al. 2005). In addition, the majority (33 out of 37) of the data points from the peregrine falcon study are from or before 2001, whereas the ringed seals data have 4 years from 1986 to 2001 and 4 years from 2001 to 2004.

Temporal trends of PBDEs in biota from the Canadian Arctic were recently reviewed by Braune et al. (2005b) and de Wit et al. (2006). Mean concentrations of ΣPBDE in eggs of northern fulmar and thick-billed murres from Prince Leopold Island, central Canadian Arctic Archipelago increased 9.1- and 4.4-fold, respectively, between 1975 and 1998. In male ringed seals, aged 0–15 years, from Holman Island, Western Canadian Arctic a 9-fold increase in ΣSPBDE was reported over the period 1981 to 2000 (Ikonomou et al. 2002). However, more recent results from this group have shown a levelling off or decline from 2000 to 2003 (Ikonomou et al. 2005). ΣPBDE also increased significantly in beluga from southeast Baffin Island over the period 1982 to 1997 (Stern & Ikonomou 2000). The increase in Canadian biota is likely reflecting the North American (>95% of the world total) use of penta-mix formulation, which according to Law et al. (2006) is likely to continue to increase for some time. The increases are typically from data series with data prior to 2001.

Several studies of PBDE concentrations in biota outside the Arctic have shown decreasing trends in the recent years. In eggs of guillemot from the Baltic Sea, a retrospective study including BDE-47, -99, and -100 covering the period 1969 to 2001 showed increasing concentrations from the 1970s to the 1980s, peaking around late 1980s and then followed by a rapid decrease (Sellström et al. 2003). A somewhat similar picture was found in pike (*Esox lucius*) from Lake Bohmen in the southern part of Sweden, where all BDE congeners show increasing trends from late 1960s up to the mid-1980s and then decrease or level off (Kierkegaard et al. 2004). In blue mussels from the Seine estuary, France, the temporal trend picture of PBDEs showed a marked increase during the period 1981 to 1991–95 followed by a levelling off and a possible beginning of decrease until 2003 (Johansson et al. 2006). These studies are all closer to the highly industrialized areas than the Arctic and the observed decreasing trend in recent years is likely to be the result of regional phasing out and restricted use of PBDEs by several Baltic and European countries.

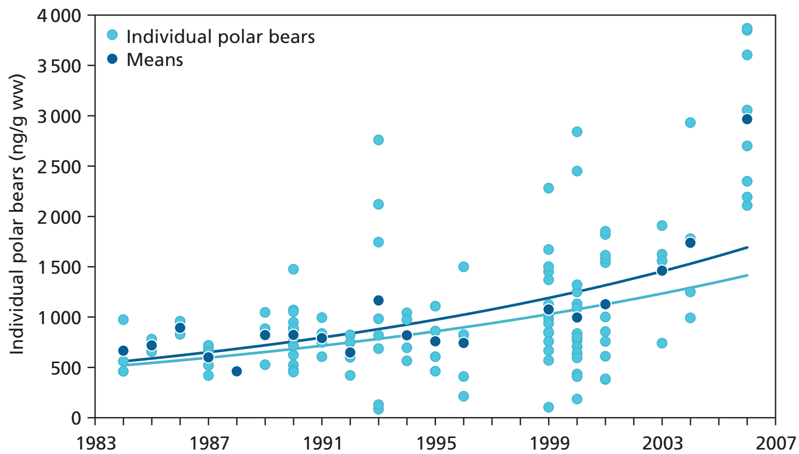

Fig. 18. Temporal trend in PFOS concentration in liver of polar bears (n=119) from Ittoqqortoormiit (East Greenland), 1984-2006 showing a significant ($p = 0.0001$) increase of 5.2% per year. The dark circles represent the mean concentrations and the light blue circles the analysed single values. The dark blue line represents an exponential curve determined with log-linear regression analysis on all values and the light blue line is calculated on means only (data from Dietz et al. 2008).

PFCs

Recent results of PFCs in East Greenland polar bears from the period 1984–2006, revealed 19 years of data being the best time series in Greenland of OHCs in biota. Bears (n=119) in the the age category of 3–5 years were selected among the 463 bears sampled over the 23 years of sampling. Increases could be documented for all 8 PFCs analysed of which 7 were highly significant. The yearly significantly increases using the exponential model varied from 2.3 to 8.5 % per year (Fig. 18; Dietz et al. 2008).

The most recent year of analysis (2006) revealed concentrations of PFCs in the following decreasing order: PFOS (2965 ng/n ww), PFNA (206 ng/n ww), PFUnA (101 ng/n ww), PFDA (88.1 ng/n ww), PFTrA (57.2 ng/n ww), PFOSA (25.2 ng/n ww), PFOA (14.0 ng/n ww) and PFDoA (12.5 ng/n ww). This means that PFOS and ΣPFCs are now higher than ΣPCBs, the highest of the OHCs found in the East Greenland polar bears, assuming that the decreases have continued after 2001 (**Paper 20**). If we extrapolate the calculated increases 100 year ahead in time the PFTrA, PFUnA, PFNA, PFOS, PFDoA, PFDA, PFOA and PFOSA would increase by 3525, 614, 465, 151, 151, 94, 49 and 2 fold respectively. If a concentration level of 77 000 ng/g lw, as documented for ΣPCBs in Baltic ringed seals (Helle et al. 1976), can be anticipated for poor reproduction in polar bears caused by PFCs as well, then reproduction failure caused by ΣPFCs and PFOS and PFTrA alone can be expected by year 2065, 2071 and 2094 respectively. Such predictions are likely to be conservative estimates as the analysed concentrations are on wet weight basis. Recalculating these into lipid weight basis the concentrations would likely double the concentrations moving the years of detrimental effects 8 to 13 years closer. For ΣPFCs this scenario lies only 46 years ahead in the future. Other effects such as poor reproductive success in harbour seal blood has been documented around 25 000 ng/g lw and mortality in half the litter has been found between 40 000 and 60 000 ng/g lw in mink muscle (Boon et al. 1987, Kihlström et al. 1992, Olsson et al. 1996). Such concentrations for ΣPFCs can be expected around 2031 and 2040–2047.

Recent investigations on ringed seal liver tissue from Qeqertarsuaq and Ittoqqortoormiit have likewise revealed a significant increasing trend in perfluorooctane sulfonate (PFOS), perfluorodecanoic acid (PFDA) and perfluoroundecanoic acid (PFUnA) concentrations in the magnitude of 8.2, 3.3 and 6.8% increase per year respectively (Fig. 19; **Paper 22**). The annual increases on the Greenland west coast were lower, being 4.7% for PFOS, 1.7% for PFDA and 5.9% for PFUnA.

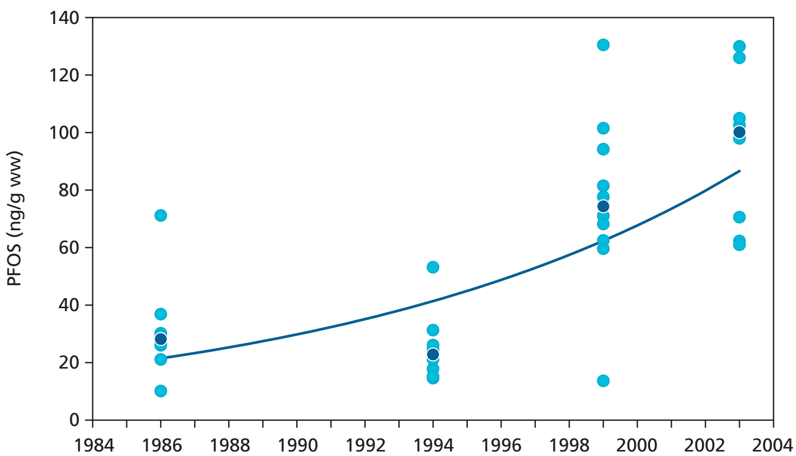

Fig. 19. Temporal trend in PFOS concentration in liver of ringed seals from Ittoqqortoormiit (East Greenland), 1986–2003. The dark blue circles represent the median concentrations. The line represents an exponential curve determined with log-linear regression analysis (**Paper 22**).

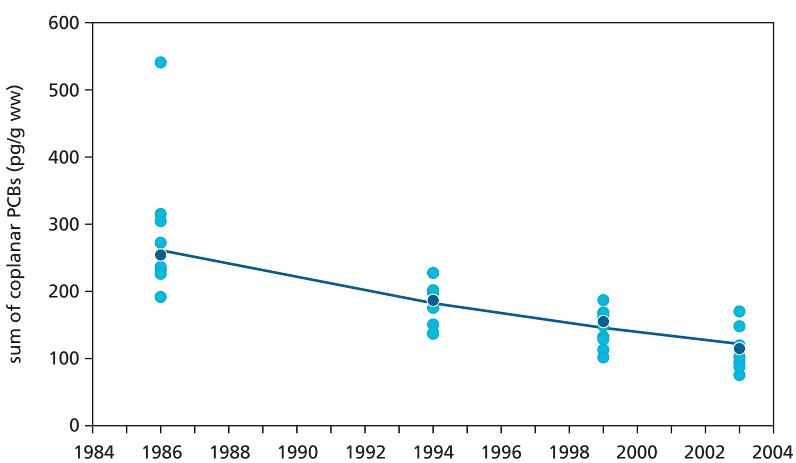

Fig. 20. Temporal significant (p=0.01) decrease of 4.5% per year of non-ortho PCBs (CB77, CB126 and CB169) concentrations in ringed seal blubber from Ittoqqortoormiit, Central East Greenland. Light blue circles are individual concentrations, dark blue circles are median values and the line represents the result of linear regression analyses of logarithmic transformed median values (**Paper 23**).

No information on time trends for PFCs was presented from the Canadian region in the review by Braune et al. (2005b). However, temporal trends in PFCs were recently investigated in liver samples from two ringed seal populations in the Canadian Arctic, Arviat, Western Hudson Bay (1992, 1998, 2004, 2005) and Resolute Bay, Lancaster Sound (1972, 1993, 2000, 2004, 2005) (Butt et al. 2007). C9-C15 PFCAs showed statistically significant increasing concentrations during 1992–2005 and during 1993–2005 at Arviat and Resolute Bay, respectively. Conversely, PFOS and PFOSA concentrations showed maximum concentrations during 1998 and 2000 at Arviat and Resolute Bay, with statistically significant decreases from 2000 to 2005. In the case of Arviat, two consecutive decreases were measured from 1998 to 2003 and from 2003 to 2005. These results indicate that the ringed seals and their food web are rapidly responding to the phase out of perfluorooctane sulfonyl fluoride based compounds by 3M in 2001 (Butt et al. 2007).

Coplanar PCBs and dioxins

Coplanar PCBs and dioxins have not yet been investigated for time trends in the extensive collection of samples available from polar bears from East Greenland. However, in a recent study by Riget et al. (**Paper 23**), East Greenland ringed seal blubber showed an annual decrease of 6.3% ($p=0.26$), 2.1% ($p=0.25$) and 4.5% ($p=0.01$) in ΣPCDDs, ΣPCDFs and non-ortho PCBs, respectively in the period between 1986 and 2003 (Fig. 20). The small number of years ($n=4$) of data available is believed to be the reason for the lack of significance of the trends in ΣPCDDs and ΣPCDFs.

Concentrations of ΣPCDD and ΣPCDF decreased in eggs of northern fulmars collected between 1975 and 1998 from Prince

Photo 5. Polar bears sampled since 1984 from the Greenlandic traditional hunt constitute the best time series for contaminant studies. Photo: R. Dietz.

Leopold Island, Northeastern Canada, whereas the pattern was less clear in thick-billed murres, mainly due to a slight increase in concentrations in 1993 (Braune et al. 2005b). Concentrations of total non-ortho PCBs also decreased in both fulmars and murres between 1975 and 1998 (Braune et al. 2005b). Stable nitrogen isotope analyses indicated that the temporal trends in OHC concentrations in seabird eggs were not generated by a shift in trophic level over time (Braune et al. 2001, 2005b).

Requirements for convincing time-trend studies

Having reviewed a large number of analyses and varying results it is relevant to consider what is actually required in terms of numbers of years of sampling and investigations before reliable conclusions can be drawn regarding temporal trends.

Riget et al. (2000b) investigated the requirements for length of time series within the Greenland environment by use of power analysis. The levels of PCBs, HCB, HCHs, DDTs, Cd, Pb, Hg and Se, and especially the variability in biota obtained during Phase 1 of the Greenland AMAP-programme, were used to illustrate the ability of the programme to detect differences in contaminant levels over time. The statistical power of t-tests for comparison of contaminant levels was illustrated according to various scenarios of magnitude of change, significance level and sample size. We concluded that the ability to detect differences was rather poor for many combinations of contaminants and media, and that long time series are needed before temporal trends are likely to be detected. It was documented that a time series of approximately 10–17 years was required (PCBs in blue mussels: 10 years; PCBs in 2–4 year old ringed seals: 12 years; Hg in polar cod (*Boreogadus saida*): 13 years; and Cd in 2–4 year old ringed seals: 17 years) to detect a linear trend with a change of 10% per year within a significance level of 5% and a power of 80%.

Bignert et al. (2004) likewise analysed the statistical power to detect temporal trends of mercury in Arctic biota, using data gathered during the past two or three decades, mostly under the auspices of AMAP Phases I and II. Most of the investigated time-series covering much of the Arctic were, at that time, too short to possess an acceptable statistical power to detect temporal trends. In order to estimate the sensitivity of a typical Hg-time-series with varying sample frequency, the result showed that the statistical power of a trend-test was seriously reduced if sampling was carried out at a lower frequency than once a year. If, for example, the desired sensitivity of the monitoring programme is to be able to detect an annual change of at least 9% per year within a time-period of 12 years, the power was approximately 80% with annual sampling. With sampling every second, third or fourth year, the corresponding power was only approximately 40, 20 and 10%, respectively.

As seen from the previous examples, very few Greenlandic time series covers more than 12 years with even fewer data points. So far only the polar bear samples obtained by NERI and GINR in collaboration are meeting this criterion. From East Greenland, samples (muscle, liver, kidney, blubber and hair) from 463 polar bears have been obtained representing 20 years of sampling within at time span of 24 years, from 1983 to 2006 (Fig. 18). This material has recently been investigated for PFCs and hopefully the sampling can be continued and funding can be obtained to get a clearer picture of how the Greenland marine biota have responded recently to changes in other contaminant exposures as well. Such effort has been planned under the IPY in the programme "Bear Health".

Part conclusion on OHC time trends

Using the best available time series, declines in levels of "legacy" OHCs, such as PCBs, DDTs, HCHs, HCB, chlordanes, dieldrin, and coplanar PCBs have been detected. Temporal trends for toxaphene, PCDDs and PCDFs are more uncertain, but may be decreasing. Increases in concentrations of a number of "new" OHCs such as the PBDEs and the PFCs took place prior to the turn of the millennium in the entire Arctic. PFCs continue to increase in Greenland, but there is some evidence that, in recent years, PFCs and PBDEs may have decreased in some other areas. However, these trends of "new" OHCs may have changed in the most recent years, and will be confirmed if monitoring continues. The Greenland trends are, in most cases, consistent with those revealed using longer and better time-series from other parts of the Arctic, and, with some delay, relative to trends observed in Northern Europe. The trends show a clear response to regulations on chemicals that have been introduced at the national and international levels. Many of these trends have, however, been investigated over too few sampling years to provide a clear picture of year to year variation, onset and end of these changes and the relative change over time, all of which could provide useful information on the response time of different contaminants at different trophic levels and for different regions. Some highly relevant sample matrices, such as polar bear samples, are available from the Greenland east coast which provides the potential of a high resolution insight into the time period from 1983 until the present.

Trophic transfer of contaminants in the marine food web

Lower trophic levels

In order to compare heavy metal concentrations in marine mammals we have also investigated lower trophic levels. We have reported data on 14 invertebrate species (**Paper 10, 12, 15**), 19 fish species (**Paper 10, 11, 12, 15**) and, somewhat higher in the food chain, 14 marine bird species (**Paper 3, 12, 15**) from Greenland waters. Some of these include data for many ecosystem species, to provide an overview on levels, biomagnification and also assessment of heavy metals and selenium within the different trophic levels (**Paper 10, 12, 15, 17**). Further details including OHCs have been presented by Johansen et al. (2004a, b).

Heavy metal biomagnification

Hg

Hg shows a clear biomagnification with increasing trophic levels in almost all tissues examined from the Greenland marine ecosystem (**Paper 10**). Soft tissue of copepods, amphipods and molluscs were low in Hg (< 0.020 µg/g ww). Muscle tissue of fish and crustaceans were intermediate, with means ranging between 0.01 and 0.327 µg/g ww, while seals, seabirds and whales were highest, ranging between 0.068 and 0.669 µg/g. In polar bears, however, muscle tissue was extremely low in Hg concentration compared to seals, seabirds and whales and even some of the fish. For liver tissue, the lowest Hg concentrations were found in fish (means: 0.005–0.569 µg/g). Liver concentrations were higher in seabirds (means: 0.046–2.67 µg/g), while seals, whales and polar bears had mean Hg concentrations up to 21.6 µg/g (**Paper 10**; see also Fig. 22). Mean kidney concentrations increased in the order fish (0.027 µg/g) < seabirds (0.105–2.05 µg/g) < seals and whales (0.177–3.29 µg/g), while polar bears were significantly higher (10.8–23.2 µg/g) (**Paper 10**). In a later study, mean values of Hg in kidney as high as 32.0 µg/g were reported for adult East Greenland polar bears (**Paper 16**). The high Hg concentrations in polar bear kidneys were higher than in any other species, but they were linked in a 1:1 molar ratio to Se (**Paper 15**; and section "Mercury and effects"). The trophic increase of mercury was likewise supported by comparisons from specific Arctic marine food chains (**Paper 12**). A recent study by Riget et al. (2007b) investigated total Hg, MeHg and stable isotopes of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) in central West Greenland to investigate trophic transfer of mercury in this Arctic marine food web. The food web magnification factor was not only comparable with those reported for other Arctic marine food web but also with quite different food webs such as freshwater lakes in the sub-Arctic, East Africa and Papua New Guinea. This suggests similar mechanisms of mercury assimilation and isotopic ($\delta^{15}N$) discrimination among a broad range of aquatic taxa and underlines the possibility of broad ecosystem comparisons using the combined contaminant and stable isotope approach. Two extensive Canadian studies comparing Hg in muscle tissue with $\delta^{15}N$ to reflect the trophic level from 27 species from Lancaster Sound in NE Canadian Arctic and 10 species (including *Calanus hyperboreus*, mixed zooplankton, 8 seabird species and ringed seals) from the North Water Polynia in Baffin Bay supported the bioaccumulative and bimagnifying characteristics of Hg (Atwell et al. 1998, Fisk 2002). The human exposure study conducted by Johansen et al. (2004a, b) from Southwest Greenland likewise supported the findings of a trophic accumulation of Hg in the marine food chains.

Cd

In contrast to Hg, Cd showed a less clear picture of biomagnification in the marine food chains. Actually, the conclusions about whether or not Cd magnifies along the food chain depend on which species and tissue groups are being compared. Mean Cd concentrations in muscle tissue increase in the order polar bear (<0.020–0.024 µg/g) < fish (<0.015–0.036 µg/g) < whales (<0.015–0.107 µg/g) < seals (<0.015–0.366 µg/g) < birds (<0.015–0.668 µg/g), which was not a typical pattern of bioaccumulation (**Paper 10**). Mean Cd concen trations in liver tissue, being 20–100 higher than in muscle tissue, followed the sequence: fish (<0.034–2.11 µg/g) < mollusc and crustacean hepatopancreas and polar bear liver (0.477–7.79 µg/g), < seabirds, whales and seals (0.853–36.6 µg/g). Mean Cd concentration in kidney tissue was low in fish (<0.127 µg/g), while polar bears, whales, seabirds and seals were highest (3.47–110 µg/g) (**Paper 10**). Fisk et al. (2003), who only analysed muscle made a comparison of Cd concentrations from *Calanus hyperberous*, mixed zooplankton, seabirds and ringed seals from the North Water Polynia, Northern Baffin Bay and concluded that Cd was not biomagnifying. The human exposure study by Johansen et al. (2004a, b) from Southwest Greenland confirmed high Cd concentrations in marine mammals and seabirds, intermediate and low concentrations in fish and low concentrations in terrestrial mammals. The findings listed above demonstrate the importance of the type of tissue selected for analysis with regard to making conclusions on biomagnification.

Part conclusion on bioaccumulation of Hgand Cd

In summary, it has been shown that Hg shows a clear bioaccumulation in food chains, whereas Cd shows both increases and lack of increases depending on which species and tissue are being compared. Generally, the highest heavy metal exposure from consumption of animals and risks of effects in animals can be expected in species feeding at high trophic levels, especially if kidney and liver is consumed.

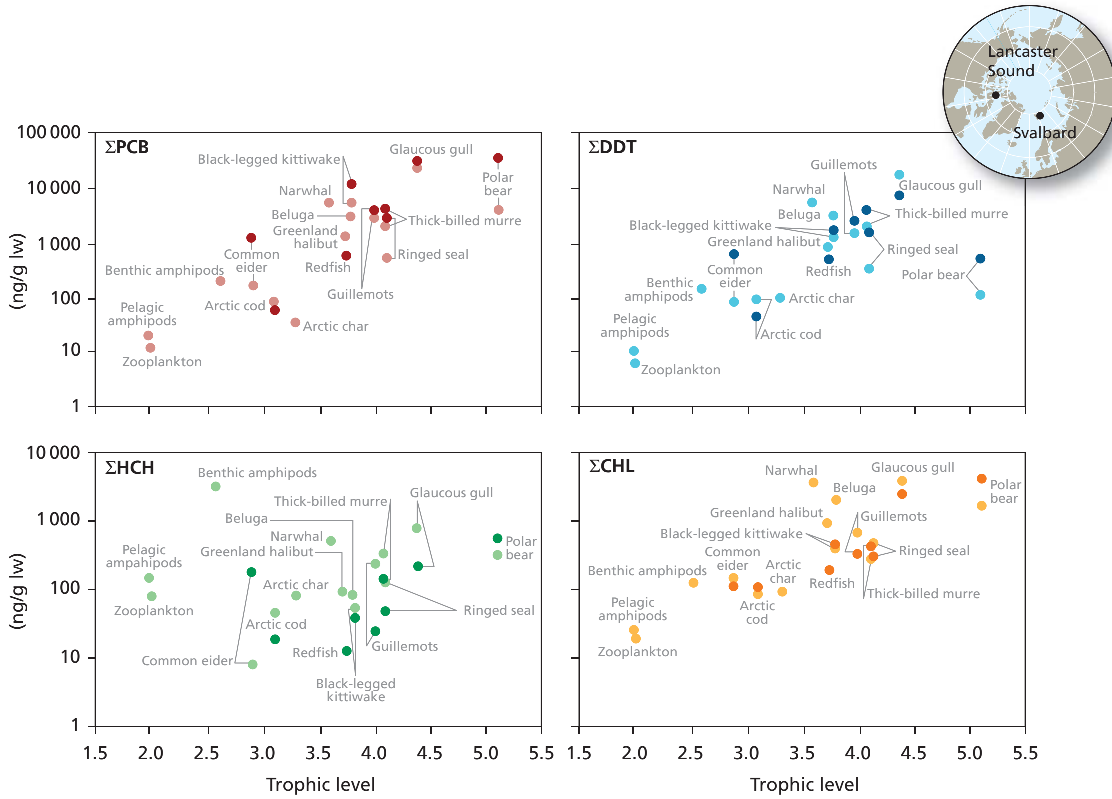

Fig. 21. Correlations between concentrations lipid weight (lw) of major OHC (and trophic levels in the marine food web around Svalbard (dark circles) and in Lancaster Sound (light circles) (de March et al. 1998).

OHC biomagnification

Prior to the AMAP process, OHC data were scarce from Greenland. Data from 1978 were available for walrus (*Odobenus rosmarus*) from Avanersuaq (Born et al. 1981) as well as data on belugas (*Delphinapterus leucas*) from Upernavik and Disko Bay 1985–1990 (Stern et al. 1994). Furthermore, data were available from eight Central West Greenland fish species (Berg et al. 1997). However, the region, period and tissues analysed varied among these three studies, preventing a comparison of trophic levels. The AMAP process generated values from blue mussels, shorthorn sculpin, polar cod, glaucous gulls, Icelandic gulls (*Larus gloucoides*) as well as ringed seals from four areas (Avanersuaq, Qeqertarssuaq, Nanortalik and Ittoqqortoormiit) between 1994 and 1995 (Cleemann et al. 2000a, b, c). These levels were compared by Dietz et al. (**Paper 17**), who concluded that ΣPCB, ΣDDT, ΣHCH and HCB increased in concentrations towards higher trophic levels. The human exposure study by Johansen et al. (2004a, b) from Southwest Greenland provided information on OHC concentrations in a large amount of additional species and tissues (liver, kidney, blubber, meat and mattaq), even though the number of samples for some species and tissue groups was low (n=5 to 20) and no age harmonization was conducted. The study included PCBs (sum of 104 congeners; ΣPCB), DDTs (sum of 6 DDT-related compounds; ΣDDT), chlordane, toxaphene (total and selected congeners), HCH, chlorobenzenes, mirex, octachlorostyrene, and endosulfan. The study by Johansen et al. (2004a, b) confirmed that OHC concentrations were highest in marine mammals and some birds, and almost without exception, OHC levels were higher in the marine than in the terrestrial environment.

In a recent study, OHC data have also become available for narwhals (*Monodon monoceros*) collected between 1985 and 1992 from Northwest Greenland (**Paper 19**) and from polar bears sampled 1999–2001 in Ittoqqortoormiit, East Greenland (**Paper 20**). These data, in comparison with earlier studies and the recent human exposure study by

Johansen et al. (2004a, b), also confirm conclusions based on the extensive number of OHC analysis from Svalbard and the Canadian High Arctic that have demonstrated that OHCs, such as ΣPCB, ΣDDT, ΣHCH and ΣCHL are bioamagnifying in the marine environment (Fig. 21; de March et al. 1998, Fisk et al. 2003, de Wit et al. 2004). Several of these comparisons were supported by stable isotope ($\delta^{15}N$ and $\delta^{13}C$) analysis, which were used as an explanatory variable for the different species. By including information of $\delta^{15}N$, the trophic level can be calculated and the effect of biomagnification can be easily visualised, as was done for e.g. ΣPCB, ΣDDT, ΣHCH and ΣCHL in species from Svalbard and Lancaster Sound (Fig 21; de March et al. 1998).

Levels of PFOS and other fluorochemicals in East Greenland polar bears were compared with fish, birds and other marine mammals from Greenland and the Faroe Island. These data showed that PFOS in polar bears was 20-fold higher than in ringed seals, which were approximately 4-fold higher than black guillemot and shorthorn sculpins (Bossi et al. 2005a). Minke whales (*Balaenoptera acutorostrata*) had PFOS concentrations similar to ringed seals (Bossi et al. 2005a). Biomagnification studies on PBDEs are less well documented. However, investigations on PBDEs in ringed seals and polar bears documents average biomagnification factors between 1.8 (BDE-154) and 71 (BDE-153), dependant of congeners, from 5 regions in the Arctic (Muir et al. 2006, **Paper 28**). Due to the high levels in top predators, the increase between ringed seals and polar bears and the similarity in physical-chemical properties of PBDEs and PCBs, it is likely that they biomagnify in a similar way to the other OHCs.

Part conclusion on biomagnification of OHCs

OHC shows a clear bioamagnification for most compounds around the Arctic. Hence the highest OHC exposures and risks of effects can be expected in species feeding on high trophic level and lipid rich prey species.

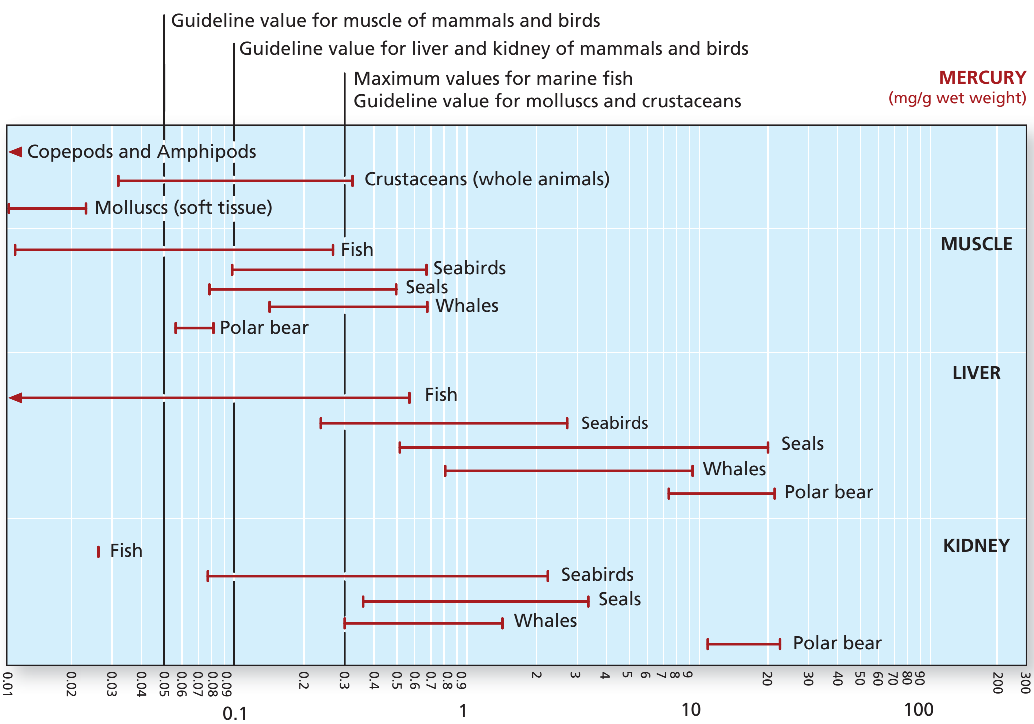

Fig. 22. Overview of ranges of concentrations of Hg in different animal groups relative to guideline values and maximum acceptable values for food products in Denmark (**Paper 10**).

Human exposure

In most regions of Greenland, the Inuit depend on hunting of marine mammals including the polar bear and marine birds for their food. Due to the high trophic position of these species, and consumption of large amounts of internal organs and blubber, concerns have been raised about the possible health effects on individuals and on the population, and questions asked about how this dietary exposure to contaminants can be reduced (Hansen et al. 1998, 2003).

The human exposure issue has been dealt with in several studies. From Greenland, one of the first attempts to describe human contaminant exposure in relation to "Provisorial Tolerable Weekly Intake" (PTWI) was conducted by Johansen et al. (1980). Here, it became evident that levels of both heavy metals and OHCs were high in diets using certain foods from the Greenland marine environment. Based on a larger sample sizes and more species, an overview article by Dietz et al. (**Paper 10**) summarised the available heavy metal information from Greenland at that time and discussed the results in relation to the guideline and maximum acceptable values for food products in relation to Danish legislation (Fig. 22). It was evident that concentrations of heavy metals such as Hg and Cd were high in a number of species and in many cases far above the guideline and maximum tissue residue values for consumed species.

Human exposure to Hg

It was concluded that the Danish Food Standard guideline limits for Hg in meat (muscle) was exceeded in almost all groups of birds (except yearlings) and mammals, whereas in crustaceans and fish Hg was below the relevant Food Standard Limits. In addition, the guideline value for liver and kidney was exceeded in all groups of seabirds and marine mammals (except for yearlings) (**Paper 10**). Mercury in adult polar bear liver and kidney exceeded the guideline value by 216- and 232-fold, respectively.

Human exposure to Cd

For Cd, similar conclusions were made. The Danish guideline muscle value was not exceeded for fish, whereas birds and mammals exceeded the value in most cases. The Food Standard maximum limits for liver and kidney were exceeded in all seabirds and marine mammals. The highest mean levels found in 5–10 year old seals from Northwest Greenland exceeded the limits 73-fold in liver and 110-fold in kidney (**Paper 10**) A similar overview of heavy metal ranges was also conducted by Dietz et al. (**Paper 12**) for Greenland relative to the rest of the Arctic. The variation in guideline values from the different Arctic countries was, however, not considered. The conclusions were similar with respect to trophic accumulation; Greenland biota showed the highest concentrations of Cd and intermediate levels of the other metals and elements that were compared (Hg, Se and Pb; **Paper 12**). However, Greenlanders still received the highest exposure due to amount of ingested traditional food (e.g. Nilsson 1997, Hansen et al. 1998, 2003, Nilsson & Huntington 2002).

Human exposure to heavy metals and OHCs in the Disko Bay area

Recently, more detailed work has been conducted in the Disko Bay area, Central West Greenland (Johansen et al. 2004a, b), where the most important species and tissues were selected on the basis of an investigation of dietary habits in this area by Pars et al. (2001). Johansen et al. (2004a, b) investigated both heavy metals and OHCs, evaluated relatively to international Provisorial tolerable weekly intake (PTWI) guidelines (Fig. 23). Dependent on their importance for consumption, between 5 and 20 of each sample was analysed for a large number of contaminants and again compared to the acceptable daily intakes (ADI) and tolerable daily intakes (TDI) recommended by FAO and WHO (Johansen et al. 2004a, b). Based on the results from Johansen et al. (2004a, b) a risk assessment has been conducted using no observed adverse effect levels (NOAEL) and margins of safety (MOS) (Nielsen et al. 2006). This investigation expressed concern about potential for health effects among Greenlanders from Hg, Cd, Pb, PCBs, chlordane and toxaphene. However, a relatively low concern for adverse health effects was expressed for chlorobencenes, HCH, and dieldrin, and low concern for Se and DDT (Nielsen et al. 2006).

Johansen et al.'s (2004a, b) study included Cd, Hg, Se, PCB, DDT, chlordane, HCH, chlorobenzenes, dieldrin and toxaphene in the major species and tissues consumed by Greenlanders. They concluded that the traditional diet was a significant source of contaminants to people in Greenland, although contaminant levels varied widely among species and tissue. The levels ranged from very low in many species and tissues to very high in a few. Furthermore, contaminant levels were very low in terrestrial species and in muscle of many marine species (e.g. Fig. 23). High OHC concentrations were typically found in blubber of marine mammals, and high metal levels in liver and kidney of seals and whales. In their study, the mean daily intakes of Cd, chlordanes and toxaphene significantly exceeded "acceptable/tolerable daily intakes" (ADI/TDI) by a factor between 2.5 and 6, respectively. Mean intakes of Hg, PCB and dieldrin also exceed ADI/TDI by up to approximately 50%. Johansen et al. (2004a, b) applied calculated figures for mean intakes. Since variation in both food intake and contaminant levels was large, some individuals will be exposed to significantly higher intakes than others. The positive outcome of this study was that the mean intakes of DDT, HCH and chlorobenzenes were well below the ADI/TDI values, and that it seemed unlikely that the TDI for these contaminants will normally be exceeded in the Greenland population. The evaluation of contaminant intake of Johansen et al. (2004a, b) pointed to seal muscle, liver, kidney and blubber as well as whale blubber as the dominant contributors of contaminants in the traditional diet. Levels in liver from Greenland halibut, snow crab, king eider, kittiwake, beluga and narwhal were also high. Kidney of beluga and narwhal were also high, but were, with the exception of toxaphene in Greenland halibut liver, not identified in this study as important sources, because they were eaten in low quantities. Johansen et al. (2004a, b) suggested that a way to minimize contaminant intake would be to avoid or limit the consumption of diet items with high contaminant levels. By assuming a traditional diet composition in their study without fish liver, bird liver, seal liver, seal kidney, seal blubber, whale liver, whale kidney and whale blubber, the intake of all contaminants would be below the TDIs. Such a change in food preference would result in a reduction of the intake of the amount of traditional food of only 24–25%, and according to Johansen et al. (2004a, b) such a change in diet would not be likely to result in deficiency of minerals, vitamins or other nutritional compounds. Recent studies by Gebbink et al. (in press a, b) indicates that a number of OHCs (dieldrin, ΣCHL, Σ-MeSO2-p,p'-DDE, Σ-MeO-PBDE, Σmirex, Σ-OH-PCBs, PCP, ΣPBDE) are higher in liver than adipose tissue.

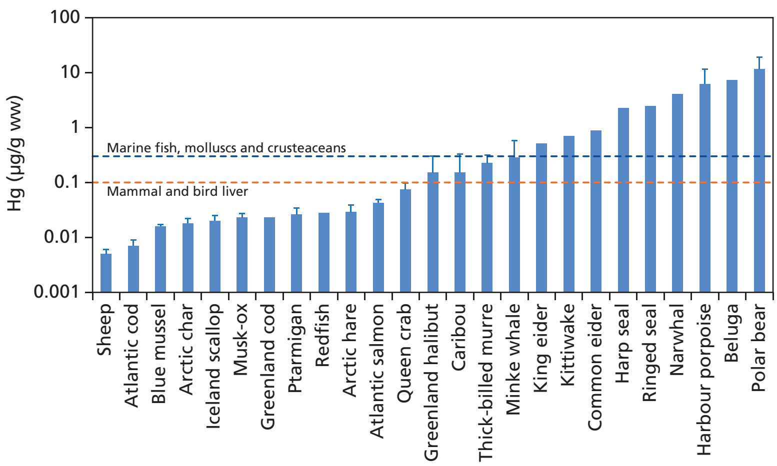

Fig. 23. Overview of Hg mean concentrations in animal liver (and whole molluscs) consumed regularly in the Disko Bay area of Central West Greenland. Guideline values and maximum acceptable values for Danish food products are indicated by lines (data from Johansen et al. 2004a, b).

Part conclusion on human exposures of contaminants in Greenland

It has been shown that traditional food including marine mammals from Greenlandic waters contains high levels of metals and OHCs. For certain organs and tissues, based on current dietary habits, recommended guideline values for tolerable or acceptable intakes of some contaminants are exceeded. The Inuit population can minimize their contaminants intake and thereby reduce their risk of effects by reducing their intake of internal organs such as liver and kidney to reduce exposure to Hg, Cd,

Photo 6. The Greenland Inuit population is exposed to high concentrations of OHCs and heavy metals through their high intake of local country food. However, levels can be reduced by advise on food intake and through international regulation of long range transported contaminants. Photo: R. Dietz.

PBDEs and PFOS, as well as blubber, adipose tissue and other fatty tissue to reduce exposure to lipophilic OHCs. Contaminant exposure can further be reduced by preferentially consuming young individuals and species of low trophic levels.

Contaminant related pathological effects and diseases

Having identified contaminant concentration, levels and trends, the next obvious step is to link these concentrations to possible biological effects. Such work has recently been included among our activities in accordance with the recommendations from the AMAP Phase I assessment. Biological effects are a large and complex subject, and can be studied at different levels of biological organization, from the molecular to the ecosystem level. Several of these aspects have been dealt with in great detail during the AMAP Phase I and II processes (**Paper 12**, de March 1998, de Wit et al. 2004, Derome et al. 2005).

Several approaches are being used to identity and estimate the risk of possible effects.

Laboratory experiments
The first approach, which has been used extensively within the AMAP process, involves comparison of tissue concentrations in relation to values derived or extrapolated from primarily laboratory experiments. Some of the difficulties in extrapolation relate to differences in sensitivity, where the same types of effects are seen but at different doses. Also laboratory animals are most often exposed to single OHCs or technical products at high doses for short periods of time, and it is difficult to extrapolate the toxic effects seen at high acute doses to possible adverse effects at lower but chronic exposures. Finally the question is whether effects observed in non-Arctic species investigated in laboratory studies can be transferred to Arctic species.

Effect studies in free ranging animals
Wild animals are generally exposed to lower concentrations of OHCs than laboratory animals in experimental studies; however the former are also typically exposed to mixtures of OHCs and to other stressors such as cold temperatures and fasting conditions, and they are exposed over their entire lifetime. They are also exposed to "weathered" chemical mixtures due to the change in composition of OHCs caused by abiotic degradation, metabolism and subsequent filtering up through the food web.

Biomarkers
Another approach used to study biological effects is examination of "biomarkers" – subtle indicators of biological responses to contaminants. These approaches have also been reviewed during the AMAP process, but so far no such analysed have been conducted in Greenland free ranging animals, largely due to the considerable logistic challenges involved in applying these methods in the relevant high trophic level biota. It was decided not to present and repeat information on tissue concentration comparisons to effects and biomarker issues in this dissertation, as this

material is quite extensive and no additional information has been provided through our own work.

Effect studies included in the present dissertation

Other approaches to study biological effects include detoxification response, effects on reproductive organs (size and tissue alterations), internal target organs such as liver, kidney and selected immunological organs, effects on cranium and bone (gross skull pathology, bone mineral composition fluctuating asymmetry), as well as immune response and clinical-chemical blood parameters, and these are being addressed in the present thesis as relevant new information is emerging from our work. Effects have only been studied in a few species in Greenland and then only in relation to species, regions and contaminants where levels are among the highest in the Arctic so as to obtain the highest likelihood of detecting possible subtle effects. The AMAP work formed an important process for obtaining the necessary overview for identifying relevant species, areas and contaminants for possible effects studies (**Paper 12**, de March et al. 1998). Mercury in Greenland polar bears and Cd in ringed seals from Greenland were considered high enough to cause concern for effects, which is why these species were investigated. As polar bears also were among the highest exposed species for OHCs, and because bears from East Greenland and Svalbard were among the highest exposed populations it was recommended in the AMAP Phase I assessment to investigate the health of the polar bears in this region (de March et al. 1998). As the polar bear is protected on Svalbard, East Greenland was the only area where internal organs could be obtained in reasonable numbers, and NERI in collaboration with GINR therefore initiated studies investigating effects of OHCs in polar bears from this area. This effort later expanded to a controlled experiment on sledge dogs which were fed foods similar to the diet of polar bears. It is important to bear in mind that the effect studies carried out on East Greenland polar bears has been carried out on bears from 1999–2001 with rather low OHC exposure. The investigations were conducted at a time where the old OHCs had been decreasing for many decades and PFCs and PBDEs had only been increasing for few decades (see also "Temporal trends of OHCs").

Mass mortality and contaminant exposure

OHCs are believed to have an immunosuppressive effect on the immune system, and thus may reduce resistance against diseases. One of the most significant and well studied disease outbreaks in marine mammals concerns the Phocine Distemper Virus (PDV), which had two major outbreaks in Danish waters in 1988 and 2002 resulting in the death of 22 000 and 32 000 harbour seals, respectively (**Paper 2, 26**). The disease is believed to originate from seals from Arctic waters (**Paper 1**, Duignan et al. 1994, 1995a, b, c, 1997, Nielsen et al. 2000). This high mortality rate of this disease is also discussed below in relation to contaminant exposure.

Mercury and effects

Human health effects of Hg

Mercury levels are high in the Greenland and Faroese human population and therefore a special focus of studies has been on human health effects. Over the past decades, epidemiological studies on human health effects related to exposure to Hg (and PCBs) have been oriented toward prenatal exposure and children's health. These have largely focused on neurological systems, but recently cardiovascular effects have gained attention (Dewailly & Weihe 2003). Clinical neurological examination of children from Qaanaaq, NWG did not reveal any obvious negative effects. However, auditory evoked potentials showed possible Hg exposure related deficiencies, although only statistically significant in a few cases (Weihe et al. 2002). In studies on the Faroe Islands, Grandjean et al. (1997) reported associations between maternal hair Hg concentration during the pregnancy period and cord blood Hg concentrations and children's performance in neurobehavioral tests, dealing with fine motor function, ability to concentrate, language, visual–spatial abilities and verbal memory. Of these, the neuropsychological dysfunction was the parameter

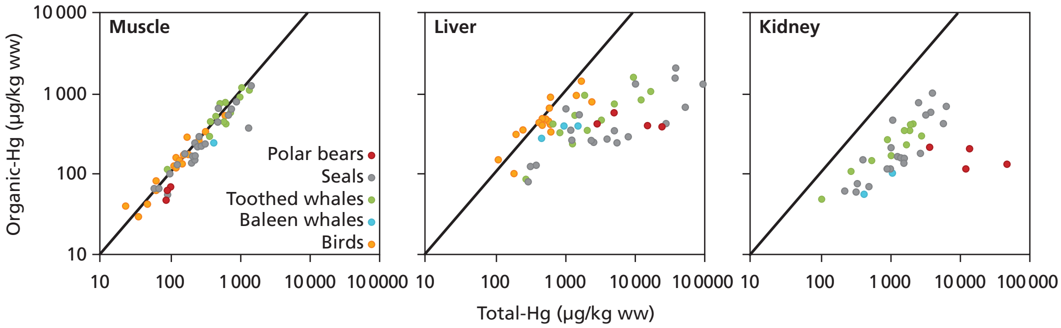

Fig. 24. Ratio between organic and total Hg in muscle (left), liver (center) and kidney (right) in various animal groups in Greenland (**Paper 4**).

with the strongest linkage to cord blood Hg concentration (Grandjean et al. 1999). Grandjean et al. (1992, 1997, 1999) had difficulties in determining whether effects such as those on language and memory function observed in children, were due to prenatal exposures to Hg, PCB, or to both. However, patterns of neurobehavioral effects attributed to developmental Hg exposure in humans resembled those seen in experimental animals in relation to motor, sensorimotor system effects and cognitive effects.

Salonen et al. (1995) suggested that the high mortality from coronary heart disease observed among fish eaters from Finland could be explained by the high Hg content in lean freshwater fish. Mercury can promote the peroxidation of lipids, resulting in more oxidized low-density lipoproteins (LDLs), which have been implicated as an initiator of arteriosclerosis. The enhanced risks of death from coronary heart disease were seen in combination with low serum Se concentration, a situation that is seldom the case in Greenland, where Se is generally present in molar surplus in the dominant marine foods, especially in whale skin (**Paper 13, 15, 16**). Selenium was hence believed to be an antioxidant that can block the Hg-induced lipid peroxidation (Salonen et al. 1982). Contrary to the situation in eastern Finland, the mortality rate from coronary heart disease in Inuits is extremely low, as they have a high consumption of marine mammals and fish being high in both Se and polyunsaturated (n-3) fatty acids (Dewailly et al. 2001a).

Organic vs inorganic Hg in Greenland food

Even though Hg in Greenland biota is not the highest in Arctic, there has been an interest in elucidating the source of the elevated Hg levels that are observed in the Inuit population. More than 95% of the methyl-Hg in foods is taken up by mammals, whereas the corresponding figure for inorganic Hg is only 15% (Berlin 1986). Dietz et al. (**Paper 4**) therefore examined muscle, liver and kidney in 20 species of birds, seals, toothed whales, baleen whales and polar bears from the Greenland marine environment for total and organic Hg. The investigation revealed that the major part of the Hg present in muscle tissue was organic (Fig. 24, **Paper 4**). This was also the case in liver when total Hg concentrations were below 2 µg/g ww, however any further increase in the total Hg (up to over 100 µg/g ww) did only result in a corresponding increase in the in-organic fraction (Fig. 24, **Paper 4**). The percentage of total Hg in the kidney that is in organic form is, on average between 10 and 20% for species other than polar bears where this percentage was <6% (Fig. 24, **Paper 4**). Seen from an animal health perspective, the low percentage of organic Hg, even at high total Hg exposures indicated that the Hg could be demethylated and stored in an inert inorganic form as also later documented (**Paper 15**). From a human consumption point of view, the low percentage of organic Hg in tissues where it accumulates, like liver and kidney, make the Hg less bioavailable and less toxic.

Inorganic Hg mediates the liver and kidney toxicity through high affinity to a variety of enzymes in e.g. microsomes and mitochon-

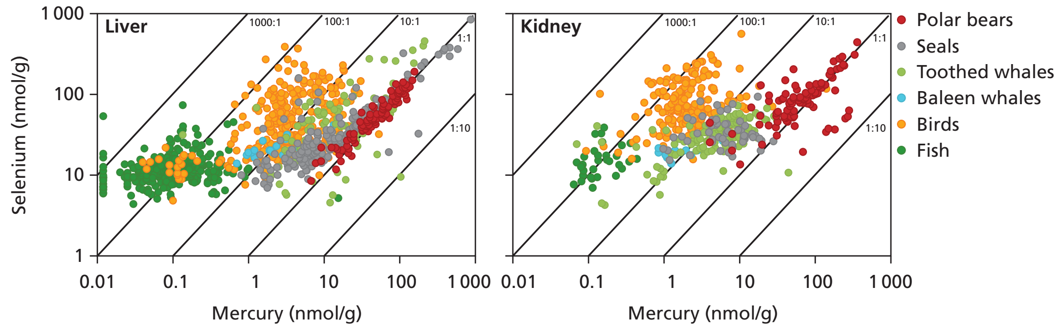

Fig. 25. The Hg-Se relationship in liver (left) and kidney (right) tissue in various animal groups including 48 marine species from Greenland. The lines represent different decadal molar ratios (**Paper 15**).

dria via SH-groups (co-enzym inhibitor) inducing cellular toxicity, although the metallothionein-Hg and Se-Hg complex bindings are believed to have a preventive effect (Goyer & Clarkson 2001). As the Hg concentrations increase it is most likely that the majority of the Hg at higher concentrations is being bound to the inert Hg-Se complex tiammanite, which the organism uses to detoxify and store the surplus of Hg. In order to evaluate this finding, we analysed the Hg-Se relationship on a molar basis for muscle, liver and kidney tissue obtained from more than 5000 individual animals (**Paper 15**). The Se/Hg approached a 1:1 ratio when concentrations increased above ca. 10 nmol equivalent of ca. 2 µg/g ww in liver as seen primarily among seals, toothed whales and polar bears. In kidney a similar pattern seemed to be present in the highest exposed animals, which were primarily polar bears (Fig. 25; **Paper 15**).

This threshold, in order of magnitude, coincides with the point where organic-Hg ceased to increase (Fig. 24; **Paper 4, 15**). When evaluating the toxicity of these concentrations, a total Hg concentration above 30 µg/g ww in both liver and kidney is believed to be lethal or harmful to wildlife and birds (Thompson 1996), whereas the threshold is believed to be twice as high in liver for marine mammals (Law 1996). The threshold for terrestrial wildlife is exceeded for a number of polar bear individuals in both kidney and liver, whereas only few seals and toothed whales are above the marine mammal liver threshold level. In fact, the mean Hg concentration presented by Dietz et al. (**Paper 16**) for East Greenland polar bear kidneys (32 µg/g ww) are above the threshold for terrestrial wildlife, whereas the corresponding figure for liver tissue was slightly below the threshold.

Mercury effects in polar bear

Even though Greenland is not, in terms of geography, the area with the highest Hg exposure, Hg concentrations in liver and kidneys were high enough to cause concern, with mean Hg in, for example adult East Greenland bears at 13.4 µg/g ww in liver and 32.0 in kidney (**Paper 16**). Consequently, some individuals exceed the lethal or harmful threshold level of 30 µg/g ww for terrestrial mammals (Thomson 1996, **Paper 12**). Sonne et al. (2007b) therefore investigated the histopathological impact of Hg on East Greenland polar bear liver and kidney tissues collected between 1999–2001. Liver Hg levels ranged from 1.1–35.6 µg/g ww and renal levels ranged from 1–50 µg/g ww in samples collected during this period. Of these, 2 liver values and 9 kidney values were above the known toxic threshold level of 30 µg/g ww in terrestrial mammals. Evaluated after age-correcting ANCOVA analyses, liver Hg levels were significantly higher in individuals with visible Ito cells, and a similar trend was found for lipid granulomas. Liver Hg levels were significantly lower in individuals with portal bile duct proliferation/fibrosis, and a similar trend was found for tubular hyalinisation in renal tissue. Based on these relationships and the nature of the chronic inflammation we concluded that the lesions were likely a result of recurrent infections and ageing but that long-term exposure to Hg could not be excluded as a co-factor (2007b).

Mercury effect on concentrations of neurochemical receptors

In two recent studies, Basu et al. (2005a, b) investigated whether Hg exposure could be related to concentrations of neurochemical receptors (muscarinic cholinergic (mACh) and dopaminergic-2 (D2) systems) in brain tissues of river otters (*Lontra canadensis*) and mink (*Mustela vison*). In both cases a negative effect of Hg was detected and it was concluded that these neurochemical receptors were useful as novel biomarkers of Hg exposure and neurotoxic effects in wildlife. Such investigations are now being conducted on brain tissue of East Greenland polar bears with collaborators in Canada.

Cadmium effects

Histopathological changes in kidneys have been described in cadmium-exposed laboratory mammals and in humans (e.g. Scott et al. 1977, Squibb et al. 1979, Friberg et al. 1986, Yasuda et al. 1995). Concentrations of Cd in the kidney of ringed seals from northwest Greenland are among the highest recorded Cd tissue concentrations in Arctic marine mammals, reaching levels that may induce renal histopathological changes (Fig. 26, **Paper 12**). Dietz et al. (**Paper 14**) investigated renal histology in 15 of 462 ringed seals within three different concentration ranges. Of these, 5 were above 200 µg/g ww. From the analysis it was concluded, that there was no evidence of classic tubular Cd induced renal lesions and that ringed seals could have adapted to the high Cd levels (**Paper 14**). However, in order to get more information on the possible effects, we conducted an additional study on 100 Northwest Greenland ringed seals using optimal fixation for histology (Sonne-Hansen et al. 2002). Thirty-one of these had Cd concentrations above 50 µg/g ww, 11 above 100 µg/g ww and one above 200 µg/g ww. Ten seals had obvious histopathological changes, categorised mainly as glomerulonephritis. However, none of these changes were consistent with classic Cd-induced tubular damage. It is known that Cd-induced metabolic dysfunctions can induce osteopenia (demineralisation) of the lumbar vertebrae in humans (Friberg et al. 1986, WHO 1992a).Therefore, the three lowest lumbar vertebrae were scanned to measure bone mineral density in order to evaluate possible Cd induced demineralization (Fanconi's syndrome). No significant correlations were however found between skeletal mineralization and Cd concentration, renal lesions, age or sex, respectively (Sonne-Hansen et al. 2002).

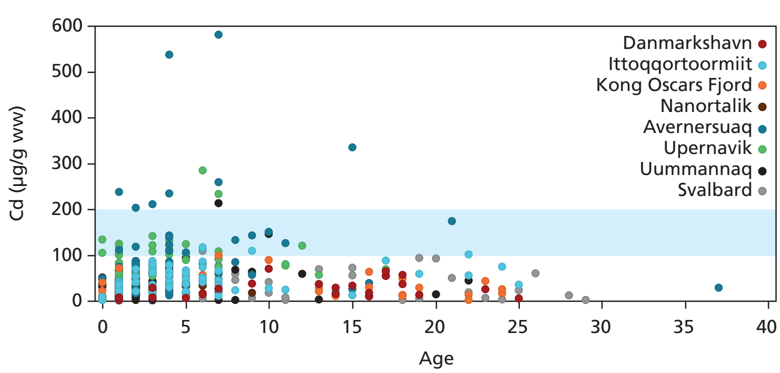

Fig. 26. Cd concentrations in Greenland ringed seals kidneys often exceed the 100 and 200 µg/g ww threshold limits for Cd effects (**Paper 12, 14**).

Cadmium and Hg effects in other species

Histopathology linked to contaminant analyses were performed on bowhead whales (*Balaena mysticetus*), beluga whales, and ringed seals from Arctic coastal Alaska. The concentrations of Cd and Hg in liver and kidneys of some animals occurred at concentrations that would be considered toxic in domesticated species, but no lesions indicating chronic heavy metal toxicosis were detected (Woshner et al. 2000, 2001a, b). Autometallography (AMG) granules were evident in belugas, where total Hg ranged up to 17.1 µg/g ww in liver and up to 82.5 µg/g ww in kidney. Mean areas occupied by AMG granules correlated well with hepatic Hg concentrations and age (Woshner et al. 2002). Another histopathological study was performed on kidney tissues of Atlantic white-sided dolphins (*Lagenorhynchus acutus*) off the Faroe Islands (Gallien et al. 2001). Kidney tissues showed Cd concentration in the range of 22.7 to 31.1 µg/g ww and Hg concentrations from

0.1 to 2.5 µg/g ww. There were abnormalities in parts of the kidneys in two of the three mature animals, as well as calcium phosphate concretions containing Cd found in the kidney tissue of the same individuals. The occurrence of metal-containing granules has also been reported in the liver of marine mammals and birds and in the respiratory tract of cetaceans (Derome et al. 2005). In invertebrates, these granules are for storage and immobilization of Cd and/or for detoxicification of the metal (Derome et al. 2005).

Part conclusion on effects of Hg and Cd in Greenland top predators

Epidemiological studies on Arctic populations indicate neuropsychological dysfunction in some humans that resemble effects seen in experimental animals. Initial investigations of histopathological and neurochemical receptor biomarkers indicate that effects of Hg can not be excluded. Lesions found in polar bear liver and kidneys were likely a result of recurrent infections and ageing, but that long-term exposure to Hg could not be excluded as a co-factor. Cardiovascular and possibly other effects of Hg in higher trophic levels may be reduced and in some cases eliminated due to the protective effect of Se being present in surplus in the Arctic marine ecosystem. Although Cd concentrations in several marine species are above threshold effect levels, Cd has so far not been shown to cause effects in Arctic wildlife.

Effects of OHCs

East Greenland, Svalbard and the Kara Sea, have been documented as the Arctic areas with the highest OCH concentrations, and the polar bear was shown to be among the highest exposed species in the Arctic during AMAP Phase I assessment. The assessment recommended that Arctic countries investigate health effects in species having tissue concentrations at or above levels of concern. For the Greenland region, it was therefore logical to start investigating the possible effects on polar bear. Greenland provided a unique opportunity to obtain samples in reasonable numbers from the traditional hunt. Therefore histopathological investigations on polar bears were started in 1999. In most other Arctic areas, investigations into effects and OHC analysis had been restricted to blood samples and adipose biopsies conducted in regions where the polar bear is protected. This was the case at Svalbard, in Russia and partly in Canada, where samples were obtained during the handling of polar bears in connection with tranquilisation and mounting of satellite collars or conventional tags (e.g. Norstom et al. 1998, Oskham et al. 2003, 2004, Brathen et al. 2004, Lie et al. 2004, 2005, Verrault et al. 2005). Hence, the polar bear studies in East Greenland have provided a unique opportunity to investigate the potential effects of contaminants on a real-world highly exposed species. Notwithstanding this unique study system, it is inevitably based on correlational and descriptive analyses. To improve the understanding and entangling the potential effects of the cocktail of exposure to contaminants and food stress, we expanded the effect investigations to include a controlled experiment in 2004–2005 using sledge dogs (*Canis familiaris*) as a surrogate for polar bears, mimicing the polar bear exposure to POPs. This was done to be able to define and compare an exposed and an unexposed group, which is not possible in free-ranging polar bears.

OHC effects on reproductive organs

Sexual organs

The reproductive organs are susceptible to changes in homeostasis induced by organochlorines (Colborn et al. 1993, Damstra et al. 2002). The functioning of reproductive organs involves a complex interaction and timing between endocrine (hormonal) and immune parameters (Damstra et al. 2002). Impaired fertility has been associated with a negative impact from environmental anthropogenic organohalogen compounds in both male and female mammals. Among the effects noted are testicular dysfunctions, such as low sperm count and altered spermatozoa morphology in males, and pathological changes such as endometriosis, leiomyomas, occlusions and stenosis in the female reproductive tract (Bergman & Olsson 1985, Bergman 1999, Campagna 2001, 2002, Damstra et al. 2002). Such effects may lead to a reduction in the number of successful births which, if it occurs at high frequencies may have an effect at the population level. Therefore reproductive organs from 55 male and 44 female East Greenland polar bears were examined to investigate potential negative impacts from OHCs (**Paper 24**). Multiple regression analyses showed a significant inverse relationship between OHCs and testes length and baculum length/weight, respectively, in both subadults (DDTs, dieldrin, chlordanes, HCHs and PBDEs) and adults (dieldrin). Baculum BMD significantly decreased with increasing chlordanes, dieldrin, PCBs, PBDEs and HCB in both subadults and adults. In females, a significant inverse relationship was likewise found between ovary length and ΣPCB and ΣCHL, and between ovary weight and ΣHCB and uterine horn length and ΣHCB. Our study suggests that there is an impact from xenoendocrine contaminants on the size of East Greenland polar bear genitalia. To what extent these findings have had an effect on the East Greenland polar bear sperm and egg quality/quantity and uterus and penis size/robustness and hence the reproduction is uncertain. In a clinical survey on East Greenland male sledge dogs revealed a rare congenital malformation of the urethra and penis corresponding to severe perineal and penile hypospadias (Sonne et al. 2008b). Such malformation means that the animal wil not be able to reproduce themselves. It was suggested the congenital malformation was due to endocrine organ pathology/tumours of the dam, enzyme/receptor defects (mutation) in the pup or in uteri exposure to environmental xenoestrogens (Sonne et al. 2008b).

Photo 7. Sledge dogs having a similar food intake as the polar bears have been used in a controlled OHC exposure experiment as a substitute for polar bears in order to control exposure and obtain an unexposed control group. Photo: R. Dietz.

Pseudohermaphroditism

Wiig et al. (1998) reported on 4 pseudohermaphroditic female polar bears examined during live-capture for satellite tagging around Svalbard. It was suggested that the enlarged clitorises were congenital and could have been caused by an enzyme defect (21-hydroxylase deficiency), androgen producing tumour or a high exposure to organochlorines during the foetal stage or early development of the reproductive organs. Therefore, we collected polar bear sexual organs in order to contribute further information on this issue. Of the 44 sampled female specimens from 1999 to 2002, only one aberrant female was identified. This was a 23-year-old female polar bear killed in an Inuit hunt in East Greenland on July 9, 1999. The bear had a significantly enlarged clitoris resembling, in size, form and colour, those of previously reported pseudohermaphroditic polar bears from Svalbard (Wiig et al. 1998, Sonne et al. 2005b). Except for the enlarged clitoris, all dimensions of the external and internal reproductive organs of the bear were similar to a reference group of 23 normal adult female polar bears from East Greenland collected in 1999–2002. The aberrant bear was a female genotype, and macroscopic examination of her internal reproductive organs indicated that she was reproductively functional. A histological examination of the clitoral enlargement revealed intense chronic ulcerative and perivascular clitoriditis similar to acral lick dermatitis frequently seen in domestic dogs. In conclusion, therefore, we did not find any signs of pseudohermaphroditic hyperplasia of clitoral tissue due to androgenic or antiestrogenic endocrine disruption in this single individual. The levels of organohalogens and TEQ values were also lower than concentration thresholds of toxicological risk. We concluded that it is possible that some of the previously found adult female polar bear pseudohermaphrodites from Svalbard may have been misdiagnosed. Therefore, future studies examining pseudohermaphrodism in wildlife should consider that certain occurrences are natural events (Sonne et al. 2005b).

OHC effects on internal organs

OHCs and liver toxicity

In rats and mink, several studies have associated acute exposure to PCBs with liver toxicity (e.g. Jonsson et al. 1981, Bergman et al. 1992a, Kelly 1993, Chu et al. 1994, MacLachlan & Cullen 1995, Parkinson 1996). In marine wildlife, chronic exposure to OHCs, such as PCBs, DDTs, and PBDEs has been associated with toxic effects on several organ systems (Bergman & Olsson 1985, Schumacher et al. 1993, Bergman 1999, Bergman et al. 2001). In our work, we initiated an investigation into liver histology of East Greenland polar bears sampled during 1999–2002 (Sonne et al. 2005a). Light microscopic changes revealed nuclear displacement from the normal central cytoplasmic location in parenchymal cells, mononuclear cell infiltrations (12–16%), mild bile duct proliferation accompanied by fibrosis (8%), and fat accumulation in hepatocytes and pluripotent Ito cells (75–100%). For adult females, hepatocytic intracellular fat increased significantly with concentrations of the sum of hexachlorocyclohexanes, as was the case for lipid granulomas and hexachlorobenzene in adult males. Based on these relationships and the nature of the chronic inflammation, we suggested that these findings were caused by factors including long-term exposure to OHCs and recurring infections (Sonne et al. 2005a).

OHCs and renal toxicity

Studies of free-ranging grey seals and ringed seals from the Baltic Sea (Bergman et al. 2001) have shown an association between organochlorines and renal lesions. In dose-response and case-control experiments with OHCs, toxic effects on renal tissue have been found in rats (Bruckner et al. 1974, McCormack et al. 1978, Wade et al. 2002), bream fish (*Abramis brama*), and asp fish (*Aspius aspius*) (Koponen et al. 2001). We investigated the kidneys of East Greenland polar bears for toxic effects due to the high levels of OHCs in their adipose tissue (Sonne et al. 2006c). OHC concentrations and adverse effects on renal tissue in 75 polar bears collected during 1999 to 2002 were analysed. Specific lesions were diffuse glomerular capillary wall thickening (found in 22% of the animals examined), glomerular mesangial deposits (74%), tubular epithelial cell hyperplasia (21%), hyalinization of the tubular basement membrane, tubular dilatation, atrophy and necrosis (36%), tubular medullary hyaline casts (15%), interstitial fibrosis (30%), and mononuclear cell infiltration (51%). With the exception of mononuclear cell infiltrations, all these parameters were correlated with age, whereas none was associated with the sex of the animals. In an age-controlled statistical analysis of covariance, increases in glomerular mesangial deposits and interstitial fibrosis were significantly correlated with ΣPBDE concentrations in subadults. In adult males, statistically significant positive correlations were found for tubular epithelial cell hyperplasia and dieldrin concentration; diffuse glomerular capillary wall thickening and ΣCHL concentrations, and tubular medullary hyalin casts and ΣCHL, ΣPBDE, ΣPCB and ΣHCH. The lesions were consistent with those reported previously in highly OHC-contaminated Baltic seal populations and exposed laboratory animals. The renal lesions were also a result of aging. However, based on the above statistical findings as well as the nature of the findings, we suggest that long-term exposure to OHCs may be a cofactor in renal lesion occurrence, although other cofactors, such as exposure to heavy metals and recurrent infections from microorganisms, cannot be ruled out (Sonne et al. 2006c). In a recent controlled experiment on sledge dogs we likewise found significantly higher frequencies of glomerular, tubular and interstitial lesions in the exposed group. Furthermore, higher urine protein:creatinine ratio and plasma urea levels were found in the exposed group, which indicated a negative impact on kidney function via tubular and glomerular dysfunctions (Sonne et al. 2008a).

Immunological organs

It has been documented that high concentrations of PCBs and/or pesticides reduce specific lymphocyte function and thus may produce impaired resistance against infections in polar bears (Lie et al. 2005). In harbour porpoise some OHCs have a direct affect on the thymus, causing atrophy, and some affect the immune system by increasing lymphocyte depletion from lymphatic organs (Siebert et al. 2002). Exposure of mice to BDE-47 suppressed the proliferation of lymphocytes and the production of antibodies (Darnerud & Thuvander 1998, Thuvander & Darnerud 1999). Likewise thymotoxic effects occurred in mice exposed to BDE-71 (Fowles et al. 1994). Siebert et al. (2002) also found a relation between elevated concentrations of DDE and spleen depletion in harbour porpoise. In wildlife, histopathological changes in thyroid a.o. organs have been correlated to concentrations of OHCs in harbour seal, grey seal, ringed seal and harbour porpoise (Bergman & Olsson 1985, Schumacher et al. 1993, Bergman et al. 2001, Siebert et al. 2002). Therefore we also collected immunological organs from East Greenland polar bears, to the extent that these samples could be identified and provided by the East Greenland hunters. Samples of lymph nodes (axillary, n=54 and inguinal, n=45), spleen (n=60), thymus (n=11) and thyroid tissue (n=5) from a total of 82 polar bears from East Greenland 1999–2002 were examined histologically (Kirkegaard et al. 2005). High secondary follicle count was found in spleen (21%) and lymph nodes (20%), and this was significantly higher in subadults compared to adults of both sexes. Most of the correlations between concentrations of OHCs and the amount of secondary follicles in lymph nodes were insignificant, but ΣPBDE showed a significant, but modest positive correlation. In spleen, a significant relation between low concentrations of OHCs in adipose tissue and few/absent secondary follicles was found with respect to ΣCHLs, ΣHCHs, HCB and dieldrin. No histopathological observations (e.g. neoplasia) were found in spleen, lymph nodes, thymus or thyroid. In conclusion, the results of Kirkegaard et al. (2005) suggested that based on the available samples exposure of polar bears to OHCs was unlikely to have resulted in adverse effects on the tissues in question, although ΣCHLs, ΣHCHs, HCB and dieldrin were related to increased secondary follicle counts in the spleen.

OHC effects on skeletal system

OHCs and Bone Mineral Density

Bone mineral composition in mammals is based on a complex set of interrelated mechanisms and is influenced by various nutritional and environmental factors (e.g. Ganong 1991, Sarazin et al. 2000, Johansson & Melhus 2001, Leder et al. 2001, Johansson et al. 2002, Promislow et al. 2002 & Michaelsson et al. 2003). In marine mammals such as grey seal (*Halichoerus grypus*), ringed seal, harbour seal (*Phoca vitulina*), and in a reptile, the alligator (*Alligator mississippiensis*), osteopenia and macroscopic pathology have been examined in bone during distinct periods of exposure to anthropogenic pollutants (Zakharov & Yablokov 1990, Bergman et al. 1992b, Mortensen et al. 1992, Schandorff 1997, Sonne-Hansen et al. 2002, Lind et al. 2003, 2004). The studies showed relationships between OHCs and exostosis, periodontitis, loss of alveolar bone structures, osteoporosis, widening of the canine opening, and enlargement of the foramen mentalia. These conditions prompted us to analyse bone mineral density (BMD) in skulls of polar bears from East Greenland sampled during 1892–2002 (**Paper 21**). Our primary goal was to detect possible changes in bone mineral content due to elevated exposure to OHC. BMD in skulls sampled in the period of introduction in nature of OHC (1966–2002) turned out to be significantly lower than in skulls sampled in the pre-OHC period (1892–1932) for subadult females, subadult males, and adult males but not adult females (Fig. 27; **Paper 21**). In addition we found a negative correlation between organochlorines and skull BMD for ΣPCBs and ΣCHL in subadults and for dieldrin and ΣDDT in adult males. For ΣPBDE in subadults, an indication of a relationship was detected (p = 0.06). We therefore concluded that disruption of the bone mineral composition in East Greenland polar bears may have been caused by organochlorine exposure (**Paper 21**).

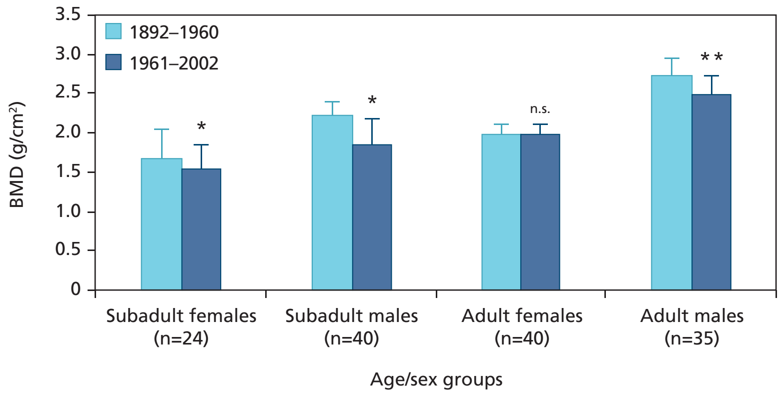

Fig. 27. BMD (g/cm^2) in skulls from East Greenland polar bears compared between 1892–1992 and 1961–2002 four age and sex groups (modified from **Paper 21**). Asterisks indicates significant (*:p ≤ 0.05 and **: p ≤ 0.01) differences and n.s. are non significant.

Gross skull pathology

Laboratory studies have shown that organochlorines induce periodontitis in mink (*Mustela vison*) (Render et al. 2000a, b, 2001), and in humans PCB seems to interfere with normal teeth outbreak (Rogan 1979, Miller 1985, Gladen et al. 1990). In various studies of wildlife including marine mammals, relationships between exposure to organochlorines and exostosis, periodontitis, osteoporosis and widening of canine alveoli have been documented (Zakharov & Yablokov 1990, Bergman et al. 1992, Mortensen et al. 1992, De Guise et al. 1995, Schandorff 1997). To investigate possible negative health impacts in regions of the polar bear range with highest exposures, a time-trend study of skull pathology was conducted on East Greenland and Svalbard polar bears sampled during 1892–2002 (Sonne et al. 2007a). Of seven different pathological changes, only tooth wear and periodontitis was in a prevalence that allowed statistical treatment. In East Greenland, the prevalence of tooth wear was significantly higher in polar bears collected in the pre-contamination period than in bears sampled during periods after introduction of and contamination of the environment by OHCs. Considering periodontitis, prevalence was not significantly different between pre-contamination and contamination periods. Polar bears from Svalbard had significantly higher prevalence of tooth wear and periodontitis than polar bears from East Greenland. Hence, we found a clear geographical difference, but no evidence for an association between skull pathology and exposure to organochlorines in East Greenland and Svalbard polar bears (Sonne et al. 2007a).

Fluctuating asymmetry

Interference of OHCs with receptors in the main endocrine pathway results in endocrine disruption and stress. This will lead to elevated blood corticosteroid levels that may induce fluctuating asymmetry (FA) (Bergman & Olsson 1985, Colborn et al. 1993, Feldman 1995, Borisov et al. 1997, de March et al. 1998, Bergman 1999, Damstra et al. 2002). Sonne et al. (2005c) therefore investigated FA in skulls of 283 polar bears sampled in East Greenland from 1892 to 2002. Thirteen useful metric bilateral traits in skull and lower jaw were measured and compared between polar bears born before 1960 (pre OHC period; n=94) and after 1961 (OHC period; n=189). The degree of fluctuating asymmetry did not differ statistically between the two periods in 10 of the 13 traits. In fact, when significant differences were found in four of the traits, the fluctuating asymmetry was lower in skulls sampled after 1960. A time trend analysis did find fluctuations over time for five traits, but the relationship was weak as the trend appeared to occur by chance due to the high number of regressions analysed (n=42). A correlation analysis of FA versus the sum concentrations of various classes of OHCs in adipose tissue from a subsample of 94 recently collected polar bears (1999–2002) did not show a trend either. Hence, this study could not document any relationship between skull asymmetry in polar bears and periods with different exposure to organohalogens (Sonne et al. 2005c). We therefore concluded that the differences were likely to be influenced by nutritional status, genetic factors, a sub-effect exposure to organohalogens or other confounding environmental factors such as temperature differences within the two investigated periods. In a recent paper Bechschøft et al. (in press) investigated eight bilateral traits from East Greenland and Svalbard with respect to trends from 1950 to 2004. Three out of 24 combinations of groups (subadults, adult female and adult males) and traits showed significant negative slope. The general decrease in FA during 1950–2000 may be explained by the general declining organohalogen concentrations found within the same period. Indications were thus found for a linkage between FA and organohalogen pollution.

OHC effects on immune response

Studies by Bernhoft et al. (2000) and Lie et al. (2004, 2005) have indicated that both serum immunoglobulin G (IgG) level, humoral (antibody response following immunization), and cellular immunity (antigen and mitogen induced lymphocyte proliferation) may be impaired by OHCs in the Svalbard subpopulation of polar bears.

In vivo studies of harbour seals fed contaminated Baltic fish likewise showed that OHCs affect humoral (antibody response) and cell-mediated (lymphocyte proliferation) immunity (De Swart et al. 1994, 1995, Ross et al. 1995, 1996a, b, c). In free-ranging species, OHC immunotoxic effects, through mitogen-induced lymphocyte response and IgG concentration, have been suggested in bottlenose dolphins (*Tursiops truncatus*) (Lahvis et al. 1995), striped dolphins (*Stenella coeruleoalba*), and harbor seal (Troisi et al. 2001) and in the St. Lawrence beluga whale (Martineau et al. 1994, De Guise et al. 1995, 1998). A connection between environmental organochlorine exposure and immunosuppression has likewise been suggested for humans (Dewailly et al. 2000, Morein et al. 2002). A few studies conducted in Nunavut supports the hypothesis that the high incidence of infections observed in Inuit children (mostly respiratory infections and acute otitis media) is due in part to high prenatal exposure to OHCs (Dewailly et al. 2000, 2001b). To determine if immunotoxicity from a typical Greenlandic natural intake of OHCs from marine mammal blubber showed a true cause-effect relationship, we conducted a controlled study on domestic West Greenland sledge dogs, these being a phylogenetically relevant substitute for the polar bear (**Paper 27**). The exposed groups were fed a diet of minke whale blubber rich in Hg, OHCs, and n-3 fatty acids, with exposure levels similar to those of Inuits and polar bears, while the control group was fed uncontaminated pork fat. The immune response after mitogen and antigen stimulation was measured using an intradermal test (IDT). The study documented that a daily intake of 50–200 g of minke whale blubber caused an impairment of the nonspecific and specific cellular immune system in the sledge dogs. Immune reactions were measured by mitogen (PHA, Con A) and antigen (KLH) intradermal testing (Fig. 28). This information together with information from the literature cited above makes it likely that Inuits and polar bears suffer from similar decreased resistance to diseases by a comparable intake of marine

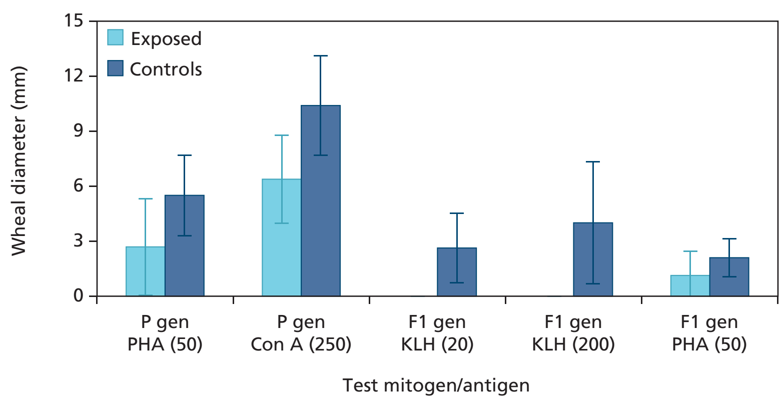

Fig. 28. Bar diagram of the significant lower intradermal reactions of exposed versus controls sledge dog bitches (P generation; n = 16) and their pups (F1 generation; n = 9) for mitogens (PHA and Con A) and antigen (KLH) and their respective concentrations (μg/ml) (data from **Paper 27**).

mammal blubber. Our study also suggested that the fatty acid composition should be taken into consideration when investigating combined immunotoxic effects of contaminated food resources in future Inuit and polar bear studies.

Part conclusion on effects of OHCs in Greenland top predators

Reduced size of reproductive organs was found in both male and female polar bears, associated with increased OHC concentrations. However, previous observation on pseudohermaphroditism in female polar bears from Svalbard could not be verified from examination of a single animal from East Greenland with an enlarged clitoris. Tissue alterations were found in liver and kidney, which could be linked to certain OHCs. However, this was not the case for immunological organs such as lymph nodes, spleen, thymus and thyroid tissue. Studies on effects on the skeletal system in East Greenland polar bears documented a reduction in bone mineral density associated with OHC exposure. However, no relationship was found between skull pathology and organohalogens. Fluctuating asymmetry in polar bears showed variable results dependant on the analyhical method used. Some of the lacking skeletal effects were probably due to subeffect exposure to OHCs, influence of nutritional status, genetic factors or other confounding environmental factors such as climate change. A daily intake of amounts of 50–200 g marine mammal blubber from Greenland is likely to cause an impairment of the immune system in top predators.

Contaminants and mass mortality epizootics among Arctic and European mammals

As discussed above, the immune system can be affected by high concentrations of OHCs. PDV has been responsible for a large number of deaths among marine mammals and is the best studied disease in marine mammals. We focussed on this highly virulent disease to consider further whether it has been possible to link the severity of this disease (the infection rate of PDV) to the exposure to OHCs.

A number of diseases other than PDV can be found in marine mammals that can affect their health and the population size, and in some cases also the health of Inuit that consume marine mammals. These include trichinosis (Born et al. 1982, Born & Henriksen 1990), toxoplasmosis (Mcdonald et al. 1990, Rah et al. 2005, Sørensen et al. 2005), brucellosis (Nielsen 2001, Tryland et al. 2001, Dubey et al. 2003, Tryland et al. 2005) or calicivirus, phocid herpesvirus, rabies virus, and influenza A virus (Smith et al. 1973, Osterhaus et al. 1985, Ødegård & Krogsrud 1981, Loewen et al. 1990, Taylor et al. 1991, Johnston & Fong 1992, Prestrud et al. 1992, Stuen et al. 1994, Zarnke et al. 1997, Lenghaus et al. 2001, Martina et al. 2003, Ganova-Raeva et al. 2004). Since no information on these diseases in relation to contaminants is available and, to date, we have only initiated and not published results on some of these diseases, these diseases are not further discussed in this dissertation. Samples collected for contaminant studies however do provide the opportunity to study other health aspects and disease patterns in the monitored animals. In this connection samples are stored in specimen's bank and hence renewed and expensive sampling can be avoided.

Effects and spreading of PDV

The potent and fairly widely distributed disease Phocine Distemper Virus (PDV) has probably been circulating in the Arctic for many centuries without being diagnosed prior to the first recorded outbreak of PDV in Europe in 1988 (**Paper 1, 2**, Heide-Jørgensen et al. 1992). The total PDV mortality in Europe exceeded 18 000–23 000 harbour seals in 1998, and was approximately 31 000 seals in 2002 (e.g. **Paper 2**, Heide-Jørgensen et al. 1992, **Paper 26**). Mass mortality events have previously been recorded in Cape fur seals in the beginning of the 19th century, harbour seals (> 1 000) in Icelandic waters in 1918, crabeater seals at the Antarctic (>3 000) in 1955, and walruses (ca. 1 200) in the Bering Strait in 1978 (see reviews in **Paper 2**, Heide-Jørgensen et al. 1992, **Paper 26**). None of these outbreaks are well described and the cause of deaths can therefore not be determined with any degree of certainty. The first well described outbreak occurred among harbour seals in New England in 1979–1980 where at least 500 seals died from an influenza-A type virus (Geraci et al. 1982).

The origin of the 1988 epizootic

Tissue samples taken from ringed and harp seals in Greenland for contaminant analysis prior to the outbreak in 1988 provided the first clue to the Arctic origin of distemper virus (**Paper 1**). A main hypothesis for the source of the 1988 epizootic is that harp seals (*Phoca hispida*) acted as the primary vector of the PDV (**Paper 2**, Henderson et al. 1992, Markussen & Have 1992). Support for this hypothesis was provided by records of mass migrations of harp seals into the southern Norwegian, Danish and Swedish waters in the winter of 1987–1988 (**Paper 1, 2**, Heide-Jørgensen et al. 1992, Markussen & Have 1992). During this migration 77 000 harp seals died in nets along the coast of Norway (Haug et al. 1991). The likely reason for this exodus was the collapse of the capelin (*Mallotus villosus*) stock in the Barents Sea (Haug et al. 1991).

Photo 8. The harp seal was most likely the primary source of PDV in 1988 due to the detected anti-body pattern in this species and the large number of harp seals observed migrating southward to lower latitudes prior to the outbreak. Photo: R. Dietz.

The origin of the 2002 epizootic

Over the years following the 1988 epizootic there were no signs that the PDV had been circulating among European harbour seals (Jensen et al. 2002, Thompson et al. 2002), and thus the new outbreak in 2002 was most likely the result of a cross-species infection. PDV has continued to circulate in the Arctic and some infectious Arctic species may have brought the disease to European waters again (**Paper 26**). However, since there were no signs of harp seals in the North Sea area prior to the 2002 PDV outbreak, the primary vector may have been another seal species (**Paper 26**). One of the possible carriers of the PDV disease is the grey seal and a number of features identifying this species as a possible carrier are discussed in Härkönen et al. (**Paper 26**).

PDV in the Arctic

Mass mortalities in the Arctic have never been observed, but there are several indications for the presence of morbilliviruses from the Arctic. Tests of PDV- and CDV-neutralizing antibodies in various pinniped samples collected prior to the 1988 PDV outbreak revealed that morbilliviruses were common among pinnipeds in the Arctic regions. PDV and CDV antibodies were detected in archived harp seal samples collected prior to the 1988 outbreak from Canadian and Greenlandic waters, the West Ice, and the Barents Sea (**Paper 1**, Markussen & Have 1992, Henderson et al. 1992, Duignan et al. 1997). Other species of Atlantic pinnipeds had also been exposed to morbilliviruses both prior to and after 1988. Among these were ringed seals in Canada and Greenland (**Paper 1**, Henderson et al. 1992, Duignan et al. 1997), and harbour seals, grey seals, hooded seals (*Cystophora cristata*), and walruses from the American and Canadian Atlantic coast (Henderson et al. 1992, Duignan et al. 1994, 1995a, 1997, Nielsen et al. 2000). With the exception of a suggested PDV outbreak in harbour seals along the Northeast coast of United States in the winter 1991–1992 (Duignan et al. 1993, 1995a), no elevated mortality has been reported in these species outside Europe. In contrast to the European Arctic, no antibodies to PDV were detected in ringed seals, spotted seal (*Phoca largha*), ribbon seal (*Phoca fasciata*), Steller sea lions (*Eumatopius jubatus*), bearded seal (*Erignathus barbatus*) and walrus from the northern Pacific (Osterhaus et al. 1988). Morbillivirus antibodies have been detected in polar bears from Alaska and Russia with prevalences ranging from 26% to 46% between different years (Follmann et al. 1996).

The reason why mass mortalities from PDV have not been registered in the Arctic is probably linked to frequent re-infection leaving little or no space for an infectious naive population in which mass mortalities can appear. The fact that a potent virus such as PDV is circulating in the Arctic without providing any detectable die off is relevant in the discussion on whether diseased or dead animals originating from exposure to contaminants are likely to be encountered in the Arctic by hunters or scientists. At present, we are investigating the recent development of PDV in ringed seals as well as the potential transference of the disease to polar bears in East Greenland.

PDV and OHCs

The geographical pattern of the PDV mortality could, at a first glance agrees with the hypothesis that OHCs may be linked to the effect and spreading of PDV. No mass mortality has been observed in the Arctic where OHC concentrations are lower than in the Northern Europe, where PDV struck hardest. Also PDV struck harder in the Kattegat region, close to the heavily polluted Baltic, than around the British Isles, where OHC exposure is lower. The role of OHC contamination, through impeding immune system function, was therefore considered an implicating factor in the 1988 epizootic, but no causal relation could be established (Hall et al. 1992a, b, Reijnders & Aguilar 2002). The fact that the rate of population increase (a function of fecundity and survival) in the last decade before the 2002 virus outbreak was close to the maximum possible, indicates that the immune system of the seals in, e.g. the Wadden Sea was not severely impeded between the two epizootics. No detailed study including OHC analysis from all regions during the two outbreaks has been carried out. PCBs and DDE levels in blubber from adult seals collected in 2002 in the heavily polluted part of the western Dutch Wadden Sea, decreased by 65% and 50%, respectively, compared to 1988 (Aguilar et al. 2002, Reijnders & Simmonds 2003). The fact that no clear decrease of the mortality appeared between the two outbreaks, that the virus was not recorded in European waters prior to 1988 when OHC levels were higher, and finally that no major mortality was observed in the Baltic, where OHC have been the highest, makes it unlikely that contamination with the immuno-suppressive OHCs has played a major critical role in the seal epizootic in the North Sea and Kattegat/ Skagerrak in the two outbreaks (**Paper 26**). Also the virus appears to have originated from, and has circulated in the Arctic, where OHC exposure is lower. A key question in understanding the latent risk of any novel disease is why some introductions of pathogens cause epidemic out-breaks, while in other cases the disease fades out. Using a novel method, Harding et al. (submitted) estimated the basic reproductive number (R0) for many different subpopulations from the two outbreaks of PDV in European harbour seals in 1988 and 2002. Interestingly, values of R0

Photo 9. The Phocine Distemper Virus outbreaks exterminating more than 22 000 seals in 1988 and 32,000 seals in 2002 in Northern Europe are the largest mass mortalities observed among marine mammals. Photo: R. Dietz.

and the mortality varied greatly among su populations, and three factors were found correlate with this variation by modifyin the "functional contact rate" namely the local population growth rates prior to the disease outbreak, the degree of metapopulation structure, and the time of infection of local groups. These parameters, previous contact to PDV and other factors can confound the linkage between immune suppression and OHC loads.

Part conclusion on PDV and contaminants
Even for incidents such as PDV outbreaks, whe considerable effort has been expended on the dete tion of dead animals and investigation of popul tion effects, it has not been possible to make a cle linkage between contaminants, immune suppre sion and number of deaths caused by the disease. A large number of confounding factors play a major role for such disease events. The epidemiology of PDV in European seals indicates that previous contact to PDV, local population growth rates, metapopulation structure, and seasonality of the infection has a major influence on the severity with which the pathogen spreads.

Marine mammal migration and stock separations

Information on migration and stock separation of marine mammals is of major importance when studying contaminants. It is important to know 1) in which areas the animals obtain their contaminant burdens during different seasons, 2) if the different species can be divided into separate stocks with potentially different contaminant exposure due to differences in feeding as well as sources and transport of contaminants. If information on 1 & 2 are available this needs to be taken into account when designing monitoring programmes and making conclusions about geographical or temporal trends in contaminant exposure.

Work on marine mammal migration and stock separations are driven by many different questions, and several scientific disciplines can be used to answer these questions. The methods most frequently used are visualised in Fig. 29.

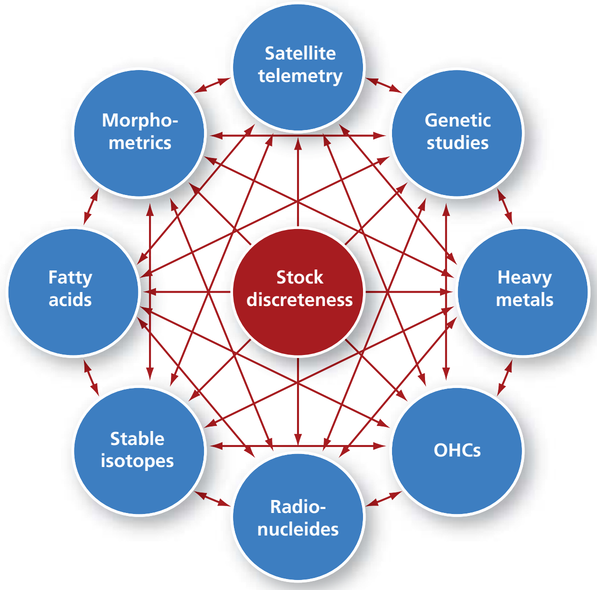

Fig. 29. Overview of disciplines used to reveal stock discreteness.

Ideally, information on population structure, distribution and seasonal migration patterns should be known for all migratory species before conducting investigations on contaminants, but this criterion is rarely fulfilled. One of the strongest tools for obtaining information on population structure is satellite telemetry; however, for some species and areas this information is not easy to obtain and, if possible this approach is supplemented by genetic studies on samples taken during tagging, through biopsy sampling or from the native hunt. Earlier, morphometric studies have also been used, but today a number of additional methods can be used to further supplement genetic analysis, including analyses for contaminants (heavy metals, OHCs and radionuclides), stable isotope and fatty acids.

It is generally accepted that satellite telemetry is a robust tool providing results which are easy to interpret. Therefore this approach is dealt with in greater detail, including discussion of papers from this dissertation. Information from studies of genetics, contaminants, stable isotopes and fatty acids relative to the stock separation issue has received less focus in my work, and this is reflected in the less detailed description, and fewer papers co-authored and selected for the thesis on these subjects.

Satellite tracking

Satellite tracking has proven to be a very reliable technique for many species, providing information that can be used elucidate the answers to a number of questions. One basic subject that telemetry can provide very relevant information on is the identification of different marine mammal stocks. Stock information is important for the monitoring of contaminants, but is also essential for the management of hunted stocks. Linking stock distribution and migration patterns to various physical parameters (e.g., ice cover, bathymetry, land barriers, food availability) increases the understanding of habitat selection, which in turn can be used to identify critical habitats (Fig. 30). Understanding of critical habitats can then be used to reduce impacts that, in addition to hunting, can result from conflicts with human activities such as resource exploitation, fishery interaction or disturbance from shipping or other noise emitting activities.

The broad community involved in satellite telemetry facilitates collaboration among institutions and funding programs over time, and also the cooperation between regions that is necessary to get a thorough picture of stock migrations and related management questions.

Fig. 30. Examples of questions that satellite telemetry can provide useful information to solve.

Information from studies of some key Arctic species of importance for our contaminant work is provided in the following.

Polar bears

The polar bear is probably the best studied species throughout the Arctic, and much of this work is based on satellite telemetry. For many years it has been possible to track polar bears year-round, and thereby to obtain information to subdivide polar bear populations into subpopulations or management units. Such information has been used in the design of studies on meta-analysis on Arctic spatial trends of heavy metals and OHCs (e.g. Born et al. 1991, Norstrom et al. 1998, **Paper 12, 16**, Smithwick et al. 2005b, Verrault et al. 2005, Muir et al. 2006). The circum-Arctic polar bear population/management areas, several of which are based on cluster analysis of satellite-tracked polar bears, are shown in Fig. 31 (Derocher et al. 1998).

For areas bordering Greenland, satellite tagging has been used to differentiate Kane Basin and Baffin Bay polar bears in Northwest Greenland (Taylor et al. 2001, **Paper 16**), and east of Greenland to distinguish the Greenland stock from Svalbard bears (Born et al. 1997b, Wiig et al. 1995, 2003). Substantial amount of information on contaminants and their effects have been obtained in connection with the telemetry work on polar bears. Many of the biopsies and blood samples for contaminant studies have been obtained from the polar bear satellite or conventional tagging programmes, especially in areas like Svalbard where the polar bear is protected, or in places where no hunt is taking place for other reasons. The extensive Norwegian OHC effect studies have been based on adipose tissue biopsies, blood analysis and vaccination programmes conducted in connection with tagging programmes carried out within the last two decades (e.g. Oskam et al. 2003, 2004, Braathen et al. 2004, Lie et al. 2004, 2005). Another example is the survey of denning behaviour in relation to contaminant exposure conducted by Skaare et al. (1994). In that study, no relationship could be detected between Svalbard female polar bear reproductive success and PCB levels, possibly due to

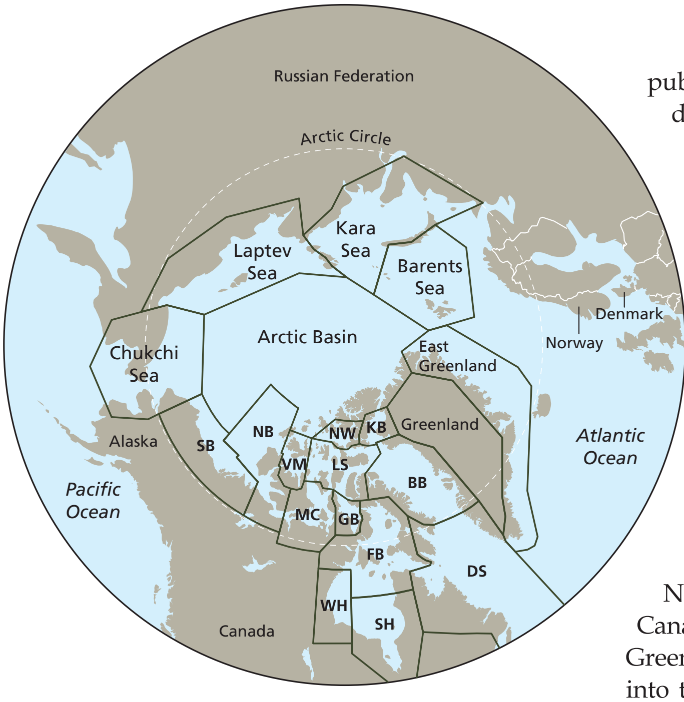

Fig. 31. Distribution of polar bear populations throughout the circumpolar basin (Derocher et al. 1998).

the limited number of bears investigated (n=10). Space-utilisation was examined for 54 female polar bears from Svalbard and the Barents Sea that were collared with satellite transmitters to provide information on their spatial positions and annual home range sizes (Olsen et al. 2003). Among the tested variables, annual home range size was the variable that affected ΣPCB5 (sum of PCB-99, -153, -156, -180, and -194) to the largest degree. Olsen et al. (2003) proposed that the positive correlation of home range size with ΣPCB5 in female polar bears was related to the higher energetic costs required, as polar bears with large home range sizes would need to consume more prey than bears with smaller home range sizes. Polar bears with large home range sizes were also more pelagic, inhabiting areas further east, closer to the ice-edge zone than animals with small home range sizes. Thus, prey choice associated with a pelagic space-use strategy may also explain the higher ΣPCB5 in polar bears with large home range sizes.

Narwhal

Narwhals have been studied for their contaminant loads but most of this information was published before satellite telemetry revealed differences in the sub-populations (Wagemann et al. 1983, Wagemann & Muir 1984, Hansen et al. 1990, Muir et al. 1992, Wagemann et al. 1996, Dietz et al. 1997b, **Paper 12**). As no hunting takes place at Svalbard, the only OHC data on narwhals has been based on blubber biopsies of 3 sub-adult narwhals tagged with satellite transmitters in 1998, (Wolkers et al. 2006a, Lydersen et al. 2007). Information on stock discreteness obtained from satellite telemetry from the region between Greenland and Canada has accumulated over the past 15 years.

Narwhals are dispersed from the central Canadian High Arctic over West and East Greenland to Svalbard, Franz Joseph land and into the polar Basin North of Russia (Hay & Mansfield 1989, Reeves et al. 1994, **Paper 7**). Although the narwhals are geographically dispersed during the summer in Greenland and Canada they are forced southward into Baffin Bay during winter, where several stocks show overlapping distribution based on their winter home-ranges (Fig. 32) (**Paper 9, 18**, Heide-Jørgensen et al. 2002, 2003b, **Paper 30**). The relatively restricted winter distribution means that their geographical exposure to food and contaminants from the same feeding grounds is similar during this time of the year. Despite the fact that food items are found in the stomachs of narwhals during summer, it has been documented that narwhals feed more during winter and may be competing for the food (Greenland halibut, *Reinhardtius hippoglossoides* and *Gonatus fabricii*) with other narwhal populations (Heide-Jørgensen et al. 1994b, Laidre & Heide-Jørgensen 2005). These similarities in winter distribution and feeding preferences may explain why no clear geographical patterns in contaminants are found from studies in the various sampling areas (**Paper 19**). Also narwhal obtained along the eastern part of Baffin Island from Pond Inlet, Clyde River, Broughton Island during autumn, winter or spring are likely to be from either the Eclipse Sound or Admiralty Inlet summering stocks (**Paper 30**). Thus, the samples obtained outside the summering months from July to mid- September are hard to separate.

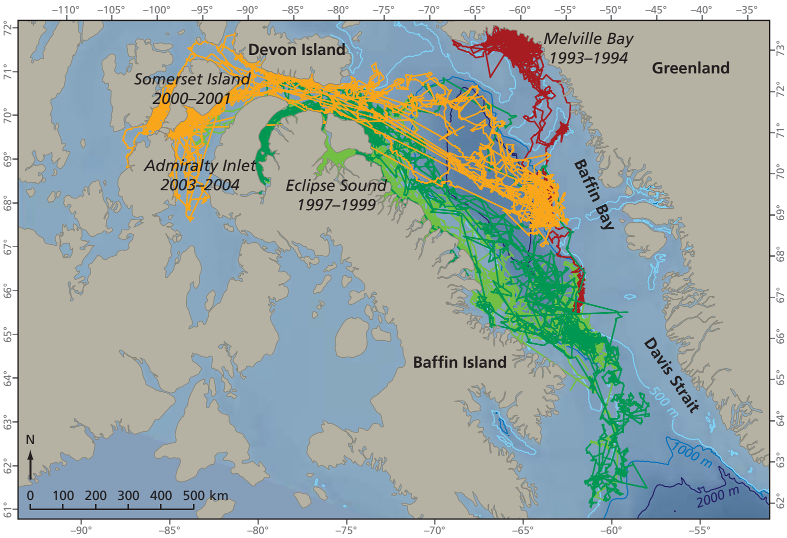

Fig. 32. The migration routes and summer and winter kernel home ranges of 88 narwhals tagged by satellite transmitters over 9 years from 4 summering cites: Red: Melville Bay (1993–1994), Light green: Tremblay Sound (1997–1999), Yellow: Creswell Bay (2000–2001) and Dark Green: Admiralty Inlet (2003–2004) (based on information from **Paper 9, 18**, Heide-Jørgensen et al. 2002, 2003b, **Paper 30**).

So far, this information has not been used in the planning of meta-contaminant analysis on narwhals. However, contaminant analysis, genetic and stable isotope analyses have been used to fill out some of the lacking information on narwhal stock relationships (see sections below). The telemetry work has also provided information on diving capabilities relevant for understanding feeding behaviour and distribution patterns linked to major prey items and a number of physical parameters such as bathymetry, ice and temperature (e.g. Heide-Jørgensen & Dietz 1995, Heide-Jørgensen et al. 2001, Laidre et al. 2002, 2003, 2004a, 2004b, Stern & Heide-Jørgensen 2003, Heide-Jørgensen & Laidre 2004, Dietz et al. 2007b). These studies have shown where narwhals seek their prey during winter, and with the information on feeding, biomagnification factors can be estimated between narwhals and their dominant prey when these are collected and analysed (e.g. **Paper 12**). If ice extent alters dramatically as a result of climate change, the narwhal distribution pattern and thus the regions where they are exposed to contaminants over the course of the year may change as well; this may also be true for some of their prey dispersal and composition as well.

Photo 10. Narwhals have been tagged with satellite transmitters since 1993 and information on many subpopulations has been obtained. Some individuals have been instrumented with other tags to obtain supplementary information on narwhal diving, feeding strategies and use of sound. Photo: R. Dietz.

Beluga whale

Belugas are distributed throughout the Arctic with a broader range than the narwhal as their distribution also extends into the Beaufort Sea, the East Siberian Sea, the Chukchi Sea and even extends down into the Northern Pacific (Brodie 1989). In the Northwest Atlantic, the beluga is also present as far south as the St. Lawrence River, Canada. Satellite tracking of beluga whales has been conducted in Canada/Greenland, Alaska, and Svalbard. Information is only included here from those studies which have importance in relation to the Greenland winter populations or contaminant work. Telemetry results are discussed with reference to genetic and contaminant comparisons in sections below.

Considerable effort has been applied to study the linkage between belugas in Greenland and Canada, to evaluate stock relations primarily in connection with hunting regulations, but this information can also be used to understand where the belugas obtain their contaminant exposure over the year. The satellite tracks presented by Heide-Jørgensen et al. (2003a) provided direct evidence that there is a link between the belugas summering in the Canadian High Artic and those in the West Greenland wintering area, which may not be too surprising as belugas are not present in Greenland waters during summer (Brodie 1989, Heide-Jørgensen et al. 1993, Heide-Jørgensen 1994). Satellite tracking provides an opportunity for direct estimation of movement probability, and pooling tracks from several years and localities to increase sample size.

The percentage of satellite tracked belugas moving to Greenland has varied between 0 and 60% dependent on year and tracking site. On average, from all years we estimated that 15% (4/26) of the belugas from the Canadian High Arctic moved to West Greenland (Richard et al. 1998a, 2001, Heide-Jørgensen et al. 2003a). Using this distribution on the population estimate, the resulting abundance estimates would be ca. 4 400 belugas wintering in West Greenland and 16 800 belugas wintering in the North Water and adjacent areas (Heide-Jørgensen et al. 2003a). These estimates do not agree with aerial surveys conducted in the North Water, where Finley & Renaud (1980) and Richard et al. (1998b) gave counts of approximately 500 whales at the surface in leads and cracks in the North Water in winter 1978 and 1994, respectively. If these figures were corrected for whales that were submerged and overlooked by the observers, the figures could maybe be as high as 3 000, but still far from the estimate of 16 800 (Heide-Jørgensen et al. 2003b). This variability stresses the need for additional studies to elucidate the inter-annual and inter-regional differences in beluga movement probability.

At Svalbard, blubber samples taken during satellite tracking operations have been used to evaluate contaminant burden in the protected stock of this region (Andersen et al. 2001, 2006, Wolkers et al. 2006a). A trans-Arctic contaminant study has been suggested for the IPY beluga programme PATOB (Pan-Arctic Tracking of Beluga Whales) that could provide an excellent tissue sampling opportunity for contaminant and genetic investigations. Also the relationship between Canada and Greenland stocks documented by satellite telemetry has been used in a long-time trend Hg IPY programme proposal, where historic samples from Canadian and Greenlandic beluga teeth collections will be compared. St. Aubin et al. (2001) investigated blood clinical-chemical parameters in 55 belugas obtained during the satellite tagging operations. Two belugas recaptured 19 and 24 days after instrumentation showed changes in leucocyte counts, hematocrit, and a variety of plasma chemical constituents, some of which indicate inflammation and a likely physiological response to handling and tagging stresses (St. Aubin at al. 2001). Most of the contaminant work carried out on belugas has been done on animals sampled from the aboriginal hunt in Alaska, Canada and Greenland. For the more contaminant exposed St. Lawrence belugas, the samples have primarily been collected from animals found dead (e.g. Martineau et al. 1994, De Guise et al. 1995, 1998). In the case of St. Lawrence belugas, this population is simply too small to allow for satellite tagging work to be combined with contaminants.

Ringed seals

Ringed seal are distributed throughout the entire Arctic and are a very important resource for the Inuit communities (Reeves 1998). For this reason ringed seal has been selected as an essential species to be monitored in the AMAP programme. Ringed seals have generally been regarded as being relatively stationary, but many recent studies have shown that especial-

Photo 11. Walruses from West Greenland have been tracked to Southeast Baffin Island, where they haul out on the rocky shores during late summer and autumn, when the ice has disappeared from the region. The walruses from these two regions are hence likely to be a common population. Photo: R. Dietz.

ly the younger ringed seals make considerable migrations. Satellite telemetry, conventional tagging and genetic studies have been used to provide information on site fidelity of the ringed seals. In Greenland, ringed seals have only been tagged in the Avanersuaq area in NW Greenland (Heide-Jørgensen et al. 1992, Teilmann et al. 1999, Born et al. 2002a, 2004). These studies have shown that there is contact with Arctic Canada and tags have been recovered as far south as Disko Island (Kapel et al. 1998). Little is known on movements from other areas of Greenland, but results from conventional taggings have revealed connections between other regions, such as Kong Oscars and Scoresby Sound Fjords (Kapel et al. 1998). This shows that at least some of the ringed seals sampled in various areas and analysed for contaminants may be receive their contaminant exposure in other regions. Ideally, knowledge of ringed seal dispersal should be obtained for all monitored areas, before ringed seals are compared for contaminants around the entire Arctic, but a lot of knowledge is still lacking on ringed seal migration patterns. An ongoing genetic study of ringed seals may be the first analysis to shed light on the population differences around most of Greenland (see genetic section and Rew et al. in press).

Walrus

Walruses presently have a disjunct Holarctic distribution with the widest gap of ca. 500 km. Three subspecies are recognized: the Atlantic walrus (*Odobenus rosmarus rosmarus*), the Pacific walrus (*Odobenus rosmarus divergens*) and the Laptev Sea walrus (*Odobenus rosmarus laptevi*) (Born 2005).

Tagging of walruses is a particular challenge and most results have been obtained from animals tagged on land. In Canada, only short term local movements have been obtained from walrus in the Canadian High North (Stewart pers. comm.). Most Greenland information on walrus movements from satellite tracking is from the National Park in Northeast Greenland, where no hunting is taking place (Born & Knutsen 1992, 1997, Born et al. 1997a, 2005). In the Baffin Bay region, where most walrus are hunted and information on stock assessment is of major importance, satellite tagging has just recently been initiated. Remote deployment techniques have even made it possible to tag walruses in the pack ice, though the tags do not last much longer than one month on average (Jay et al. 2006, Dietz & Born unpublished). Ongoing investigations have revealed a connection between central West Greenland and Southeast Baffin Island, Canada, indicating that walruses from these regions are receiving the same contaminant exposure (see Fig. 33).

At Svalbard, walruses have also been successfully tagged and in this region samples were also analyzed for contaminants (e.g. Wiig et al. 1993, Gjertz et al. 2001, Wolkers et al. 2006b).

Other marine mammals

Results from other Greenland marine mammals continue to accumulate for other marine mammals, including harp seals, hooded seals, bowhead whales, humpback whales, minke whales and blue whales, with the primary goal of these investigations being stock management and protection, understanding the distribution and habitat preferences in relation to resource exploration and exploitation, and understanding the changing ecosystem relative to a changing climate. Some of these studies have been linked to genetic sampling and analysis, but no dedicated effort has been addressed towards contaminants thus far, and therefore these studies are not considered further here.

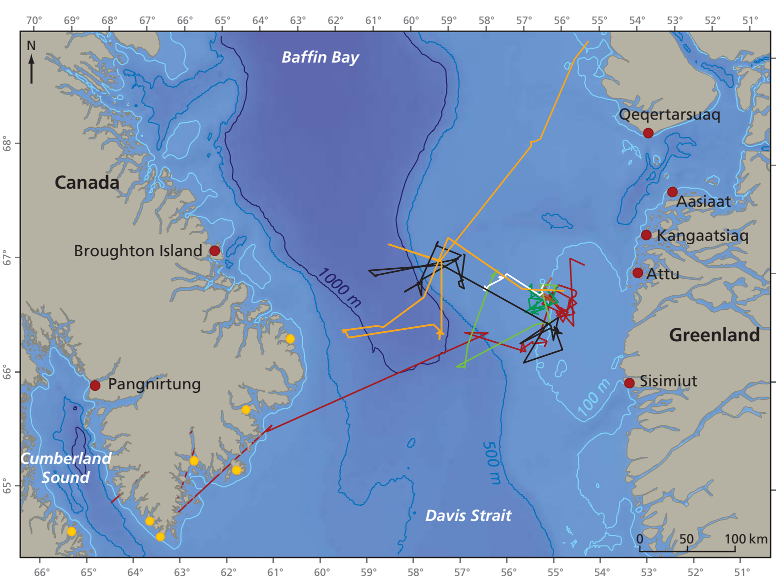

Fig. 33. Preliminary results from the walrus satellite tagging in West Greenland in the spring 2005 and 2006. Major cities (red dots) and haul out sites (yellow dots) shown (Dietz & Born unpublished data).

Contaminant studies

As previously mentioned, marine mammal stock information is of major importance for geographical comparison of contaminant levels. However, there are a number of examples where contaminants have been used to gain an insight in marine mammal stock relations in Greenland and adjacent areas.

Narwhals

Although satellite tracking effort has been applied to the Greenland sites Inglefield Bredning (1990, 2002–2006) and Uummannaq district (1994–1995, 2006), no usable long-term information has been obtained from these regions. To provide information on a possible link in narwhal stocks between these two regions, contaminant analyses were compared from these areas (**Paper 19**). No consistent difference in metal levels (Hg, Cd and Se) between narwhals from Avanersuaq and Uummannaq was found and few statistical differences in mean of sum and individual OHCs (PCBs, DDTs, HCBs, HCHs, trans-nonachlor and toxaphenes) concentrations among the regions were observed. It was therefore concluded that Uummannaq and Avanersuaq are likely to be visited by the same narwhals or by whales with similar contaminant exposure (**Paper 19**). This hypothesis was consistent with the telemetry work, where none of the tagged whales from the

Photo 12. Chemical and DNA analyses from the narwhal hunt has been used to compare narwhal populations and their possible relateness. Photo: R. Dietz.

other 4 summering populations were found to visit Uummannaq, and therefore it would be likely that the whales entering Uummannaq in early winter could come from Inglefield Bredning in Avanersuaq district. PCB and DDT concentrations in West Greenland narwhals were approximately half as high as those found in East Greenland and Svalbard, indicating clear geographical differences among these stocks. This difference between East and West Greenland was in agreement with the genetic information obtained by Palsbøll et al. (1997), demonstrating no population exchange between these regions.

Belugas

Innes et al. (2003) used OHC analyses to detect differences between beluga whales from Canada and West Greenland. Beluga caught by hunters from various hamlets in the Arctic differed in the concentrations of organochlorine contaminants in their blubber. By applying Canonical Discriminant Analysis (CDA) it was possible to separate all seven sampling locations from each other. The analysis provided evidence that most beluga caught by hunters from Grise Fiord, Canada were not the same as beluga caught while migrating along West Greenland. But as previously outlined, these results are at variation with those from the more direct investigations using satellite telemetry (Heide-Jørgensen et al. 2003a). The samples for OHC analyses in Grise Fiord (1984: n=15; 1985: n=5; 1987: n=8) could have been too few and by chance only represented animals wintering in the North Water (see also Richard et al. 1998b). All compared areas were not sampled in the same year, therefore changes over time in OHC levels may also have affected the results of the comparison. The Disko belugas were samples in 1992, whereas the Upernavik and Grise Fiord beluga samples were from 1985–1990 and 1984–1987 (Innes et al. 2002). Whether a year-to-year alternation might exist in the choice of wintering area, i.e. North Water vs. Central West Greenland is uncertain, but according to O'Corry-Crowe et al. (2002) monodontiids have an extreme site fidelity to their selected summering and wintering grounds in the Arctic.

Minke whales

Detection of possible stock relations among minke whales from North Atlantic minke whales has proved particularly challenging. Only limited results from satellite telemetry have been obtained for this species, so the best information available to date is based on a large number of analysis from minke whales hunted in Greenlandic and Norwegian licensed whaling operations over a relatively short time period from 6 May to 31 October 1998. These analyses included contaminants that were used to detect possible stock differences and similarities among minke whales from seven North Atlantic IWC management areas. Overall, the seasonal and spatial distribution of samples was believed to be representative of the Greenlandic and Norwegian catches. Samples were also used to study the regional variation in muscle ^{137}Cs concentrations (Born et al. 2002b), OCH burdens (Hobbs et al. 2003) in the blubber, and various elements including Hg and Cd in soft tissues and baleen (Born et al. 2003). Recently, a relatively novel approach was also adopted to investigate population sub-structure by combining information on regional variation in certain fatty acids (FA), OHCs and heavy metals (Hg and Cd) (Born et al. 2007).

Using the combination of heavy metals, OHCs and fatty acids (FA) in a Canonical Discriminant Analysis (CDA) albeit was possible to separate the minke whales into four sub-populations: West Greenland, a Central Atlantic group represented by whales from Jan Mayen, a Northeast Atlantic group (Svalbard, Barents Sea and northwestern Norway), and the North Sea. The study indicated that a multi-elemental approach based on long-term deposited compounds with different ecological and physiological pathways can be used for identification of sub-populations of marine mammals. These results were also consistent with those obtained from the genetic studies by Andersen et al. (2003).

Part conclusion on satellite tagging linked to contaminants

Understanding marine mammal distribution is of major importance for planning contaminant studies and making valid conclusions from such studies. In some regions, contaminant samples and samples for biological effect parameters can only be obtained during tagging operations. Satellite tagging and contaminant analysis from the same animals has potential for linking contaminant levels with dispersal, behaviour and possible effects on the tagged animals. In cases where tagging has proven difficult to conduct, contaminant analyses can provide information on animal stock relationships.

Genetic, stable isotope and fatty acids studies

Genetic studies are widely used today to establish the extent of contact between subpopulations. We have been involved in a few studies using genetics to separate stocks dealing with narwhals, minke whales and ringed seals (Palsbøll et al. 1997, Andersen et al. 2003, Rew et al. in press), which are some of the key species in our and the AMAP contaminant programmes.

Narwhals

Based on mtDNA, Palsbøll et al. (1997) detected five sub-populations of narwhal from Northern Baffin Bay, Eastern Greenland, Uummannaq district, the 1994 sassat at Kitsissuarsuit and the remaining western Greenland localities including Melville Bay, Upernavik district & Disko Bay, except for the 1994 sassat samples. A later study comparing eight areas/years with a larger sample size documented the same general pattern but also added more details (Riget et al. 2002). As satellite tracking of narwhals from Inglefield Bredning has only resulted in short-term results (Heide-Jørgensen unpublished), genetics and telemetry cannot be compared from this region. The genetic comparisons in mtDNA between a sample from Avanersuaq and Uummannaq narwhals showed significant differences in the comparison carried out by Palsbøll (1997), but using a larger volume of material and splitting up the sample years from Avanersuaq into two periods a connection was shown between these populations in one of two comparisons (Riget et al. 2002). In the comparisons where similarities between the populations were detected, the results agreed with the contaminant studies conducted by Dietz et al. (**Paper 19**). Stable isotope $\delta^{15}N$ values were significantly higher in samples from Uummannaq in 1993 compared to samples from Avanersuaq in 1984 and 1985, indicating that the narwhals in Uummannaq were feeding at a higher trophic level (**Paper 19**). However, a difference in trophic feeding level over an 8-year period could easily take place, even if narwhals from Avanersuaq are connected to those at Uummannaq.

Belugas

In an overview study of genetic relationships of Canadian and adjacent stocks of beluga whales by de March et al. (2002), it was shown that belugas from Lancaster Sound were significantly different from those caught in West Greenland (Upernavik and Disko Bay) based on mitochondrial DNA haplotype distribution. This finding agrees with the OHC studies of Innes et al. (2002) but contradicts the evidence collected by satellite tracking of known individuals moving between the two areas (Heide-Jørgen et al. 2003a). An overall genetic difference was also reported for belugas between Creswell Bay and West Greenland in 1996, where the beluga that moved from Creswell Bay to West Greenland was included in the genetic analysis reported by de March et al. (2002). Whilst no difference was found for samples taken in Creswell Bay in 1993, unfortunately no whales were tagged in that year. In both 1996 and 2001, the satellite tagged whales visited several West Greenland beluga hunting grounds. These results document that genetic sampling can show variable answers dependent on the sampling year, which is also the case for satellite telemetry (Heide-Jørgensen et al. 2003a).

Photo 13. Samples from too few pods may effect the answers to stock discrimination. Photo: R. Dietz.

Monodontiid sampling

The results above bring into question how clearly genetic studies can discriminate between stocks of narwhals and belugas collected from the hunt. Palsbøll et al. (2002) suggested that, due to the nature of the genetic sampling programs, genetic studies are more likely to discriminate pods of related whales, rather than stocks in different areas. The samples for genetic studies are often collected from harvest events, where whales from the same pod are killed. Consequently, there is a high risk of obtaining samples from related individuals. This same sampling bias may be true for whales from satellite-tagging studies at a specific site; however, in many harvest situations, entire pods of a single family unit are sampled for genetic studies, which results in a higher degree of interrelatedness than individual whales that are live captured over a period of days in estuaries. For this reason there is a need to conduct genetic analyses on the same animals as are tracked by telemetry, so that movements and genetics can be compared directly and so that family relations in group sampling and migration patters can be compared.

Ringed seals

Rew et al. (in press) detected no significant levels of genetic heterogeneity among ringed seals from Avanersuaq, Upernavik and Kangaatsiaq (Northwest Greenland) and Danmarkshavn and Kong Oscars Fjord (Northeast Greenland). Nanortalik ringed seals showed genetic differences to Northwest and Northeast Greenland seal, whereas Northeast Greenland and Svalbard were similar. Divergence was also observed between Northwest and Northeast Greenland. These results document that year-round open water and all-year solid ice cover reduces connectivity among seal populations. These existing barriers are likely to change as a result of climatic change (Rew et al. in press).

Minke whales

A study by Andersen et al. (2003) documented the existence of four genetically differentiated minke whale sub-populations: West Greenland, Central North Atlantic-East Greenland-Jan Mayen area, NE Atlantic (Svalbard, the Barents Sea and northwestern Norway), and the North Sea. The genetic four region pattern was detected in a heavy metal study by Born et al. (2003) and a multi-element study Born et al. (2007). Comparisons of OHCs and fatty acids suggested a three region model where the North Sea and Jan Mayen differed respectively (Hobbs et al. 2003, Møller et al. 2003), whereas ^{137}Cs and stable isotopes showed other patterns (Born et al. 2002b, 2003).

Part conclusion genetics, stable isotopes and fatty acids

Genetics provide the strongest supplementary discipline to satellite tagging of animals for obtaining information on population differences. However, results of both techniques are dependent on sample size and methods. Other markers, such as stable isotopes and fatty acids can be used to understand feeding behaviour and link these to contaminant loads but can also provide supplementary information on stock structures.


Photo: R. Dietz

3

Conclusions

The present study has documented contaminant exposure within the Greenland ecosystem. Suggested conclusions to the seven thesis points claimed in the introduction are as follows:

(i) Basic parameters such as age and sex of the animal, tissue type, and season of collection, are likely to affect contaminant concentrations in biota.

(i) Older animals tend to have higher concentrations of Hg and Cd than younger animals in the Greenland marine ecosystem. In some cases the increase with age levels-off in old animals and for Cd in liver and kidney a decrease relative to the maximum level may be seen in old animals. Differences among sexes are seldom recorded for Hg and Cd. Although not consistent among all species, OHC groups or studies, adult males tend to have higher concentrations of OHCs than juvenile and adult females in the Greenland marine ecosystem. In mammals, fat soluble contaminants can be transferred from females to offspring through gestation and lactation, giving mature females a mechanism for excreting these compounds and thereby reducing their body burden. Mercury concentrations are highest in liver, Cd is highest in kidney and OHCs are highest in adipose or liver. The dynamics behind the pathways and accumulation of contaminants are complex and may be driven by many processes. Seasonal differences should, if possible, be taken into account in geographical and temporal trend comparisons.

(ii) Ecosystem structure, differences in trophic level, biomagnification characteristics, and climatic differences will have an affect on contaminant concentrations in biota.

(ii) Due to the longer food chains and hence higher trophic levels of most marine top predators, Hg, Cd and OHC loads in these species are higher than those found in terrestrial ecosystems. Clear biomagnifications are observed for Hg, OHCs and to certain extent Cd throughout the Arctic marine food chain. Differences in trophic level of food, and possible impacts of climatic variability and change are important information that, if possible, should be taken into account in geographical and temporal trend comparisons and future predictions.

(iii) Geographical patterns can be detected in contaminant concentrations within Greenlandic and other Arctic marine mammal populations, reflecting regional loading of the systems.

(iii) Clear geographical trends in contaminants can be detected within the Arctic. Northwest Greenland and the central Canadian Arctic have the highest concentrations of Hg, Central West Greenland and Northwest Greenland have the highest concentrations of Cd, while East Greenland together with Svalbard and Kara Sea have the highest levels of most lipophilic OHCs. This information can be used to identify maximum human exposure, where possible biological effects due to high levels of contaminants are most likely to occur, and where the lowest exposed animals can be obtained as reference groups.

(iv) Temporal trends in contaminant levels can be detected for key species in the Greenland ecosystem, reflecting global trends in emissions and pathways.

(iv) Investigations of animal hard tissue and other long-term environmental archives have revealed long-term increases of Hg with a substantial anthropogenic contribution. East of Greenland this increase levelled-off somewhere around the 1960s-1970s, followed by significant declines. In West Greenland, Hg increased throughout the 20th century but detailled information concerning trends during recent decades is sparse. Some West Greenland time series, as well as Central Canadian Arctic time-series indicate a continued increase of Hg.

The best available time series for “legacy OHCs” such as PCBs, DDTs, HCHs, HCB, chlordanes, dieldrin, and coplanar PCBs show a decline in levels of these contaminants. Time-series on toxaphene, PCDDs and PCDFs are more uncertain, but may be decreasing. Increases in concentrations of a number of “new” OHCs such as the PBDEs and the PFCs took place prior to the turn of the millennium in the entire Arctic. PFCs continue to increase in Greenland, but there is some evidence that PFCs and PBDEs over recent years this trend may have decreased or reversed in some areas.

The Greenland results are, in most cases consistent with longer time-series from other parts of the Arctic, but with a delay relative to the trends observed in Northern Europe. The observed trends show a response to regulations restricting the use of chemicals, which have been introduced at the national and international level. Most of the time-series based on contaminants analyses in soft tissue however, currently still include too few sampling years to provide a clear picture of interannual variation, and onset- and end of the relative change over time. Some highly relevant sample matrices, including polar bear samples available for the east coast of Greenland provide the potential for a high resolution insight into trends over the time period from 1983 until today, with sampling continuing.

(v) The most highly exposed groups in the Arctic system, i.e. top predators, may be affected by contaminants, however even well defined and examined epidemiologic disease outbreaks such as PDV can be hard to link to contaminant levels due to confounding parameters.

Photo 14. Increased OHC concentrations have been linked to reduced size in both male an female polar bear reproductive organs. Photo: R. Dietz.

(v) Epidemiological studies on Arctic human populations indicate neuropsychological dysfunction in some humans that resemble effects seen in experimental animals. The first histopathological and neurochemical receptor biomarkers investigations indicated that effects of Hg can not be excluded. Cardiovascular and maybe other effects of Hg at higher trophic levels may be reduced and in some cases eliminated by the protective effect of Se, which is present in surplus in the Arctic marine ecosystem. Although Cd concentrations in several marine species are above threshold effect levels, Cd has not so far been proven to cause effects in Arctic wildlife.

Reduced size of reproductive organs was found for both male and female polar bears in relation to increased OHC concentrations. However, previous observation on pseudohermaphroditism in female polar bears from Svalbard could not be verified from examination of a single animal from Greenland that was found with an enlarged clitoris. Tissue alterations were found in liver and kidney that could be linked to certain OHCs. However, this was not the case for immunological organs such as lymph nodes, spleen, thymus and thyroid tissue. However, no relationship was found between skull pathology and organohalogens. Fluctuating asymmetry in polar bears showed variable results dependant on the analyhical method used. Some of the lacking skeletal effects were probably due to subeffect exposure to OHCs, influence of nutritional status, genetic factors or other confounding environmental factors such as climate change. A daily intake of amounts of 50–200 g marine mammal blubber from Greenland may cause an impairment of the immune system in top predators.

For well-defined mass mortality events, such as the two PDV outbreaks, where considerable effort was expended on the detection of deaths and investigation of possible population effects, it has not been possible to establish any clear linkage between contaminants, immuno-suppression and number of deaths caused by the disease. A large number of confounding factors play a major role in such disease

Photo 15. Satellite telemetry work have identified polar bear population throughout the Arctic. Photo: R. Dietz.

events. The epidemiology of PDV in European seals showed that previous contact to PDV, local population growth rates, metapopulation structure, and seasonality of the infection has a major influence on the degree of spread of the pathogen.

(vi) *The Inuit population can reduce their contaminant intake by following food recommendations and thereby reduce their risk of being affected by contaminants.*

(vi) The Inuit population can minimize their contaminants intake and risk of health problems by reducing their intake of internal organs (Hg, Cd and PFCs), fatty tissue (OHCs) and preferentially eating low trophic species. Intake of young animals will result in lower Cd and Hg and OHC exposure. For OHCs, consumption of adult female animals may result in lower intakes compared to consumption of adult males. International legislation and changes in anthropogenic processes are beginning to reduce the unintended transboundary exposure of the Arctic ecosystem, which will result in reduced contaminant intake of the Greenlandic and other Arctic population. At the same time, these foods are sources of important nutrients and changes in diet can bring other health risks. Food advisories should hence be developed by relevant health authorities in consultation with local people, to prevent that changes in diet will result in unintentional adverse affects on health.

(vii) *Population structure studies are highly relevant to both the conduct of contaminant monitoring and to the interpretation of information on contaminant patterns and trends in the Arctic.*

(vii) Marine mammal distribution is of major importance for planning contaminant studies and ensuring the valid interpretation of results of such studies. New information based on satellite telemetry is accumulating for key species such as polar bear, narwhals, belugas, walrus and some baleen whales, for some regions and seasons in Greenlandic and adjacent waters. Other species like ringed seal have only been studied in Northwest Greenland, whereas other toothed whales and bearded seals have yet to be studied.

In some regions outside Greenland, contaminant samples and samples for biological effect parameters can only be obtained during tagging operations. Satellite tagging and contaminant analysis from the same animals has the potential for linking contaminant levels with dispersal, behaviour and possible effects on the tagged animals. In cases where tagging has proven difficult to conduct, genetic studies on hunted animals or using biopsies represents the best supplementary discipline to satellite tagging for resolving population relationships. Contaminants have also been used elucidate population relationships. Other disciplines such as stable isotopes and fatty acids may be used to provide supplementary information on stock structures, in addition to information to understand feeding behaviour and explain and normalize contaminant loads.


Photo: R. Dietz

4

Recommendation for future investigations

Photo 16. Further investigations are needed to resolve the impact of climate change on contaminant pathways and exposure. Photo: R. Dietz.

The following recommendations are presented for future contaminant studies relating to the scope of the present dissertation:

Basic parameters
It is suggested that statistical analysis and interpretation of data on contaminants in biota incorporate relevant biological data such as age, sex, season, stable isotopes, etc., to understand linkages and improve reliability of comparative studies.

Ecosystem
There is a need to study unresolved and "new" chemicals throughout the marine food webs in order to clarify and understand the uptake, transfer and biomagnification of these contaminants. This is particularly important if changes in climate and ocean currents alter food webs, species distributions or pathways of delivery of contaminants.

Temporal and geographic trends
Temporal trend monitoring of heavy metals and OHCs, including "new" and current-use chemicals, in biota should be continued to obtain longer, more statistically reliable time-series. Time-trend datasets can support human and wildlife exposure estimates and predictions of future scenarios. Such time-series also enable assessment of whether measures taken under international conventions and agreements are effective in reducing concentrations of contaminants of concern in the Arctic environment and ecosystems.

Studies of long-term historic time-series of Hg in biota hard tissue should be supported to extend soft-tissue data series and elucidate relationships to pre-industrial baseline levels, and linkages to historical anthropogenic contamination. Such studies should also investigate the relationship between soft- and hard-tissue concentrations in biota, to improve linkages between long-term time-series and contemporary concentrations.

Effects
Effects of heavy metals and OHCs in Arctic species that exhibit tissue levels of concern should be conducted. Initiatives should continue to refine and develop methods for detecting subtle biological effects related to contaminants. Effects monitoring can include critical tissue effect thresholds, relationships between indicators of exposure (e.g. biomarkers, histopathological investigations, behavioural and reproductive parameters) and other observed effects in Arctic biota.

More research is also needed on toxicity mechanisms of many OHCs, including establishment of effects thresholds for 'new' substances and metabolites. If possible, such data should be integrated with information on effects on population level and general health.

Climate

Dramatic changes in climate are predicted to take place in the coming decades in the Arctic. Some species will benefit from an increase in temperature and others will suffer. Scenarios linking climate change, contaminant pathways and contaminant levels have been discussed to a minor extent, but further investigations are needed. Rapid climatic change provides obvious challenges for understanding such relationships. The extensive sample collections and experience obtained during the contaminant programmes implemented in recent years should be employed to study such changes, including changes in nutritional and trophic status in species of concern. Combined effects of climate change and contaminants, including increased mobilization of contaminants during starvation in high trophic marine species should also be investigated.

Research to better understand the processes of transport, abiotic factors and climate change that may influence spatial patterns and temporal trends should be encouraged.

Population relations

Marine mammal distribution studies should be continued to resolve population differences and relationships of importance for contaminant studies. Sampling for contaminant studies during tagging operations should be encouraged for protected species and to better link contaminant levels with dispersal, behaviour and possible effects on the tagged animals. Other mutually advantageous combinations of work under different disciplines, including use of available tissue samples, contaminants, telemetry, genetic studies, stable isotopes and fatty acids can provide additional information valuable for addressing a large number of inter-related questions.

Photo 17. There are still challenges in tagging marine mammals to document population discreteness and linkages. Photo: R. Dietz.


Photo: R. Dietz

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Photo: R. Dietz

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7

Appendices

Legislation and International Conventions

Several of the Arctic countries have enacted national legislation, or in the case of Denmark, Finland and Sweden, EU legislation, that bans or restricts the production and use of certain chemicals. Other chemicals are subject to voluntary agreements with, for example, industry. These measures have been introduced at different times in different countries. The eight Arctic countries have now banned most "legacy" OHCs; e.g. chlordane (1967 and 1996), toxaphene (1969 and 1996), PCB between 1970 and 1995, DDT, aldrin, dieldrin (1970 and 1996), HCB (1977 and 1996) in the period between 1969 and 1996 (de March et al. 1998).

One of the conclusions of AMAP Phase I was that many contaminants have a global distribution and reach the Arctic as a result of long-range transport from sources regions far to the south. National measures by the Arctic countries, therefore, cannot on their own mitigate much of the contamination of the Arctic. Solving problems associated with contaminants in the Arctic, therefore, requires measures at the global scale. This Arctic message was heard, and had an effect on international negotiations that were underway at the time; the outcome being new international agreements on actions to reduce OHC and heavy metal contamination at the global level.

One of the conclusions in the AMAP POPs Phase II assessment by de Wit et al. (2004) was the explicit statement that: *"One of the most important accomplishments of Arctic research concerning OCs, and the AMAP Phase I POPs assessment was the role it played in the negotiations of a global agreement to ban the "dirty dozen" OCs (PCBs, DDT, etc.)."* The AMAP Phase II assessment will make a similar contribution to ongoing work, both to support these agreements and work to strengthen agreements with respect to Hg and the "new" OHCs, in particular brominated and fluorinated compounds.

The AMAP information assisted the negotiation of a number of international Conventions. Among these were of the Protocols on persistent organic pollutants (POPs) and heavy metals to the United Nations Economic Commission for Europe's Convention on Long-range Transboundary Air Pollution (LRTAP Convention), the establishment of a global agreement on POPs (the Stockholm Convention), as well as the Basel Convention and the Rotterdam Convention. Persistence, long-range transport potential, bioaccumulation, and toxicity are screening criteria under these conventions, which are applied to proposals to add substances to the agreements. Information from investigations conducted by colleagues and ourselves have been compiled and assessed under the outspieces of AMAP and have provided much relevant information on heavy metals and OHCs.

Further information on the UNECE LRTAP Convention, UNEP Chemicals, and The Stockholm Convention on Persistent Organic Pollutants, The Rotterdam Convention and the Basel Convention is presented below.

International Convention details

LRTAP Convention

The history of the LRTAP Convention can be traced back to the 1960s, when scientists demonstrated the interrelationship between sulphur emissions in continental Europe and the acidification of Scandinavian lakes. The 1972 United Nations Conference on the Human Environment in Stockholm signalled the start for active international cooperation to combat acidification. Between 1972 and 1977 several studies confirmed the hypothesis that air pollutants could travel several thousands of kilometres before deposition and damage occurred. This also implied that cooperation at the international level was necessary to solve problems such as acidification. In response to these acute problems, a High-level Meeting within the Framework of the ECE on the Protection of the Environment was held at ministerial level in November 1979 in Geneva. It resulted in the signature of the Convention on Long-range Transboundary Air Pollution by 34 Governments and the European Community (EC). The Convention was the first international legally binding instrument to deal with problems of air pollution on a broad regional basis. Besides laying down the general principles of international cooperation for air pollution abatement, the Convention sets up an institutional framework bringing together research and policy.

Since 1979 the Convention on Long-range Transboundary Air Pollution has addressed some of the major environmental problems of the UNECE region through scientific collaboration and policy negotiation. The Convention has been extended by eight protocols that identify specific measures to be taken by Parties to cut their emissions of air pollutants. The Convention, which now has 50 Parties, identifies the Executive Secretary of UNECE as its Secretariat. Of the 8 protocols, 4 are directly relevant in relation to the contaminants in the Arctic dealt with in this thesis. These are:

> The 1998 Protocol on Persistent Organic Pollutants (POPs); 25 Parties. Entered into force on 23 October 2003.
>
> The 1998 Protocol on Heavy Metals; 27 Parties. Entered into force on 29 December 2003.
>
> The 1991 Protocol concerning the Control of Emissions of Volatile Organic Compounds or their Transboundary Fluxes; 21 Parties. Entered into force 29 September 1997.
>
> The 1984 Protocol on Long-term Financing of the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP); 41 Parties. Entered into force 28 January 1988.

Information below is based on the relevant home pages for the conventions.

UNEP Chemicals

UNEP Chemicals is the main catalytic force in the UN system to ensure the sound management of hazardous chemicals. It promotes chemical safety by providing countries with access to information on toxic chemicals, helping to build countries' capacity to manage risks posed by chemicals throughout their life-cycle, and by supporting global actions that address chemical issues of international concern. Examples include the Stockholm Convention on Persistent Organic Pollutants (POPs), the Rotterdam Convention on the Prior Informed Consent (PIC) Procedure for Certain Hazardous Chemicals and Pesticides in International Trade, and the negotiations for a Strategic Approach to International Chemicals Management (SAICM). The information below has been obtained through the UNEP homepage (www.unep.ch).

The Stockholm Convention

The Stockholm Convention on Persistent Organic Pollutants (POPs) was negotiated under UNEP's auspices and adopted by a Conference of Plenipotentiaries in May 2001. It entered into force on 17 May 2004, and by 1 November 2004 had 82 Parties. The Stockholm Convention is a global treaty to protect human health and the environment from POPs through measures designed to reduce and eliminate their release. Currently Parties are required to take action on an initial 12 specified POPs. UNEP provides the secretariat to the Convention and implements actions to support its implementation including: creating awareness of the POPs issue, the Convention, its provisions and implementation actions; preparing guidelines for best available techniques and best environmental practices for unintentionally produced POPs; and establishing and maintaining databases and an information clearinghouse on POPs. UNEP organized the first Conference of the Parties of the Convention, which was held in Uruguay, in May 2005.

The Rotterdam Convention

The Rotterdam Convention on the Prior Informed Consent (PIC) Procedure for Certain Hazardous Chemicals and Pesticides in International Trade were negotiated under the auspices of UNEP and the UN Food and Agriculture Organization (FAO) and adopted by a Conference of Plenipotentiaries in September 1998. The Convention entered into force on 24 February 2004 subsequent to its ratification by 50 countries. The PIC procedure requires exporters trading in listed hazardous substances to obtain the prior informed consent of importers before proceeding with trade. Between 1 and 5 million cases of pesticide poisoning occur each year, mostly in the developing world. Thousands of these cases are fatal. In developed countries, the most hazardous pesticides are either banned or strictly controlled, and farm workers who use them wear protective clothing and equipment. In developing countries—which use only 25 per cent of global pesticide production but account for 99 per cent of deaths—such safeguards are less common. As well as preventing shipment of listed hazardous chemicals without prior informed consent, the Rotterdam Convention enables Parties to alert each other about possible risks. Whenever a government bans or restricts a chemical for health or environmental reasons, this is reported to all Parties. UNEP provides the secretariat for the Rotterdam Convention jointly with FAO and organizes capacity building for the national implementation of the Convention's procedures. The Convention held its first Conference of the Parties in September 2004 in Geneva, Switzerland. At the meeting 14 new hazardous chemicals were added to an initial watch list of 27 substances.

Basel Convention

The Basel Convention was adopted on 22 March 1989 by the Conference of Plenipotentiaries which was convened at Basel from 20 to 22 March 1989. The Basel Convention entered into force in 1992. The central goal of the Basel Convention is "environmentally sound management" (ESM), the aim of which is to protect human health and the environment by minimizing hazardous waste production whenever possible. ESM means addressing the issue through an "integrated life-cycle approach", which involves strong controls from the generation of a hazardous waste to its storage, transport, treatment, reuse, recycling, recovery and final disposal. Many companies have already demonstrated that "cleaner production" methods which eliminate or reduce hazardous outputs can be both economically and environmentally efficient. The United Nations Environment Programme's (UNEP) Division on Technology, Industry and Economics works to identify and disseminate "best practices" (http://www.unepie.org/). In the coming decade, more emphasis will be placed on creating partnerships with industry and research institutions to create innovative approaches to ESM. One of the most critical aspects of ESM is lowering demand for products and services that result in hazardous by-products. Consumers need to educate themselves as to the methods used in production processes and think about what they buy every day.

This dissertation – based on 30 selected English articles and book contributions – was accepted for public defense by the Faculty of Science, University of Copenhagen to acquire the doctor's degree in natural sciences.

Seven thesis points is being addressed within three thematic topics; marine contaminant loads, health effects of contaminants and marine mammal migration and stock separations. The contaminant part provides a review of key determining parameters (age, sex, season, food and climate), trends (geographic and temporal), bioaccumulation, biomagnification and human exposure. The biological health effect section deals with observed effects of contaminants in top predators in the Arctic marine ecosystem as well as a discussion on mass mortality epizootics among Arctic and European mammals.

Finally, marine mammal distribution and stock separations are discussed based on information from satellite telemetry, contaminant studies as well as genetic, stable isotope and fatty acids profiles.

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