Available online at www.sciencedirect.com
SCIENCE DIRECT®
Science of the Total Environment 351–352 (2005) 369–390
Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Humankind
www.elsevier.com/locate/scitotenv

Chlorinated hydrocarbon contaminants and metabolites in polar bears (*Ursus maritimus*) from Alaska, Canada, East Greenland, and Svalbard: 1996–2002

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Received 28 May 2003; received in revised form 4 October 2004; accepted 20 October 2004
Available online 22 August 2005

Abstract

A suite of chlorinated hydrocarbon contaminants (CHCs) including organochlorine pesticides (OCPs) and by-products, polychlorinated biphenyls (PCBs), and methyl sulfone ($MeSO_2$) PCB and *p*,*p*′-dichlorodiphenyldichloroethylene (*p*,*p*′-DDE) metabolites were determined in adipose tissue of 107 adult and sub-adult polar bears, almost exclusively females, sampled between 1996 and 2002 from populations spanning Arctic and Subarctic regions of Alaska, Canada, East Greenland, and Svalbard. The East Greenland and Svalbard populations of polar bears were distinguished by higher proportions of dichlorodiphenyldichloroethane (DDT)-related compounds, nonachlors, oxychlordane, and higher-chlorinated and persistent PCB

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doi:10.1016/j.scitotenv.2004.10.031

congeners (hepta- to nona-chlorinated). Conversely, Alaska, the westernmost population of the North American Arctic, was characterized by higher proportions of relatively volatile compounds such as hexachlorocyclohexanes (HCHs) and pentachlorobenzene (PnCBz), lower-chlorinated PCB congeners (tri- to penta-chlorinated), and lower proportions of oxychlordane. Geometric mean (GM) with 95% confidence limits (CL) ΣHCH concentrations were highest in Alaska male polar bear fat samples (GM 593; CL 363−909 ng g^{-1} lipid weight), ΣDDT concentration were highest in East Greenland female samples (GM 309; CL 249−490 ng g^{-1} l.w.), and Σ_{42}PCB (GM 5972; CL 4637−9129 ng g^{-1} l.w.) and Σ$MeSO_2$-PCB (GM 198; CL 162−279 ng g^{-1} l.w.) concentrations were highest in female samples collected from Svalbard. The distribution of Σ-chlordane-related compounds (ΣCHL), ΣCBz, mirex, and dieldrin was relatively uniform among the populations of polar bears investigated. The present 1996−2002 data of female polar bear fat samples was compared to spatial assessments of female polar bear fat samples collected between 1989 and 1993 from comparable populations. The two-point temporal comparisons showed a general decrease for age-adjusted mean concentrations of ΣCHL, *p*,*p*′-DDE, Σ_{42}PCB, Σ$MeSO_2$-PCB and 3-$MeSO_2$-*p*,*p*′-DDE over a period of approximately 10 years. However, concentrations of dieldrin were comparatively unchanged. Comparisons of present 2001−2002 concentrations in fat of female polar bears from Western Hudson Bay showed great consistency with temporal trends (1991−1999) previously reported for the same region, i.e. the apparent non-decreasing trend of ΣCHL, β-HCH, ΣHCH and dieldrin, and the apparent declining trend for ΣPCB. However, present concentrations of α-HCH and ΣCBz were elevated, and ΣDDT was notably lower in Western Hudson Bay samples compared to the last measurements in fat samples collected in 1999, which was not in accord with reported temporal trends for this region. As a result of their relatively high degree of contamination, East Greenland and Svalbard polar bears are at higher health risk of contaminant exposure among Arctic and Subarctic populations. In addition to continued biomonitoring, further research on health and population status is needed to evaluate the impact from chronic exposure of polar bear populations to CHCs and their metabolites.

Keywords: Canadian arctic; Polar bears; *Ursus maritimus*; Arctic; Chlorinated hydrocarbon contaminants; Metabolites; Geographical trends; Temporal comparisons

1. Introduction

Over the last 15 years, monitoring of environmental contaminants in the polar bear (*Ursus maritimus*) has received special attention because of its ability to accumulate a wide range of chlorinated hydrocarbon contaminants (CHCs) and their metabolites. Owing to their lipophilicity and persistence, CHCs have high bioaccumulative potential and biomagnify at higher trophic levels in arctic food chains (Muir et al., 1988; Borgå et al., 2001; Fisk et al., 2001). Hence, the polar bear, which is at the top of the Arctic marine food chain, reaches some of the highest concentrations of CHCs of any arctic mammal species (de March et al., 1998; Muir et al., 1999; Fisk et al., 2003; AMAP, 2004). as recently reviewed by Braune et al. (2005–this issue) the polar bear serves as an ideal sentinel species for biomonitoring spatial and temporal trends of CHCs, such as chlordane (CHL)-related compounds and polychlorinated biphenyls (PCBs), as a result of its wide distribution throughout the Arctic and Subarctic and its key role as apex predator in the marine environment (de March et al., 1998; Muir et al., 1999; Muir and Norstrom, 2000; Fisk et al., 2003; AMAP, 2004). As described in Norstrom et al. (1998), there are discrete populations of polar bears in the Canadian Arctic. Combined with knowledge of sea ice and land barriers, 12 polar bear management zones were defined. In general, polar bears in some areas are philopatric with little exchange among populations, although overlap can be significant over the winter. The average marine area over which a polar bear subpopulation integrates contaminants is in the order of 25,000 km^2 in Alaska. The area may be much smaller in areas where food supply is plentiful year round, such as in Viscount Melville Sound in the northwest area of the Canadian high Arctic.

Global contamination by environmentally persistent CHCs in wildlife and humans has been widely documented (de March et al., 1998; Macdonald et al., 2000; Fisk et al., 2003; AMAP, 2004). Emerging contaminants such as methyl sulfone ($MeSO_2$) PCB and *p*,*p*′-dichlorodiphenyldichloroethylene (*p*,*p*′-DDE) metabolites have been documented as impor-

tant classes of lipophilic contaminants as they have been detected in a growing number of species, including marine mammals from the Arctic (Letcher et al., 2000). In recent years, concentrations of $MeSO_2$-PCB and 3-$MeSO_2$-*p*,*p*′-DDE metabolites have been determined in fat, liver, and blood of polar bears from Canadian and East Greenland populations (Letcher et al., 1995, 1998, 2000; Sandala et al., 2004). These metabolites have been associated with a number of biological effects in mammals in vivo and in vitro such as endocrine system modulation (Letcher et al., 2000, 2002). Studies dealing with the effects of CHCs on polar bear health have raised significant concerns. As recently summarized by Fisk et al. (2005–this issue), the current understanding of contaminant-induced biological effects in polar bears points to evidence that chronic exposure to CHCs and their metabolites may compromise endocrine functions and homeostasis (Sandau, 2000; Norstrom, 2000; Skaare et al., 2001a; Letcher et al., 2002; Haave et al., 2003; Oskam et al., 2003, 2004; Braathen et al., 2004), immune functions (Bernhoft et al., 2000; Norstrom, 2000, 2001; Skaare et al., 2001b; Larsen et al., 2002; Lie et al., 2004a, 2005; Kirkegaard et al., 2005), cub and reproductive female survival (Derocher et al., 2003), reproduction and development (Wiig et al., 1998), and hepatic P450-enzymes induction (Bandiera et al., 1995; Letcher et al., 1996).

Since CHCs were first reported in polar bear tissues nearly 30 years ago (Bowes and Jonkel, 1975), monitoring of contaminants in polar bears has gained greater notoriety. Subsequently, systematic surveys of the geographical distribution of CHCs in polar bears have been undertaken throughout most of the Arctic and Subarctic regions, and an extensive database of CHC measurements is available from the 1980s and 1990s, and of measurements dating as far back as late 1960s (Norstrom et al., 1988; Norheim et al., 1992; Norstrom and Muir, 1994; Letcher et al., 1995, 1998; Bernhoft et al., 1997; Norstrom et al., 1998; Andersen et al., 2001; Norstrom, 2000; Kucklick et al., 2002; Derocher et al., 2003; Lie et al., 2003; Olsen et al., 2003; Dietz et al., 2004, Sandala et al., 2004). To date, the highest degree of contamination among circumpolar polar bear populations has been reported from the western Russian Arctic, followed by Svalbard (Norstrom et al., 1998; Andersen et al., 2001; Lie et al., 2003), whereas the lowest concentrations of contaminants were reported from Alaska (Bering/Chukchi Sea) polar bear samples (Norstrom et al., 1998; Kucklick et al., 2002).

Concentrations of CHCs appear to have declined generally throughout the Arctic and Subarctic over the last decade (Braune et al., 2005–this issue). A circumpolar assessment of polar bear adipose tissue samples from 1989−1993 showed a general decrease of sum (Σ) CHL, *p*,*p*′-DDE, and dieldrin concentrations compared to those in 1982−1984, although no significant change was reported for ΣPCB concentrations. Furthermore, in an assessment of temporal trends (1991–1999; except 1996) of CHCs in the fat of Hudson Bay polar bears, Norstrom (2000) found a significant decline in α-hexachlorocyclohexane (α-HCH), Σ-chlorobenzene (ΣCBz), and ΣPCB, although no significant change was observed for ΣCHL, Σ-dichlorodiphenyldichloroethane (ΣDDT), β-HCH, and dieldrin. Following this trend, concentrations of PCBs, inferred from concentrations of CB-153, in polar bear plasma collected from Svalbard were found to decline significantly during the period 1990−1998 (Henriksen et al., 2001; Derocher et al., 2003). Furthermore, Dietz et al. (2004) reported recently a decline in ΣCHL, ΣDDT, ΣPCB, and dieldrin concentrations in East Greenland polar bear fat samples collected between 1990 and 1990−2001.

Concentrations of CHCs in polar bears have been monitored consistently in Hudson Bay throughout the 1980s and 1990s (Norstrom, 2000), and during the 1990s at Svalbard (Henriksen et al., 2001; Derocher et al., 2003). However, comprehensive studies on the spatial trends of contaminants including most regions of the Arctic and Subarctic were last carried out for polar bears in the late 1980s and early 1990s (Letcher et al., 1995; Norstrom et al., 1998; Andersen et al., 2001; Lie et al., 2003). To address this data gap we have determined a suite of CHCs including organochlorine (OC) pesticides and by-products, PCBs, and $MeSO_2$-PCB/-*p*,*p*′-DDE metabolites in adipose tissue samples of polar bears from populations spanning the Subarctic and Arctic regions of Alaska, Canada, East Greenland, and Svalbard. The objective of this assessment was to investigate geographic differences in concentrations and congener patterns of OCs, PCBs, and $MeSO_2$-PCBs/-*p*,*p*′-DDE in most of the regions that are inhabited by polar bears. Additional objectives were to compare the present 1996−2002 con-

centrations in fat of female polar bears with those of earlier assessments from comparable populations, and to comment on the current CHC concentrations and the potential implications of exposure on polar bear health. There are companion papers documenting new organohalogen contaminants of concern, polybrominated diphenyl ethers (PBDEs) (Muir et al., submitted for publication), and perfluorinated acids (Smithwick et al., in press) in fat and liver, respectively, in polar bears from the same locations across the western Arctic.

2. Materials and methods

2.1. Field sampling

Samples of adipose tissue from 107 adult and subadult polar bears were collected during 1996−2002 from Alaska, six regions of Canada, East Greenland, and Svalbard (Table 1). At Svalbard, fat biopsy samples were collected from free-ranging polar bears tranquilized for research purposes by remote injection of a drug (Zoletil®)-filled dart (Palmer Cap-Chur Equipment, Douglasville, GA, U.S.A.) fired from a helicopter (Stirling et al., 1989). In Alaska, Canada, and East Greenland, subcutaneous fat samples were collected from harvested polar bears as part of the native subsistence hunt. Subcutaneous fat/fat biopsy samples were collected from the base of the tail (rump fat) within 12 h post mortem for Alaskan, Canadian, and East Greenland bears, or shortly following capture for anaesthetized Svalbard bears, and stored in individual Zip-Lock bags. All samples were kept at −20 °C or at lower temperature before sample preparation and extraction. For details on sample storage and preparation, see Norstrom et al. (1988, 1998), Dietz et al. (2004) and Sandala et al. (2004). The dates and locations of sampling and sex were recorded for each individual, and a vestigial premolar tooth (PM_1) was extracted for age determination. The age of the individuals was determined by counting annual growth layers in the cementum of the decalcified stained tooth according to methods described elsewhere (Stirling and Archibald, 1977; Calvert and Ramsay, 1998; Dietz et al., 2004).

For samples collected at Svalbard, capture and handling methods were approved by the Norwegian

Table 1
Sampling year, sample size, age (mean, range and number of animals >5 years old), percentage extractable lipid content in adipose tissue (mean ± 1 SD and range), and geographic co-ordinates of nine Arctic and Subarctic populations of polar bears

	Alaska (males)	Alaska (females)	Amundsen Gulf	W. Hudson Bay	Foxe Basin/Gulf of Boothia	Lancaster Sound/ Jones Sound	N. Baffin Island	S. Baffin Island	E. Greenland	Svalbard
Sampling year	1996–2002	1996–1999	2001–2002	2001–2002	2001–2002	2001–2002	2001–2002	2001–2002	1999–2002	2002
Sample size	7	5	10	14	8	6	10	16	16	15
	8.8	7.8	6.7	9.8	4.9	9.8	6.1	7.8	11.5	11.8
Age	3–14	3–14	2–22	5–14	3–9	4–19	4–11	4–20	4–20	5–25
	5	3	4	14	3	5	6	14	15	15
% Lipid	62.3 ± 21.1	80.5 ± 9.05	94.9 ± 4.62	59.8 ± 13.3	88.7 ± 5.86	83.6 ± 9.45	85.0 ± 13.3	91.8 ± 5.21	96.5 ± 5.31	56.3 ± 15.1
	32.2–81.4	64.7–87.0	85.3–100	36.1–90.0	80.7–95.8	72.0–93.6	60.0–100	83.3–100	83.5–100	23.0–80.7
Co-ordinates	63–71° N	70–71° N	70–72° N	59–61° N	69–70° N	74–77° N	70–73° N	63–68° N	69–74° N	77–80° N
	143–171° W	148–156° W	116–126° W	94–95° W	80–92° W	81–95° W	70–80° W	63–71° W	20–25° W	14–27° E

Animal Research Authority and the Governor of Svalbard. For samples from Canada, appropriate permits and community approval were obtained from the Nunavut Wildlife Research Permit, the Wildlife Management Advisory Committee, and the Inuvialuit Game Council and diverse Hunters and Trappers Committees. Samples from Alaska were transported under U.S. export and Canadian import permits under the Convention on International Trade in Endangered Species (CITES) and the U.S. Marine Mammal Protection Act of 1972. Sampling in Alaska was coordinated by individual hunters, the U.S. Fish and Wildlife Service, and the Alaska Nanuuq Commission. Samples from East Greenland were transported under Danish export and Canadian import permits under the CITES.

2.2. Chemical analyses

Chromatographic materials used for analysis were as follows: Florisil® (Magnesium silicate, F100−500, 60−100 mesh) and basic aluminium oxide (Brockman activity grade I, 60−325 mesh) (Fisher Scientific, Ottawa, ON, Canada), and silica gel (Grade 62, 60−200 mesh, 150 Å) (Aldrich Chemicals, Milwaukee, WI, U.S.A.). Deactivation of these materials was achieved with doubly distilled, *n*-hexane washed H_2O. Bio-beads S-X3 (200−400 mesh) were purchased from Bio-Rad Laboratories (Hercules, CA, U.S.A.). All solvents were of pesticide residue grade quality.

OC and PCB standard mixtures were supplied by the Canadian Wildlife Service (Ottawa, ON, Canada). 1,3,5-tribromobenzene (Accu-Standard, New Haven, CT, U.S.A.) was used as the OC/PCB recovery efficiency standard. 3-CH_3SO_2-2-CH_3-2′,3′,4′,5,5′-pentachlorobiphenyl ($MeSO_2$-PCB-IS) was used as internal standard for $MeSO_2$-PCBs and 3-$MeSO_2$-*p*,*p*′-DDE, and was kindly supplied by Dr. Åke Bergman (University of Stockholm, Department of Environmental Chemistry, Stockholm, Sweden), who also supplied the $MeSO_2$-PCB/-*p*,*p*′-DDE standard mixtures.

The concentrations of OCs, PCBs, and $MeSO_2$-PCBs/-*p*,*p*′-DDE in polar bear adipose tissue were determined based on quantification methods described by Dietz et al. (2004) and Sandala et al. (2004) with modifications described here. Briefly, accurately weighed adipose tissue samples (0.5 g) were homogenized with anhydrous sodium sulphate (6 : 1 ratio by weight), and spiked with OC/PCB recovery and $MeSO_2$-PCB-IS standards. Samples were extracted using *n*-hexane:dichloromethane (DCM) (55 : 45), and the lipids and other biogenic materials were removed by gel permeation chromatography (GPC). The percentage extractable lipid content was determined gravimetrically by evaporating the first GPC fraction (130 mL). The remaining GPC fraction (170 mL) was concentrated and eluted through a 33% KOH/silica gel (1.5 g) column with *n*-hexane:DCM (1 : 1), and concentrated to 1 mL. Instead of a KOH/silica gel column, samples from Alaska and East Greenland were chromatographed on activated silica gel (8 g, 1.1 cm internal diameter (i.d.) column), and eluted with hexane followed by *n*-hexane:DCM (1 : 1) to separate OCs from most other PCBs. Each sample was then transferred quantitatively to a Florisil® column (8.0 g, 1.2% H_2O deactivated) and four fractions were collected. The first fraction (F1), containing PCBs, was eluted with 38 mL of *n*-hexane. The second fraction (F2), containing OCPs and by-products, was eluted with 34 mL of 15% DCM/*n*-hexane. The third fraction (F3), containing heptachlor epoxide and dieldrin, was eluted with 54 mL of *n*-hexane:DCM (1 : 1). Finally, the fourth fraction (F4), containing $MeSO_2$-PCBs/-*p*,*p*′-DDE, was eluted with 80 mL of 7% methanol/DCM. The F1, F2 and F3 were concentrated to 1 mL by weight density (37 °C), and to the final volume of 100 μL in 2,2′,4-trimethylpentane (iso-octane) under a gentle flow of nitrogen in preparation for gas chromatography with micro electron capture detection (GC-μECD) analysis. The F4 was further eluted on a basic alumina column (3.0 g, 2.3% H_2O deactivated) with 50 mL *n*-hexane:DCM (1 : 1) to remove possible co-eluting artifacts, concentrated to 1 mL by weight density (37 °C), and then to the final volume of 100 μL in iso-octane in preparation for GC-μECD analysis.

Aliquots of the final extracts were injected automatically on a GC (Agilent 6890; Agilent Technologies, Palo Alto, CA, U.S.A.) equipped with a splitless injector (Agilent 7673; Agilent Technologies, Palo Alto, CA, U.S.A.) and a ^{63}Ni μECD detector (270 °C). Compound separation was completed using a fused silica DB-5 capillary column (60 m, 0.25 mm i.d., 0.25 μm film thickness) (J and W Scientific, Folsom, CA, U.S.A.) with H_2 as the carrier gas and 5% methane/95% argon makeup gas. Quantification

of OCs/PCBs was performed using an external standard. For quantification of $MeSO_2$-PCBs/-*p*,*p*′-DDE, an internal standard approach was employed based on peak area of the GC-μECD relative response factors. The OCs/PCBs and $MeSO_2$-PCBs/-*p*,*p*′-DDE were identified on the basis of their retention time on the GC-μECD DB-5 column, and verified by matching retention times with authentic standards. Using the same GC conditions and a 30 m DB-5 GC column, GC-mass spectrometry in the electron-capture, negative ion (ECNI) mode (SIM; $[M]^+$and $[M+2]^+$molecular ions) was used to confirm the isomer or structure identity of individual $MeSO_2$-PCB congeners and 3-$MeSO_2$-*p*,*p*′-DDE. Chromatographic data was interpreted using HP ChemStation Plus, Rev. A.07.01 (Hewlett-Packard, Palo Alto, CA, U.S.A.).

The following compounds were determined: 18 routinely-analyzed OCs and by-products (CHL-related compounds (ΣCHL: oxychlordane, *trans*-chlordane, nonachlor III (MC6), *trans*-nonachlor, *cis*-nonachlor, heptachlor epoxide), HCH isomers (ΣHCH: α-HCH, β-HCH, γ-HCH), CBzs (ΣCBz: 1,2,3,4-TeCBz, PnCBz, HCB), and DDT-related compounds (ΣDDT: *p*,*p*′-DDT, *p*,*p*′-DDE, *p*,*p*′-DDD), dieldrin, mirex, and octachlorostyrene). Nonachlor III co-elutes with *cis*-chlordane, however, the majority of this peak has been shown to be nonachlor III in polar bear fat (Muir et al., 1988; Norstrom et al., 1998). Furthermore, in the PCB-containing fraction F1, CB-99 was fully resolved from the portion of nonachlor III that has been found to elute in this fraction as well, and can co-elute when using a shorter 30 m DB-5 (or comparable) GC column. Fourty-two PCB congeners including co-elutions were determined ([IUPAC numbers (Ballschmiter and Zell, 1980) listed in order of their retention time] 31/28, 52, 49, 47/48, 44, 42, 71/41/64, 74, 70/76/98, 66/95, 56/60, 101, 99, 97, 81/87, 110, 151, 149, 118, 146, 153, 105, 141, 179, 163/138, 158, 178, 182/187, 183, 128, 174, 177,156 (note: CB-156 includes traces of CB-171/202 for Canadian and East Greenland samples as they co-elute on a DB 5 column), 200, 172, 180, 170/190, 201, 203/196, 208/195, 194 and 206. Methylsulfone metabolites were also determined, i.e., 3-$MeSO_2$-*p*,*p*′-DDE and 20 $MeSO_2$-PCB congeners (Σ$MeSO_2$-PCB: 3-/4-$MeSO_2$-CB-52, 3′-/4′-$MeSO_2$-CB-49, 3-/4-$MeSO_2$-CB-64, 3-/4-$MeSO_2$-CB-70, 3′-/4′-$MeSO_2$-CB-101, 3′-/4′-$MeSO_2$-CB-87 (note: 3′-$MeSO_2$-CB-87 may include traces of 5-$MeSO_2$-CB-110 as they co-elute on a DB 5 column), 4-$MeSO_2$-CB-110, 3-/4-$MeSO_2$-CB-149, 3′-/4′-$MeSO_2$-CB-132, 3′-/4′-$MeSO_2$-CB-141, 4′-$MeSO_2$-CB-174). The nomenclature of $MeSO_2$-PCBs has been abbreviated and simplified based on the systematic numbering technique applied to PCB congeners (Guitart et al., 1993; Maervoet et al., 2004).

2.3. Quality control

The results were within the accredited requirements for precision, linearity and sensitivity. Standard reference materials (SRMs) (1588a: organics in cod liver oil; 1945: organics in whale blubber) from the National Institute of Standards and Technology (Gaithersburg, MD, U.S.A.) were used to confirm the accuracy and reproducibility of the analytical methods. OC pesticide and PCB concentrations were within 15% of the SRM certified values, while $MeSO_2$-PCB concentrations were within 15% of the SRMs values reported by Hoekstra et al. (2003) and Sandala et al. (2004). The mean recoveries (±1 SD) of 1,3,5-tribromobenzene recovery standard and $MeSO_2$-PCB-IS recovery/internal standards were 91 ± 13% and 96 ± 15%, respectively. Concentrations of $MeSO_2$-PCBs and 3-$MeSO_2$-*p*,*p*′-DDE were recovery-corrected as a consequence of using the internal standard quantification approach. Generally, the method limit of quantification (MLOQ) for individual analytes of OCs, PCBs, $MeSO_2$-PCBs, and 3-$MeSO_2$-*p*,*p*′-DDE was based on a signal to noise ratio of 10, and was 0.01 ng g^{-1} lipid weight. The repeatability (inter-day comparison) of the GC-performance was tested by repeated injections (duplicates) of standard compounds and polar bear samples for each block of five samples analyzed. Method blank samples were also run with each block of five samples to assess for co-eluting interferences. Blank analysis indicated no significant interference co-elutions with analyte peaks, and duplicate standard compounds and polar bear fat samples demonstrated 5% or less variation of contaminant concentrations.

The chemical analyses were performed at the Great Lakes Institute for Environmental Research (GLIER) (University of Windsor, Windsor, ON, Canada), and additional chemical analyses were performed at the National Water Research Institute (NWRI) (Environment Canada, Burlington, ON, Canada). These labo-

ratories are certified as laboratories for determination of OC pesticides and PCBs according to the requirements of the Canadian Environmental Analytical Laboratory (CAEAL) program of the Canadian Standards Association, and are participants in the Northern Contaminant Program's Quality Assurance Program (Stokker, 2003).

2.4. Data analyses

Individual polar bears were grouped according to their location of capture or harvest by native hunters (Fig. 1). The present locations were within geographical areas that have been identified as distinct polar bear populations (Norstrom et al., 1988, 1998). This assessment was restricted to female polar bears because previous analyses indicated that there was less of an age effect on CHC concentrations, particularly highly-chlorinated PCBs, in adult females compared to males (Bernhoft et al., 1997; Norstrom et al., 1998; Dietz et al., 2004). However, samples from Alaska were comprised of a nearly equivalent number of males and females. Therefore, statistical analyses were performed using standardized analyte concentrations of male samples from Alaska to equivalent concentrations in females calculated from standardization coefficients reported by Norstrom et al. (1998). The variance due to the effect of differences in individual

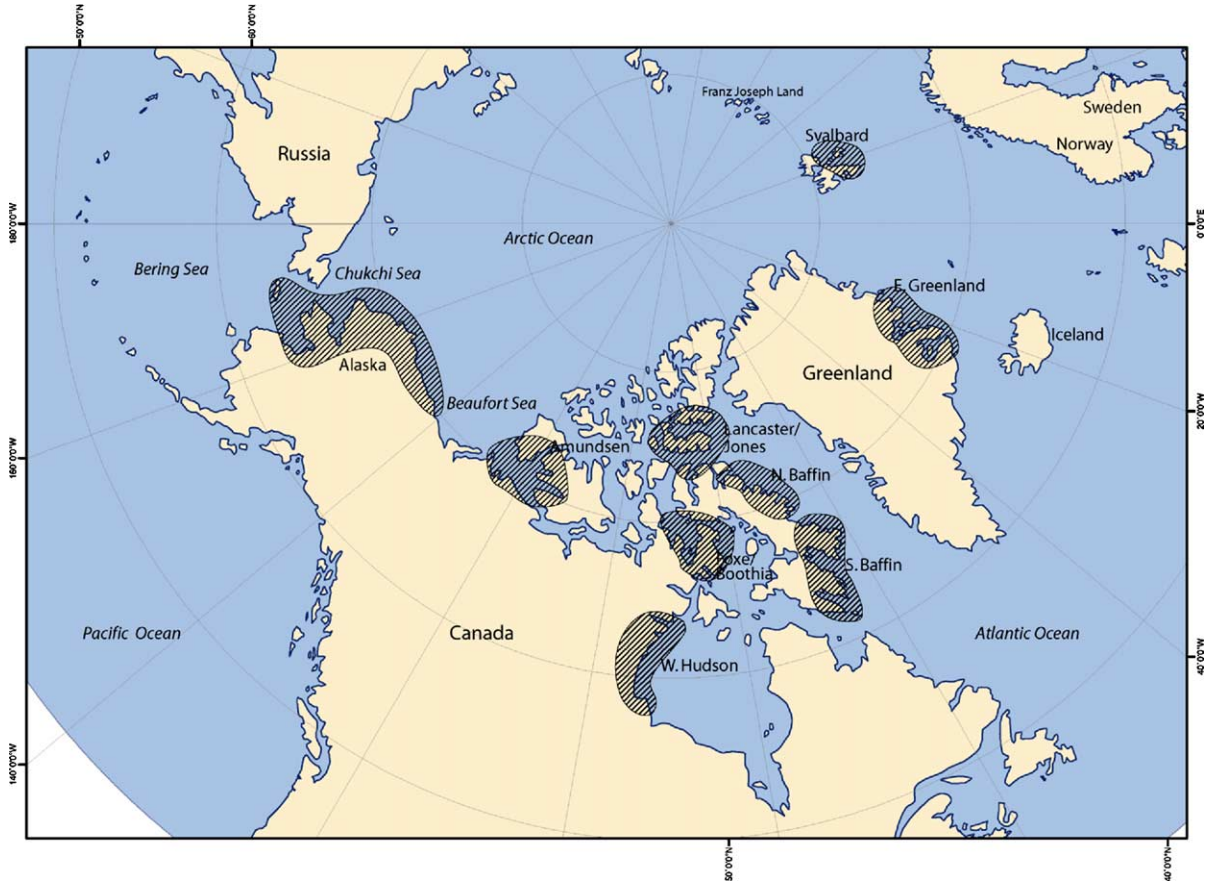

Fig. 1. Map of Arctic and Subarctic regions showing the nine sampling locations (hatched areas) of polar bears: Alaska, Amundsen Gulf, Western Hudson Bay, Foxe Basin/Gulf of Boothia, Lancaster Sound/Jones Sound, Northern Baffin Island, Southern Baffin Island, East Greenland, and Svalbard.

extractable lipid content in adipose tissue on OC, PCB, $MeSO_2$-PCB, and 3-$MeSO_2$-*p*,*p*′-DDE concentrations was minimized by using lipid-normalized concentrations (ng g^{-1} lipid weight). To avoid missing values in the data computation, analytes below the MLOQ were assigned a random value between 0 and the MLOQ.

Statistical analyses were carried out using the statistical package STATISTICA; version 6.1.409.0 (StatSoft, 2003). Statistical significance was set at $p \leq 0.05$. Variables that did not approximate the normal distribution using the Shapiro–Wilk's *W* test (Zar, 1999) were $\log_{10}$-transformed. As a result, concentrations of analytes and age of the bears were $\log_{10}$-transformed to meet the assumptions of the statistical tests. Summary statistics for any analytes were computed only if at least two-thirds of the samples contained detectable concentrations.

The geographical difference in congener patterns in polar bear adipose tissue across the populations was examined by determining the relative proportion of 15 OCs to Σ_{15}OC (oxychlordane, *trans*-chlordane, nonachlor III (MC6), *trans*-nonachlor, *cis*-nonachlor, heptachlor epoxide, α-HCH, β-HCH, PnCBz, HCB, *p*,*p*′-DDT, *p*,*p*′-DDE, *p*,*p*′-DDD, mirex, and dieldrin) and the major 20 PCB congeners to Σ_{20} PCB (CB-31/28, 47/48, 56/60, 101, 99, 118, 146, 153, 163/138, 182/187, 183, 128, 156, 200, 180, 170/190, 201, 203/196, 194, and 206), and investigated using a principal component analysis (PCA) (Hair et al., 1998). On average Σ_{20}PCB made up 94% (range 87−99%) of Σ_{42}PCB. Relative proportions of individual OCs and PCBs were arcsin-transformed and PCA was performed on the correlations. Principal components (PCs) with eigenvalues above 1 were considered to account for a significant contribution to the total variance according to the latent root criterion (Hair et al., 1998). Factor loading rotation was computed using VARIMAX to give a clearer separation of the PCs (Hair et al., 1998). Analytes with high correlation coefficients (i.e. factor loadings) ($r > \pm 0.60$) on one PC and with relatively low correlation coefficients ($r < \pm 0.35$) on any other PCs were considered significant (Hair et al., 1998).

Correlations and geographical differences in OC, PCB, $MeSO_2$-PCB and 3-$MeSO_2$-*p*,*p*′-DDE concentrations, and PCs were tested by a General Linear Model (GLM; Type III Sums of Squares) (StatSoft, 2003). Age contributed significantly to the variation of ΣOC (sum of all OC compounds and by-products), Σ_{42}PCB, and Σ$MeSO_2$-PCB concentrations in a multivariate analysis of variance for samples where all analytes were analyzed (GLM; $F_{3,80} = 7.9$, $p < 0.001$). It has been shown that concentrations of some lipophilic CHCs, notably CHLs and PCBs, are higher in sub-adult polar bears compared to adults, particularly in cubs of the year, because of inheritance from lactational transfer from mother to cub (Polischuk et al., 2002). The significant number of sub-adult polar bears <5 years of age in the present dataset probably accounts for most of the age effect. Given the small sample size of some populations of polar bears, it was not possible to divide the dataset further in age-groups (i.e. sub-adult and adult). Therefore, the analysis of covariance were performed on age-adjusted means using the Tukey's post-hoc test for unbalanced sample sizes (StatSoft, 2003). Levene's test was further computed on variables to test for homogeneity of variance between populations; an assumption of the analysis of covariance model (StatSoft, 2003). Correlations are expressed using either *partial r*, i.e. the correlation between two variables that remained after controlling for age, or the Pearson correlation coefficient *r*.

Two-point temporal comparisons were made by comparing present 1996–2002 age-adjusted mean concentrations with mean concentrations reported in earlier studies of female polar bear adipose tissue samples from comparable populations.

3. Results and discussion

3.1. Concentrations and congener patterns

Among the organochlorine (OC) pesticides and by-products analyzed, CHLs were major contaminants in adipose tissue of all populations of polar bears, and were largely dominated by oxychlordane, which made up on average two-thirds of ΣCHL (Table 2). Likewise, CHLs were the major OCs in subcutaneous fat of polar bears collected from Svalbard during 1990−1994 (Bernhoft et al., 1997), similar to reports on concentrations by Norstrom et al. (1998) in fat of polar bears sampled during 1989−1993 from Alaska, Canada, East Greenland, and Svalbard. In the present study, ΣCHL concentrations were lowest in polar

bears from Alaska (Tukey's test; $p<0.05$), representing approximately 40% of age-adjusted mean ΣCHL determined in the eight other populations (Table 3). Similarly, earlier studies have reported lower ΣCHL concentrations in polar bear fat collected from a comparable region (northeastern Alaska and Bering/Chukchi Sea) (Muir et al., 1988; Norstrom et al., 1988, 1998). Nevertheless, omitting the results from Alaska, ΣCHL concentrations had a fairly uniform distribution across the present polar bear range where no significant difference was observed (Fig. 2). This uniformity in the distribution of ΣCHL concentrations was in accord with findings for polar bear samples (Norstrom et al., 1998), ringed seal (*Phoca hispida*) samples (Weis and Muir, 1997), and geographical variation of ΣCHL concentrations in air and seawater from the Northern Hemisphere (Iwata et al., 1993).

ΣHCH concentrations showed the steepest negative west–east gradient across the populations studied (Fig. 2). ΣHCH concentrations were significantly highest in Alaska bears compared to Western Hudson Bay and to populations east of Lancaster Sound/Jones Sound (Tukey's test; $p<0.02$), and lowest in bears from Svalbard (Tukey's test; $p<0.001$). There was a six-fold difference in age-adjusted mean ΣHCH concentrations between bears from Alaska and Svalbard (data not shown). Muir and Norstrom (2000) and Norstrom et al. (1988) also reported the highest ΣHCH concentrations in polar bear fat samples from Alaska (Bering/Chukchi Sea). This may indicate an ongoing contribution of HCHs from China, southeastern Asia, and North America (de March et al., 1998). The west−east geographical trend for ΣHCH was in general agreement with results of polar bears spanning the regions from Svalbard eastwards to the Chukchi Sea (Lie et al., 2003), measurements of HCHs in seawater (de March et al., 1998; Macdonald et al., 2000), and results of ringed seals from the Canadian Arctic eastwards to the Russian Arctic (Muir and Norstrom, 2000). Furthermore, latitude was negatively correlated with the α-HCH : ΣHCH ratio and was the most pronounced latitudinal gradient measured for this study (Fig. 3). No correlation was found between longitude and the α-HCH : ΣHCH ratio. The contribution of the more water-soluble α-HCH to ΣHCH, relative to β-HCH, was thus highest at the southernmost populations of the distribution range of polar bears.

The spatial distribution of ΣCBz and dieldrin was fairly uniform across the study area, similar to the distribution of mirex when omitting the results from East Greenland where mirex was below the MLOQ in 70% of the samples (Table 2). These figures may suggest that ΣCBz, dieldrin, and mirex are relatively well equilibrated in surface waters throughout the Arctic and Subarctic. Conversely, ΣDDT concentrations increased gradually from west to east (GLM; partial $r=0.36$, $p<0.001$), and were highest in polar bear samples from East Greenland (Tukey's test; $p<0.05$), although not significantly higher than samples from Svalbard and Western Hudson Bay (Fig. 2). Results for *p,p′*-DDE (Table 2), which was the dominant component of ΣDDT (90% of ΣDDT on average), were not in accord with results reported by Norstrom et al. (1998) who found low concentrations of *p,p′*-DDE in female polar bears from Svalbard, similar to concentrations in the eastern Canadian Arctic.

Among the total suite of OCs, PCBs, and methyl sulfones analyzed in this present study, Σ_{42}PCB dominated ostensibly the composition pattern across the investigated populations, except for Foxe Basin/Gulf of Boothia and Lancaster Sound/Jones Sound, where ΣCHL accounted for the highest proportion (Table 2). CB-153 was consistently the major PCB congener in fat samples, followed by CB-180,-163/138,-170/190, -99, and -194, which combined comprised 82% (range 69−91%) of Σ_{42}PCB. Σ_{42}PCB concentrations increased most steeply with longitude (GLM; partial $r=0.56$, $p<0.001$) (Fig. 2), and increased also with latitude (GLM; partial $r=0.29$, $p=0.003$). A west-to-east gradient for ΣPCB concentrations has also been noted in earlier regional assessments for polar bear samples (Letcher et al., 1995; Muir and Norstrom, 2000; Andersen et al., 2001), thus indicating a larger scale atmospheric/oceanic transport of PCBs from European and Eurasian sources to the Greenland Sea/Barents Sea (e.g. de March et al., 1998). The difference in age-adjusted Σ_{42}PCB between Foxe Basin/Gulf of Boothia (lowest) and Svalbard (highest) was six-fold (Table 3), which was higher relative to results reported by Norstrom et al. (1998) who made a similar comparison. Nevertheless, the general increase in Σ_{42}PCB with longitude varied between congeners. This was reflected by a decreasing CB-99 : CB-180 ratio from

Table 2
Unadjusted geometric means[a] (GM) with 95% confidence limits (CL), and ranges (*R*) for individual concentrations (ng g^{-1} lipid weight) or sums (Σ) of a selection of organochlorine pesticides and by-products, ΣPCB, Σ$MeSO_2$-PCB, and 3-$MeSO_2$-*p*,*p*′-DDE in adipose tissue of polar bears from nine Arctic and Subarctic populations

		Alaska (males)[b]	Alaska (females)	Amundsen Gulf	W. Hudson Bay	Foxe Basin/Gulf of Boothia	Lancaster Sound/ Jones Sound	N. Baffin Island	S. Baffin Island	E. Greenland	Svalbard
Oxychlordane	GM	313	843	1340	1372	1355	1393	1697	1259	1071	1200
	CL	54.4–870	634–1086	998–2000	1126–1815	869–2227	890– 2096	1380–2154	1078–1780	639–2032	972– 1668
	R	90.3–1351	593–1054	531–2766	804–2657	613–2953	691–2386	1051–2713	315–2498	433–6022	577–2826
ΣCHL	GM	502	1095	2007	2345	2061	1943	2457	1819	1776	1517
	CL	155–1186	870–1350	1527–2880	2009–2874	1371–3337	1306–2790	1942–3198	1553–2461	1190–3008	1234–2081
	R	171–1770	836–1316	844–3831	1559–3979	970–3985	1104–3141	1672–4578	582–3607	845–8132	706–3648
ΣHCH	GM	593	404	379	260	498	381	297	280	137	71.3
	CL	363–909	298–525	278–591	231–302	280–850	268–530	265–336	242–352	115–182	60.1–91.1
	R	398–1269	332–550	136–802	183–399	295–1310	216–551	208–362	116–514	80.6–306	41.0–144
HCB	GM	84.5	85.7	75.5	75.3	87.3	107	152	87.1	60.0	90.4
	CL	52.0–138	6.84–206	57.4–104	56.7–117	34.5–173	80.1–139	73.2–309	70.2–126	37.9–115	71.3–143
	R	35.1–157	41.7–230	43.9–146	39.3–229	46.3–305	74.9–146	76.7–620	33.2–249	25.5–311	37.9–229
ΣCBz	GM	113	118	113	97.5	127	148	191	111	79.1	105
	CL	70.4–181	26.8–247	90.0–148	76.0–141	73.7–208	119–182	107–344	91.8–150	55.6–132	83.9–160
	R	58.7–203	70.0–277	71.1–190	55.9–257	73.4–329	111–186	108–656	42.0–275	36.5–323	49.0–248
p,*p*′-DDE	GM	73.8	131	90.4	200	73.2	54.7	123	112	268	190
	CL	46.0–124	76.0–200	56.2–150	171–250	16.9–197	32.9–83.7	0.00–452	90.6–155	216–424	165–233
	R	28.2–129	90.9–204	45.6–273	94.5–349	23.8–337	36.6–100	60.7–1157	63.9–280	82.8–830	91.0–304
ΣDDT	GM	92.2	149	103	210	88.3	65.2	137	126	309	209
	CL	59.8–150	83.9–230	63.6–173	178–267	17.1–240	38.7–102	0.00–507	102–174	249–490	180–260
	R	28.2–174	105–242	49.0–312	94.5–380	26.0–428	41.0–119	65.9–1297	77.6–329	95.4–960	96.9–352

Σ_{42}PCB	GM	2980	2838	2174	2579	1138	1847	2802	2709	5414	5972
	CL	2012–4416	1968–3881	1486–3460	2211–3172	647–2045	1197–2701	2144–3912	2372–3325	4011–8584	4637–9129
	R	1718–4784	1872–3939	943–5458	1631–4419	514–2958	1140–3153	1406–4870	1269–4266	2359–20,485	2868–16,043
AMAP Σ_{10}PCB[c]	GM	1971	2015	1450	1728	737	1219	1863	1879	3724	4032
	CL	1323–2909	1415–2732	992–2312	1476–2140	397–1375	772–1811	1415–2631	1637–2329	2703–6018	3107–6277
	R	1205–3232	1321–2773	612–3630	1115–3037	324–2053	753–2132	924–3259	861–3112	1597–14,885	1,895–11,016
$\Sigma MeSO_2$-PCB	GM	151	NA	129	109	95.9	124	178	99.4	NA	198
	CL	68.6–254		97.6–201	88.2–143	71.7–130	89.0–171	152–216	103–177		162–279
	R	111–293	NA	38.2–269	70.5–255	59.8–165	63.2–170	109–244	0.91–273	NA	76.4–456
3-$MeSO_2$-*p*,*p*′-DDE	GM	0.26	NA	1.65	0.74	0.65	1.49	–	1.55	NA	–
	CL	0.09–1.12		1.27–2.26	1.04–2.00	0.10–2.95	1.00–2.17		1.56–3.23		
	R	ND–1.04	NA	0.95–2.95	ND–3.29	ND–5.56	0.84–2.29	ND–3.02	ND–5.50	NA	ND–2.48
Mirex	GM	9.28	8.35	6.87	11.2	1.74	5.59	7.84	4.65	–	10.2
	CL	4.38–16.4	6.20–10.8	4.43–11.7	9.29–14.2	0.00–10.7	1.92–11.3	5.77–11.6	3.97–6.15		8.49–17.8
	R	6.46–24.8	6.46–11.0	2.97–19.5	6.85–22.5	ND–20.8	2.86–14.8	4.39–16.0	2.36–8.09	ND–20.3	1.93–27.3
Dieldrin	GM	90.6	111	173	152	198	65.2	171	55.4	171	160
	CL	61.2–134	74.1–156	125–254	122–226	62.4–470	19.7–170	121–295	62.6–143	148–212	134–203
	R	49.7–153	68.0–154	99.3–351	51.1–360	73.8–794	12.6–188	51.1–426	1.75–238	110–328	89.3–340
Octachlorostyrene	GM	12.7	14.3	ND				ND		13.1	16.4
	CL	7.21–21.3	9.46–20.2		–	–	–		–	10.5–18.3	11.9–25.1
	R	6.41–26.7	9.73–20.4	ND	ND–14.6	ND–6.77	ND–16.1	ND	ND–9.90	5.98–38.1	8.79–58.5

ND: not detected.

NA: not analyzed.

[a] Geometric means were computed only when at least two-thirds of the samples contained detectable concentrations.

[b] Concentrations not standardized to equivalent concentrations in females.

[c] AMAP Σ_{10}PCB: CB-31/28, 52, 101, 105, 118, 138, 153, 156, and 180.

Table 3
Two-point temporal comparisons between present 1996–2002 age-adjusted mean concentrations (ng g^{-1} lipid weight) of a selection of organochlorine pesticides, Σ_{42}PCB, ΣMeSO$_2$-PCB, and 3-MeSO$_2$-*p*,*p*′-DDE in adipose tissue of female polar bears and concentrations reported by Norstrom et al. (1998) and Letcher et al. (1995) for fat samples collected during 1989–1993 from comparable Arctic and Subarctic populations

	ΣCHL			*p*,*p*′-DDE			Dieldrin			Σ_{42}PCB			ΣMeSO$_2$-PCB			3-MeSO$_2$-*p*,*p*′-DDE		
	Norstrom et al. (1998)	This study	Diff. (%)	Norstrom et al. (1998)	This study	Diff. (%)	Norstrom et al. (1998)	This study	Diff. (%)	Norstrom et al. (1998)	This study	Diff. (%)	Letcher et al. (1995)	This study	Diff. (%)	Letcher et al. (1995)	This study	Diff. (%)
Alaska [R1, R2][a]; [1][b]	1206	852	−29	195	94	−52	37	99	168	3797	2395	−37	138	NA		0.8	NA	
Amundsen Gulf [R4][a]; [2][b]	2146	1752	−18	93	90	−3	80	164	105	3772	2040	−46	247	117	−53	1.8	1.2	−31
W. Hudson Bay [R12][a]; [10][b]	2738	2561	−6	302	200	−34	107	158	48	3965	2687	−32	218	117	−46	2.1	0.9	−57
Foxe Basin/Gulf of Boothia [R8][a]; [6][b]	2679	1743	−35	97	73	−25	216	185	−14	2245	1052	−53	145	85	−41	0.6	0.5	−23
Lancaster Sound/Jones Sound [R7][a]; [5][b]	2753	2070	−25	197	55	−72	122	67	−45	3087	1902	−38	215	131	−39	1.9	1.7	−8
N. Baffin Island [R9][a]; [7][b]	2339	2266	−22	278	123	−59	138	166	20	3244	2698	−46	287	168	−41	3.6	ND	
S. Baffin Island [R14][a]; [8][b]	2825	1794	−36	268	112	−58	174	55	−68	4564	2692	−41	470	99	−79	4.4	1.5	−65
E. Greenland [R15][a]; [12][b]	5044	1978	−61	278	268	−4	384	179	−53	22,419	5693	−75	633	NA		11	NA	
Svalbard [R16][a]	3162	1742	−45	253	190	−25	189	169	−11	12,775	6369	−50	NA	221		NA	ND	

ND: not detected.
NA: not analyzed.
[a] Comparable locations reported by Norstrom et al. (1998) for populations of female polar bears (adipose tissue samples).
[b] Comparable locations reported by Letcher et al. (1995) for populations of polar bears (pooled adipose tissue samples).

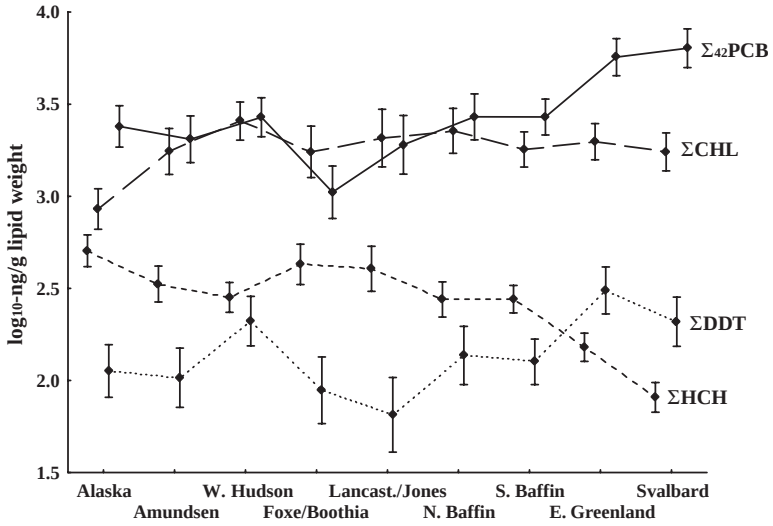

Fig. 2. Age-adjusted mean ΣCHL, ΣHCH, ΣDDT, and Σ_{42}PCB concentrations (log_{10}-transformed ng g^{-1} lipid weight) plotted with 95% confidence intervals (vertical bars) in adipose tissue of female polar bears from nine Arctic and Subarctic populations listed in order of longitude.

west to east (Fig. 3), thus showing a greater increase of higher-chlorinated and recalcitrant PCB congeners eastwards, as also reported by Letcher et al. (1995) and Andersen et al. (2001). The CB-99 : CB-180 ratios for samples across the Arctic reflect the regional signatures and atmospheric input originating from different types of technical PCB formulations. For example, eastern Arctic inputs are likely influenced by the Soval PCB mixture, which is used mainly in Russia. However, the average weight percent distribution of the PCB isomer groups in the Soval mixture has been reported to be fairly close to Aroclors 1254 and 1242 (Ivanov and Sandell, 1992). A similar but non-significant trend for CB-99 : CB-180 ratio was observed along latitude (GLM; partial $r=-0.17$, $p=0.08$), demonstrating greater local input of relatively volatile and less-persistent PCBs in southern regions of North America.

The dominant $MeSO_2$-PCB congeners in polar bear fat were consistently 3′-/4′-$MeSO_2$-CB-101, 4′-$MeSO_2$-CB-87 and 4-$MeSO_2$-CB-149, which, when combined, accounted for 49% (range 20−63%) of Σ$MeSO_2$-PCB. Σ$MeSO_2$-PCB concentrations were positively correlated to Σ_{42}PCB (Pearson; $r=0.44$, $p<0.001$), and followed consequently a similar increasing trend from west to east, and from south to north. Similarly, 3-$MeSO_2$ *p*,*p*′ DDE was correlated to its precursor, *p*,*p*′ DDE (Pearson; $r=0.46$, $p<0.001$). Due to the high variance within populations, no significant geographical difference was found for concentrations of Σ$MeSO_2$-PCB and 3-$MeSO_2$-*p*,*p*′-DDE between the populations examined (Table 2).

Principal component analysis (PCA) revealed notable geographical differences in the congener make up of 15 compounds of OCs to Σ_{15}OC, and 20 major PCBs to Σ_{20}PCB. PCA yielded 3 principal components (PCs) with eigenvalues greater than 1 for OCPs and PCBs examined separately (Table 4). The PCs extracted accounted for a relatively low cumulative percentage of the total variance (Table 4). In order to investigate regional differences, mean factor scores for the nine populations of polar bears were plotted using PC1 and PC2, which explained 43% and 55% of the total variance for OCs and PCB group composition, respectively (Fig. 4). For OCs, PC1 was positively correlated with *trans*-nonachlor, *cis*-nonachlor *p*,*p*′-DDE, *p*,*p*′-DDD, and *p*,*p*′-DDT, and PC2 was negatively correlated with PnCBz, α-HCH, and β-HCH (Table 4). For PCBs, PC1 was positively correlated with CB-47/48 and -118, and negatively correlated with CB-180 (Table 4). Moreover, PC2 was positively correlated with CB-146, -163/138, -183, and -203/196 (Table 4). The plot of PC1 vs PC2 for OCs demonstrates that Alaska was distinguished by

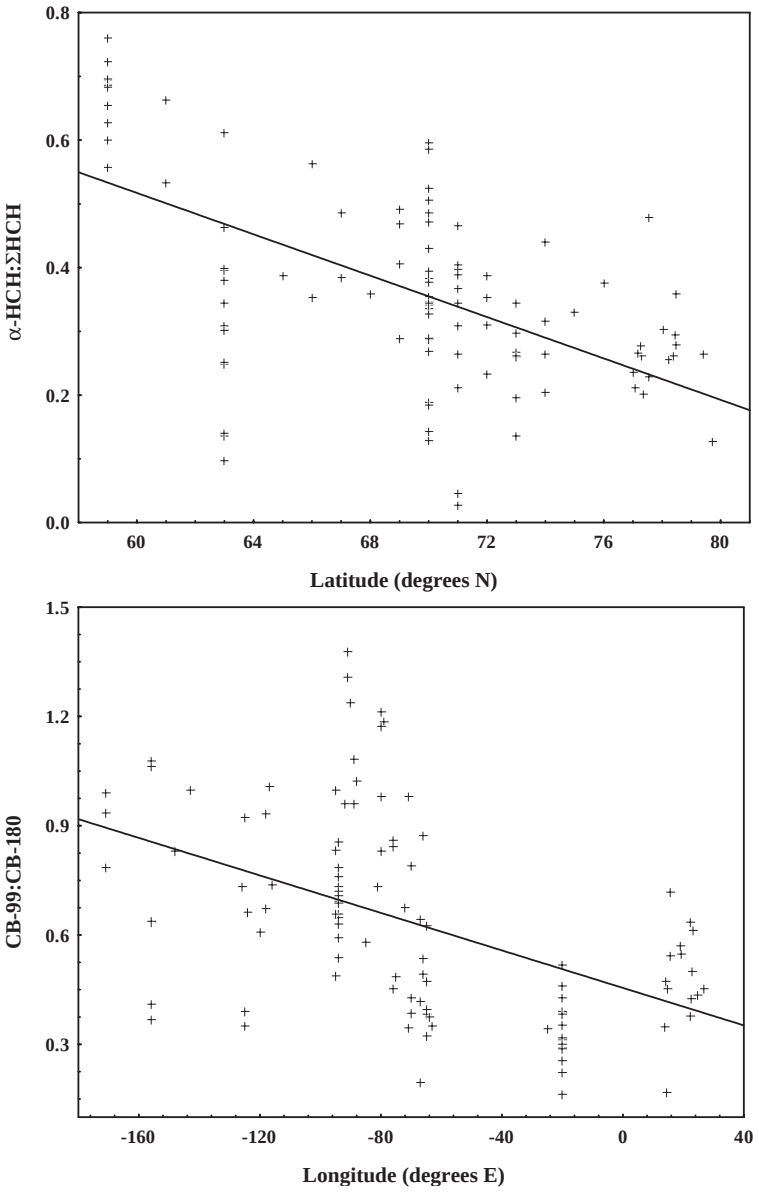

Fig. 3. Age-adjusted correlations between the α-HCH:ΣHCH (GLM; partial $r=-0.59$, $p<0.001$) and CB-99:CB-180 (GLM; partial $r=-0.44$, $p<0.001$) ratios, and latitude and longitude, respectively, for female polar bear adipose tissue samples from nine Arctic and Subarctic populations.

high proportions of relatively volatile PnCBz and HCHs (α, β), and low proportions of oxychlordane (Fig. 4). East Greenland and Svalbard were characterized by high proportions of persistent DDT-related compounds, nonachlor (*cis, trans*), and oxychlordane (Fig. 4). For PCBs, PCA plot scores shows the bears from Alaska and Foxe Basin/Gulf of Boothia were distinguished by high proportions of lower-chlorinated PCB congeners (tri- to penta-chlorinated), whereas bears from East Greenland were characterized by higher proportions of higher-chlorinated and more persistent PCBs (hepta- to nona-chlorinated) (Fig. 4).

Table 4
Factor loadings (i.e. correlation coefficients) of a selection of individual organochlorine pesticides (OCs) and by-products, and PCBs to the three principal components (PCs) 1, 2 and 3, extracted from arcsine-transformed proportions of 15 individual OCs to Σ_{15}OCs, and 20 major PCB congeners to Σ_{20}PCB

	PC1	PC2	PC3
Nonachlor III (MC6)	−0.17	0.25	−0.60
trans-Nonachlor	0.71	0.07	−0.18
cis-Nonachlor	0.78	−0.17	0.04
Heptachlor epoxide	0.02	−0.04	−0.84
α-HCH	−0.21	−0.68	−0.35
β-HCH	0.10	−0.75	0.33
PnCBz	−0.13	−0.79	0.11
p,p′-DDE	0.79	0.34	0.05
p,p′-DDD	0.72	0.07	0.31
p,p′-DDT	0.79	0.20	0.18
Eigenvalues	3.84	2.64	1.94
% total variance	25.6	17.6	12.9
CB-31/28	0.24	−0.29	−0.67
CB-47/48	0.79	0.03	−0.18
CB-118	0.65	0.33	0.01
CB-146	0.30	0.84	0.31
CB-163/138	0.32	0.78	−0.26
CB-183	−0.09	0.85	−0.08
CB-128	0.25	0.15	−0.83
CB-180	−0.88	0.05	0.34
CB-201	−0.15	0.15	0.73
CB-203/196	−0.20	0.77	0.30
Eigenvalues	6.6	4.32	2.4
% total variance	33	21.6	12

Only significant compounds are tabulated, i.e. compounds with correlation coefficients $> \pm 0.60$ on one PC and $< \pm 0.35$ on any other PCs. Eigenvalues and percentages of the total variance explained by each PC are shown.

3.2. Temporal comparisons

Temporal trends of OC pesticide, PCB, and methyl sulfone metabolite concentrations in polar bear adipose tissue could not be determined from the present assessment as published data for comparable populations, omitting a comprehensive dataset for Hudson Bay, were measurements over a limited period of time. Therefore, two-point comparisons were made between present 1996−2002 age-adjusted mean concentrations and those reported in earlier surveys of female polar bear fat samples collected at similar regions. However, direct comparisons between means did not allow for the correction of variation of concentrations within each polar bear populations or confounding factors known to influence contaminant concentrations, e.g. sex ratio, age composition, nutritional and reproductive status, feeding habits and methodological differences (i.e. sampling substrate, analytical techniques, number of congeners measured as sums). Studies on polar bears have reported significant oscillations of CHC concentrations throughout the years, which are quite often related to differences in biological conditions of the individuals rather than variations of CHC concentrations in the abiotic environment or in the food chain of polar bears. The likelihood of misinterpreting the data increases by not adjusting for confounding factors that vary systematically over time in a non-standardized dataset (Henriksen et al., 2001). As a result, substantial errors are introduced by means of direct comparisons between surveys of contaminants for polar bears. Henriksen et al. (2001) recommended that monitoring studies of polar bears should be based on a 14-year sampling period, and that sampling should take place annually at the same location.

Nevertheless, age-adjusted mean ΣCHL concentrations in the present 1996−2002 study were lower (18−61%) compared to 1989−1993 data of female polar bear fat samples from comparable populations (Norstrom et al., 1998), except for Western Hudson Bay where mean ΣCHL was nearly unchanged (Table 3). Our figures for Western Hudson Bay (2001−2002) showed great consistency with temporal trends of CHCs in Hudson Bay polar bear adipose tissue throughout the 1990s that showed a non-decreasing trend of ΣCHL (Norstrom, 2000). Norstrom et al. (1988) noted that ΣCHL concentrations appeared to have declined throughout the Arctic and Subarctic during the 1980s, especially in Hudson Bay, suggesting that current CHL concentrations in this particular region, from the 1990s and onwards, may have leveled off. A comparison between the 1990-dataset and the data for samples collected during 1999−2001 from East Greenland female polar bears revealed a 60−75% decrease in age-adjusted mean ΣCHL, which was in accord with the 61% decline observed in the present study for the same location (Table 3).

Present age-adjusted mean concentrations of α-HCH and β-HCH for Western Hudson Bay female bears fell within the same range as reported for 1991−1999 in the same region (data not shown) (Norstrom, 2000). Norstrom (2000) reported no significant change in ΣHCH concentrations for Hudson Bay polar bear fat during the last decade, mostly due

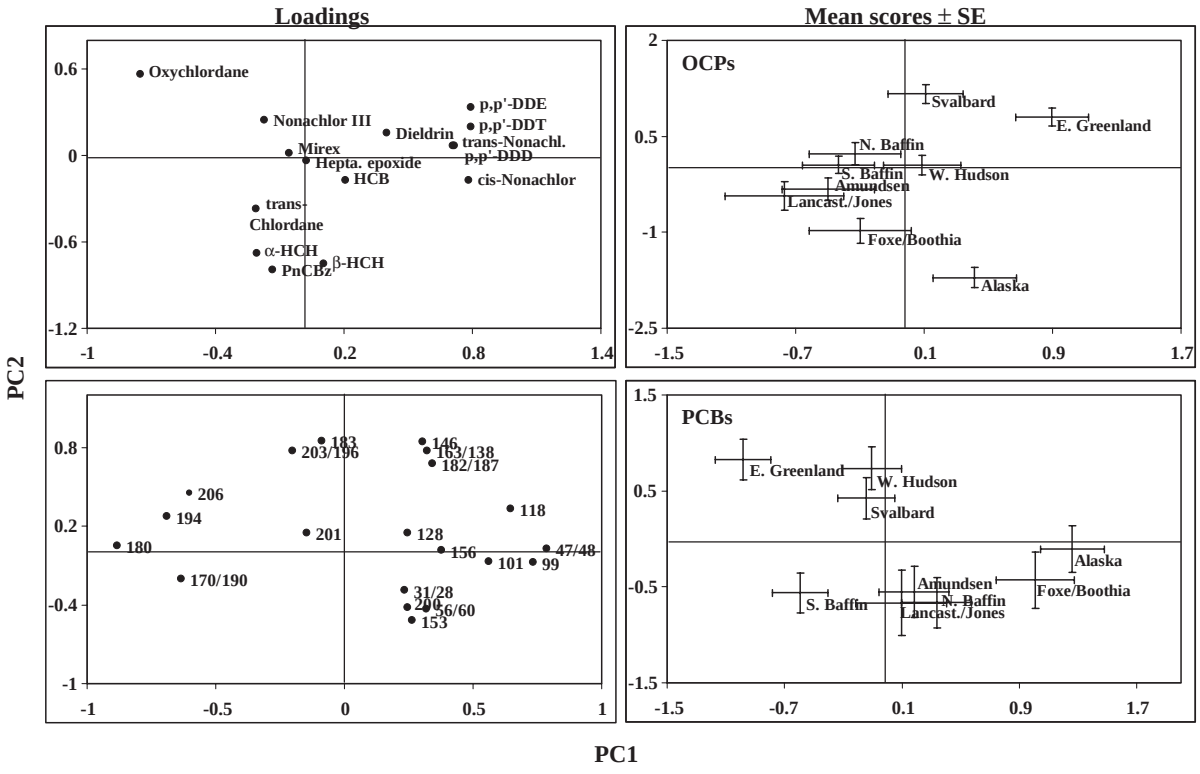

Fig. 4. Plots of factor loadings of 15 organochlorine (OC) pesticides and by-products and 20 major PCB congeners, and mean factor scores (± 1 standard error (SE) as vertical and horizontal bars) on the first two principal components (PCs). PC1 and PC2 combined described 43% and 55% of the total variance for OCPs and PCBs, respectively.

to the general increase of β-HCH proportions to ΣHCH over the last 30 years. In contrast, Dietz et al. (2004) reported a roughly 2.5-fold decrease of ΣHCH in polar bear fat collected from East Greenland between 1990 and 1999−2001, which was consistent with the decline observed in plasma concentrations of polar bears from Svalbard during 1991−1996 (AMAP, 2004). Long-range transport of HCHs from European/Eurasian sources to Svalbard and East Greenland may have declined to a larger extent relatively to regional inputs from North America to the Canadian Subarctic and Arctic.

Age-adjusted means of *p*,*p*′-DDE concentrations for female polar bears were lower (25−72%) than concentrations reported at comparable regions for 1989−1993 (Norstrom et al., 1998), although this tendency was not consistent for all populations as *p*,*p*′-DDE concentrations for Amundsen Gulf and East Greenland were comparatively unchanged (3−4% decrease) (Table 3). According to Norstrom (2000), there was no clear sign of decreasing concentrations of *p*,*p*′-DDE over the last 10 years in polar bear fat samples from Hudson Bay. In fact, there has been a tendency for increasing ΣDDT concentrations in Hudson Bay bears since 1995. Present 2001−2002 age-adjusted mean concentrations of ΣDDT for Western Hudson Bay bears (Table 3) were approximately 20% of those during the period of active DDT use pre-1970, and approximately 30% lower than last measured in 1999 by Norstrom (2000). On the other hand, concentrations of ΣCBz, which have previously been documented to have decreased significantly in polar bear samples from Hudson Bay since 1991 (Norstrom, 2000), were inconsistent with the present findings from the same location where concentrations were literally the same as before 1995 (Table 2). A comparison of our results for dieldrin with comparable polar bear female populations with the 1989−1993

dataset (Norstrom et al., 1998) showed that changes in age-adjusted mean concentrations varied from a 168% increase in Alaska female bears to a 68% decrease in Southern Baffin Island bears (Table 3). Irregularities in the oscillations of dieldrin concentrations were also observed by Norstrom (2000) for Hudson Bay bears monitored throughout the 1990s. The decline (57−73%) of age-adjusted mean dieldrin concentrations noted by Dietz et al. (2004) for East Greenland sub-adult and adult female polar bear adipose tissue samples collected between 1999−2001 and 1990 was consistent with our results showing a 53% decrease for this population (Table 3). Concentrations of dieldrin in polar bear fat may be more influenced by biological factors such as clearance rate, fasting cycles, and reproductive status. As a result, Norstrom et al. (1998) argued that the polar bear may not be a reliable biomonitoring species for concentrations of dieldrin.

A comparison between Σ_{42}PCB concentrations measured during the 1989−1993 assessment of female polar bear fat samples (Norstrom et al., 1998) and the current age-adjusted mean Σ_{42}PCB concentrations revealed a consistent decrease (32−75%) for comparable populations (Table 3). Similarly, for polar bear plasma samples collected at Svalbard, Henriksen et al. (2001) reported a 36% decrease of CB-153 concentrations between surveys of 1991−1994 and 1995−1998. In addition, Dietz et al. (2004) reported a 78% decrease in age-adjusted mean Σ_{42}PCB concentrations in fat samples of East Greenland polar bears between the 1999−2001 and 1990-datasets, which was virtually the same decline observed in the present study (75%) for this population (Table 3). Concomitantly, Norstrom (2000) observed a statistically significant downward trend of ΣPCB concentrations in Hudson Bay polar bears throughout the 1990s. Present 2001−2002 age-adjusted mean Σ_{42}PCB concentrations for Western Hudson Bay (Table 3) was nevertheless unchanged compared to the 1999 data for Hudson Bay bears (Norstrom, 2001). Norstrom (2001) predicted equilibrium with global distribution of PCBs in Hudson Bay and argued that further decreases in concentrations may be slower than observed during the 1980s. Presently, datasets available on temporal trends of PCBs in polar bear fat are in general agreement, suggesting that PCB concentrations have declined gradually and consistently in the Arctic and Subarctic over the last decade.

Similar to Σ_{42}PCB, age-adjusted mean Σ$MeSO_2$-PCB concentrations in polar bear fat were consistently lower (39−79%) relative to concentrations in composite adipose tissue samples collected during 1989−1993 from comparable populations in Alaska and Canada (Letcher et al., 1995) (Table 3). In fact, there was a consistent and proportional decrease between Σ_{42}PCB and Σ$MeSO_2$-PCB concentrations since the last comprehensive studies (1989−1993) on spatial trends of CHCs and methyl sulfones in female polar bear fat (Letcher et al., 1995; Norstrom et al., 1998). This was also reflected by a virtually unchanged mean (± standard deviation) Σ$MeSO_2$-PCB : ΣPCB ratio (0.06 ± 0.03) over approximately 10 years (Letcher et al., 1995).

3.3. Implications of current concentrations

Despite the high tissue and blood concentrations of CHCs in polar bears compared to other arctic animals, polar bears are documented to have a superior biotransformation capacity towards this class of anthropogenic compounds (Muir et al., 1988; Letcher et al., 1998). The high biotransformation capacity of polar bears results in variable retention and accumulation of CHC metabolites. Some CHC metabolites may have biological effects in organisms. Persistent $MeSO_2$-PCBs and 3-$MeSO_2$-*p*,*p*′-DDE have been reported in polar bear fat and blood, and the retained chlorinated phenolics hydroxylated (OH))-PCBs, 4-OH-heptachlorostyrene, and pentachlorophenol have been reported in polar bear blood or plasma (Letcher et al., 1995, 1998; Sandala et al., 2004; Sandau, 2000). These methyl sulfone and chlorinated phenolic contaminants have demonstrated biological activity from an endocrine standpoint from in vitro and in vivo studies (Brouwer et al., 1998; Letcher et al., 2000, 2002; Sandau, 2000). A possible causative linkage has been established between concentrations of several CHCs and their metabolites, and detrimental health effects reported for polar bears (de March et al., 1998; AMAP, 2004; Fisk et al., 2005–this issue; Lie et al., 2004a, 2005). Σ_{42}PCB concentrations in adipose tissue of some individuals sampled from Western Hudson Bay, East Greenland, and Svalbard exceed the threshold

concentrations associated with the NOEL for kit survival in mink (AMAP, 2004). Moreover, Σ_{42}PCB concentrations in polar bears from East Greenland and Svalbard surpass in some cases those of harbour seal (*Phoca vitulina*) blubber linked to immunosuppression and depressed vitamin A (de March et al., 1998). Comprehensive risk assessment studies showed that CHC concentrations were significantly correlated to e.g., immunological and endocrine biomarkers in polar bears from Svalbard (Skaare et al., 2002; AMAP, 2004; Fisk et al., 2005–this issue; Bernhoft et al., 2000; Lie et al., 2004a, 2005). To date, few studies on the biological effects of CHC exposure have been carried out for any polar bear population, and particularly for North American populations: consequently, there is a substantial knowledge gap.

As a result of their high CHC concentrations in adipose tissue, East Greenland and Svalbard bears are at higher health risk among Arctic and Subarctic populations. Recently, investigations of a number of effect parameters from East Greenland polar bears including histopathological examinations of e.g., liver, kidneys, adrenals, and thyroids, and bone structures have been documented (Kirkegaard et al., 2005; Sonne et al., 2005a,b). Further health risk assessments are warranted to clarify the extent to which these particular populations status and health is affected.

3.4. Summary

The results of both univariate and multivariate analysis of the present dataset describe major differences in concentrations and proportions of OC pesticides, PCBs, and $MeSO_2$-PCBs and 3-$MeSO_2$-*p*,*p*′-DDE in adipose tissue of Arctic and Subarctic polar bear populations. The easternmost populations of the polar bear range investigated in this study were distinguished by higher proportions of DDT-related compounds, nonachlor III, oxychlordane, and higher-chlorinated and more persistent PCBs (hepta- to nona-chlorinated). This suggests increased loading from European and Eurasian sources of DDTs, CHL-related compounds, and higher-chlorinated PCB congeners. The accumulation of heavily chlorinated PCBs in polar bear adipose tissue may be due to the exposure of specific commercial mixtures of PCBs, e.g. Aroclor 1254, in the polar bear food chain (Letcher et al., 1995; de March et al., 1998). However, biological factors such as regional variation in diet, food chain structure, and biotransformation ability could also affect the bioaccumulation/biomagnification pattern for these classes of lipophilic compounds (Muir et al., 1988; Norstrom et al., 1988; Olsen et al., 2003). Inversely, western polar bear populations of the North American Arctic were characterized by a significantly higher exposure to relatively volatile compounds such as HCHs and PnCBz, and tri- to penta-chlorinated PCB congeners, suggesting a stronger influence of Southeast Asian and North American-sourced inputs for these classes of contaminants (Iwata et al., 1993; Macdonald et al., 2000).

Concentrations of ΣDDT, Σ_{42}PCB, and Σ$MeSO_2$-PCB showed increased gradient from the westernmost to the easternmost range of the polar bear. The concentrations of these highly lipophilic compounds were highest in female polar bears from East Greenland and Svalbard. In contrast, concentrations of ΣHCH increased at sites going towards the western part of North America, being highest in Alaska female polar bears. No significant gradient was observed for classes of compounds like ΣCHL, ΣCBz, mirex, and dieldrin, which were distributed relatively uniformly over the studied area. The compounds that increased in concentration with latitude were Σ_{42}PCB and Σ$MeSO_2$-PCB. These gradients along latitude and longitude were in general agreement with previous findings reported for polar bears (Norstrom et al., 1988, 1998; Muir and Norstrom, 2000; Andersen et al., 2001; Lie et al., 2003), beluga whales (*Delphinapterus leucas*) (Muir et al., 1990), ringed seals (Schantz et al., 1993; Muir et al., 2000), and general spatial trends in the Arctic environment (de March et al., 1998; Muir et al., 1999; Macdonald et al., 2000; Fisk et al., 2003; AMAP, 2004).

With respect to adipose tissue samples collected during 1989−1993 from comparable female polar bear populations (Letcher et al., 1995; Norstrom et al., 1998), there was a general decrease for the age-adjusted mean concentrations of ΣCHL, *p*,*p*′-DDE, Σ_{42}PCB, Σ$MeSO_2$-PCB, and 3-$MeSO_2$-*p*,*p*′-DDE for the present 1996−2002 female polar bear samples. However, concentrations of dieldrin were unchanged relative to those reported for these female polar bear

populations sampled during 1989−1993. The present figures of temporal comparisons showed great consistency with temporal trends observed for Svalbard polar bear plasma concentrations of CB-153 throughout the 1990s (Henriksen et al., 2001), and ΣCHL, ΣPCB, and dieldrin concentrations measured in East Greenland female polar bear adipose tissue collected during 1999−2001 relative to samples from 1990 (Dietz et al., 2004). Compared to Norstrom (2000), the Western Hudson Bay female polar bear fat samples of the present 2001−2002 assessment displayed trends that were consistent with the apparent non-decreasing trends of ΣCHL, β-HCH, ΣHCH, and dieldrin concentrations over the last 10 to 12 years, and the apparent declining trend for ΣPCB. In contradiction with results by Norstrom (2000), who reported a downward trend for α-HCH and ΣCBz, and no significant change for ΣDDT (mostly *p*,*p*′-DDE), the concentrations of α-HCH and ΣCBz in the present 2001−2002 survey were elevated compared to the last measurements reported for 1997−1999. For ΣDDT, the present mean concentration in Western Hudson Bay was notably lower than last measured in fat samples collected in 1999.

Acknowledgements

Numerous individuals have made contributions to this study in addition to the co-authors. We would first like to thank the local hunters, field biologists and other associated personnel and people in northern communities who participated and assisted in sampling collection and processing. Those involved in sampling, cataloguing and summarizing of biological and geographical information included Julie Beauchesne (Government of Nunavut, Department of Sustainable Development), Marsha Branigan (Government of the Northwest Territories, Department of Resources, Wildlife and Economic Development), Martyn Obbard (Government of Ontario, Ministry of Natural Resources), Bryan Wakeford (Environment Canada, Canadian Wildlife Service), Jonas Brønlund (Ittoqqortoormiit/Scoresby Sound), and Erik W. Born (Greenland Institute of Natural Resources). Magnus Andersen (Norwegian Polar Institute) is thanked for the collection of samples at Svalbard. Rodica Lazar (GLIER), as well as the involvement of other GLIER lab technicians are thanked for their assistance in the chemical analyses. Laboratory personnel and students at NWRI (Mary Williamson, Xiaowa Wang, and Marla Smithwick), NWRC (Wendy Calvert), and NERI (Maja Kirkegaard) are thanked for sample processing, distribution and/or additional chemical analysis. Dr. Katrine Borgå provided valuable comments on the manuscript.

The majority of funding for this study was from by the Northern Contaminants Program, Department of Indian and Northern Affairs of Canada. Supplemental funding was provided by the Canada Research Chairs (CRC) Program and a Premier's Research Excellence Award (to R.J. Letcher). Collection and analysis of samples from Alaska, East Greenland and Svalbard received financial support from the U.S. Fish and Wildlife Service, the Danish Co-operation for Environment in the Arctic (DANCEA), and the Norwegian Pollution Control Authority, respectively.

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