Arch. Environ. Contam. Toxicol. 35, 354–367 (1998)

ARCHIVES OF
Environmental
Contamination
and Toxicology

Chlorinated Hydrocarbon Contaminants in Polar Bears from Eastern Russia, North America, Greenland, and Svalbard: Biomonitoring of Arctic Pollution

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Received: 2 December 1996/Accepted: 5 February 1998

Abstract. Adipose tissue samples from polar bears (*Ursus maritimus*) were obtained by necropsy or biopsy between the spring of 1989 to the spring of 1993 from Wrangel Island in Russia, most of the range of the bear in North America, eastern Greenland, and Svalbard. Samples were divided into 16 regions corresponding as much as possible to known stocks or management zones. Concentrations of dieldrin (DIEL), 4,4′-DDE (DDE), sum of 16 polychlorinated biphenyl congeners (ΣPCB), and sum of 11 chlordane-related compounds and metabolites (ΣCHL) were determined. In order to minimize the effect of age, only data for adults (320 bears age 5 years and older) was used to compare concentrations among regions. Concentrations of ΣPCB were 46% higher in adult males than females, and there was no significant trend with age. Concentrations of ΣCHL were 30% lower in adult males than females. Concentrations of ΣPCB, ΣCHL, and DDE in individual adult female bears were standardized to adult males using factors derived from the least-square means of each sex category, and geometric means of the standardized concentrations on a lipid weight basis were compared among regions. Median geometric mean standardized concentrations (lipid weight basis) and ranges among regions were as follows: ΣPCB, 5,942 (2,763–24,316) μg/kg; ΣCHL, 1,952 (727–4,632) μg/kg; DDE, 219 (52–560) μg/kg; DIEL, 157 (31–335) μg/kg. Geometric mean ΣPCB concentrations in bears from Svalbard, East Greenland, and the Arctic Ocean near Prince Patrick Island in Canada were similar (20,256–24,316 μg/kg) and significantly higher than most other areas. Atmospheric, oceanic, and ice transport, as well as ecological factors may contribute to these high concentrations of ΣPCB. ΣCHL was more uniformly distributed among regions than the other CHCs. Highest ΣCHL concentrations were found in southeastern Hudson Bay, which also had the highest DDE and DIEL concentrations. In general, concentrations of ΣCHL, DDE, and DIEL were higher in eastern than western regions, suggesting an influence of North American sources. Average ΣPCB concentrations in bears from the Canadian Arctic were similar to those in 1982–84, while average ΣCHL and DDE concentrations were 35–44% lower and DIEL was 90% lower. However, the significance of these temporal trends during the 1980s is not conclusive because of the problems of comparability of data.

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Global contamination by environmentally persistent chlorinated hydrocarbon (CHC) pesticides and industrial chemicals has been well documented (Barrie *et al.* 1992; Iwata *et al.* 1993, 1994; Tanabe *et al.* 1994). Many of these compounds are resistant to biological and physical degradation processes. Although oceans appear to be the main sinks (Tanabe and Tatsukawa 1986) and ocean currents play a role (especially under ice-covered seas), global distribution is mainly via the atmosphere (Iwata *et al.* 1993). Global distillation or fractionation of CHCs followed by condensation in cold polar waters has been proposed as a mechanism whereby the polar regions may become sinks for some CHCs (Wania and Mackay 1993). Evidence for this was provided by Iwata *et al.* (1993), who calculated that there was a net flux of chlordanes and PCBs from the atmosphere to the oceans that tended to increase with latitude as the oceans became colder. Global fluxes of HCH were more complex because of the influence of current sources in Asia. We can therefore conclude that CHCs will persist in

polar environments, and there even may be the potential for concentrations of some CHCs to increase. However, there may also be a reversal of trends occurring for some compounds. Recent evidence of oversaturation of Arctic waters, especially under the polar ice cap, indicates that they are becoming a net source of HCH, rather than a sink, to the atmosphere (Jantunen and Bidleman 1995).

There are mg/kg concentrations of a number of CHCs in the blubber of Arctic cetaceans and polar bear (Norstrom and Muir 1994). These include polychlorinated biphenyls (PCBs), chlordane (CHLs), dieldrin (DIEL), and 4,4′-DDT. Persistent metabolites such as oxychlordane and 4,4′-DDE (DDE) may be more prevalent than the precursor chemical. Relative concentrations of CHCs in arctic marine mammals depends on trophic level and metabolic capability of the species (Norstrom and Muir 1994). Chemicals such as PCBs are also of concern to humans consuming these mammals. Concentrations of PCBs are about 2–10 times higher in breast milk of Inuit from northern Québec than in the southern Caucasian population (Dewailly *et al.* 1994). Knowledge of the circumpolar distribution and temporal trends in concentrations of CHCs is therefore important in determining the sources and potential significance of these contaminants to arctic marine and maritime wildlife and humans.

It is difficult and expensive to monitor CHCs in air, water, and low trophic-concentration biota with the intensity required to define geographical and temporal distribution, especially in the Arctic. The state of knowledge of the dynamics of these chemicals in the arctic environment is also inadequate to permit predictions of bioaccumulation from concentrations in water or air to biologically significant concentrations in species at the top of the food web, including humans. An alternative is to monitor concentrations directly in a species that accumulates and integrates contaminants over a known area of the marine environment. The polar bear is an excellent candidate biomonitor for the arctic marine environment. Polar bears are the principal mammalian predators at the top of the arctic marine food chain. They are distributed widely throughout the arctic and subarctic circumpolar regions. Their diet consists mainly of ringed seal (*Phoca hispida*) (Stirling and Archibald 1977). The home range of ringed seals is small. Based on site tenacity and territoriality, Smith and Hammill (1981) estimated that male ringed seals may occupy the same, under-ice habitat for as much as 9 months of the year. Ringed seals therefore integrate CHC contamination in their diet over a limited area.

Individual polar bears may roam over several thousand km in a year, depending on sea ice conditions. Discrete populations in Canada have been defined by a variety of techniques, including mark-recapture and mark-kill. Combined with knowledge of sea ice and land barriers, 12 polar bear management zones were defined (Taylor and Lee 1995). These are considered to be the best estimates of home ranges, but the boundaries are not definitive in many cases. Based on dissimilarity in CHC, mercury, and cadmium residue patterns within zones (Norstrom *et al.* 1988; Braune *et al.* 1991), the polar bear stocks could probably be further subdivided. In some areas, polar bears are philopatric and there is little exchange among populations, although population ranges may overlap significantly in winter (Derocher and Stirling 1990). The average marine area over which a polar bear subpopulation integrates contaminants is in the order of 250,000 km^2 in Alaska (Amstrup and Gardner 1991), but may be much smaller in areas where food supply is plentiful year round, such as in Viscount Melville Sound (Taylor and Lee 1995). The polar bear is therefore a meso-scale indicator of arctic and subarctic marine pollution in most of its distribution.

PCBs and DDT were first discovered in polar bear tissues in the early 1970s by Bowes and Jonkel (1975), a few years after they were identified as environmental contaminants. In 1982–84 a systematic survey of the geographical distribution of CHCs in polar bear fat and liver throughout the Northwest Territories of Canada was undertaken. Relatively high concentrations of PCBs and the pesticide metabolite oxychlordane were found in polar bears, along with lesser amounts of chlordane-related compounds, DDE, dieldrin, HCB, and α-HCH (Muir *et al.* 1988; Norstrom *et al.* 1988).

To study the geographical distribution of CHCs in various regions of the arctic marine environment using polar bear fat as a biomonitor, a sampling survey was organized through the Polar Bear Specialists Group of the International Union on Conservation of Nature. A collaborative study among conservation managers and research biologists from Canada, the US, Greenland, Russia, and Norway was initiated with the Canadian Wildlife Service as lead agency. During the period 1989–93, samples were collected from most of the western hemisphere range of the bear. The fat samples were analysed for 29 individual CHC compounds by gas chromatography/mass spectrometry. The results are presented in this paper as concentrations of DIEL, DDE, and the sum of the CHC compound classes, ΣCHL and ΣPCB, in adipose tissue on a lipid weight basis. The results are discussed in terms of the sources and geographical distribution of these CHCs in the arctic and subarctic marine environments.

Methods

Sample Sites and Sample Description

Samples of subcutaneous adipose tissue from harvested bears and biopsies from bears tranquilized for research purposes were collected from Wrangel Island in the East Siberian Sea, eastward through the polar bear's complete range in North America to east Greenland and Svalbard. Samples were collected from the spring of 1989 to the spring of 1991 in all areas except Wrangel Island and the Arctic Ocean near Prince Patrick Island, where biopsy samples were taken in the spring of 1993. Samples were assigned to 16 regions (Figure 1) based partly on the location of the bear when the sample was taken and partly on management zones in the Canadian Arctic. Therefore, these regions do not necessarily correspond to distinct stocks. A narrative description of the regions and their relationship to the Canadian polar bear Management Zones (Taylor and Lee 1995) is given in Table 1. Bears from R1 are considered to be in the same stock as R2, but since all but one of the bears from R1 were females emerging from dens after a long fast, samples from R1 were kept distinct. A total of 561 samples from bears of all ages were analysed.

Sampling Techniques and Storage

Polar bear adipose tissue is relatively uniform in fatty acid composition throughout the various fat depots (Pond *et al.* 1992). We have found that distribution of CHCs is similar among these depots in 12 male bears from R7 (CWS unpublished data), although there is a significant

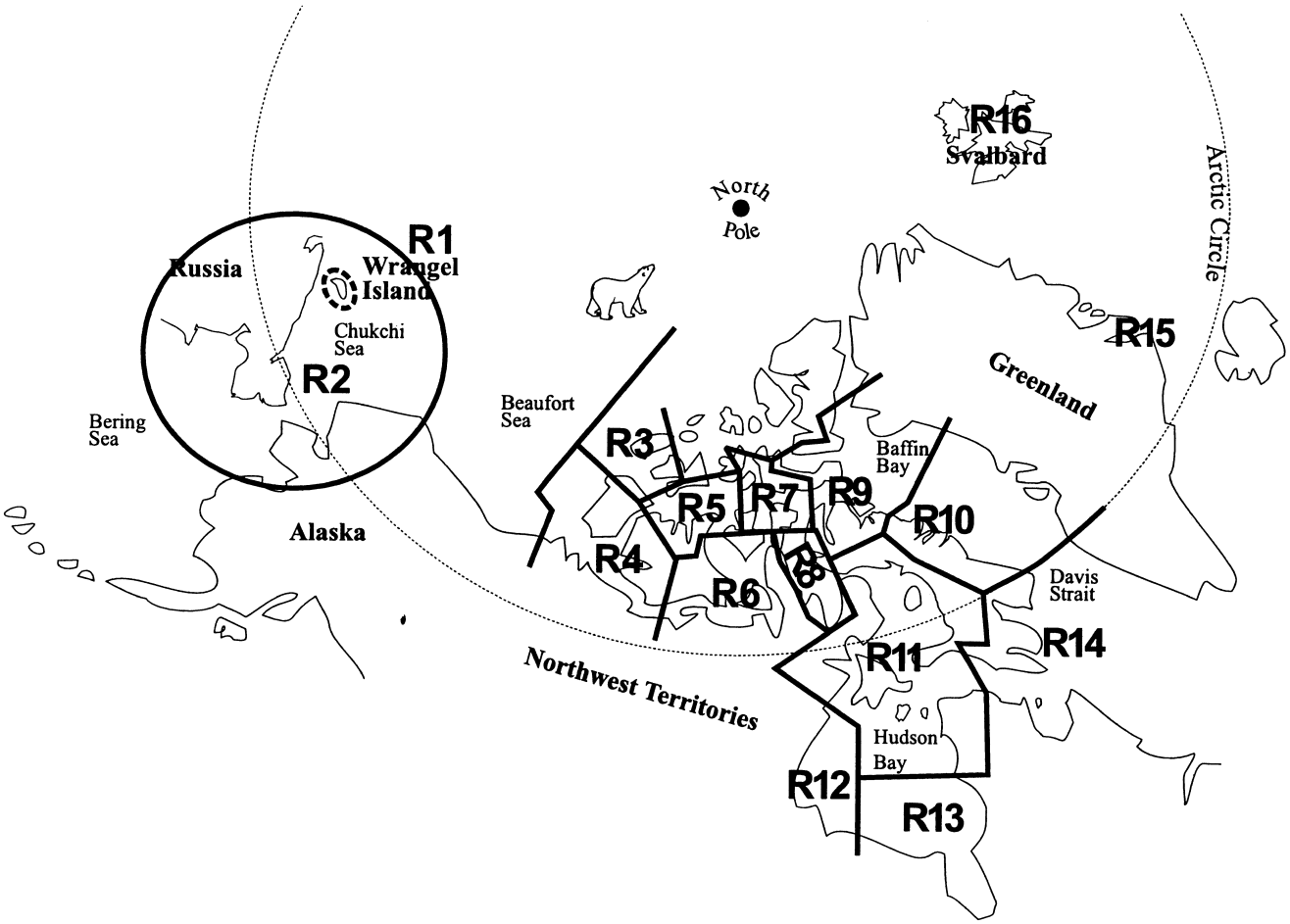

Fig. 1. Regions used to distinguish geographic differences in CHC exposure among polar bears in the western hemisphere. Detailed descriptions of regions is given in Table 1

difference in concentrations in adipose and liver lipids (Norstrom *et al.* 1988; Letcher *et al.* 1996). Bernhoft *et al.* (1997) found little or no discrimination in distribution of CHCs among adipose, plasma, and milk lipids. It is therefore legitimate to compare concentrations in fat taken from any area in the bear.

Necropsy fat samples (50–100 g) were scraped from the rumps of bears killed by aboriginal hunters in the Northwest Territories and Québec. Subcutaneous samples were taken from various areas of the hide from Greenland and Alaska bears. The samples were stored in Whirlpak linear polyethylene bags or solvent-cleaned glass containers, frozen, and shipped to the National Wildlife Research Centre (NWRC) in Hull, Canada. At NWRC the samples were partially thawed, and the whole external surface of the fat mass was carefully excised with a clean scalpel to remove possible contamination from the sampling process. The core of the fat sample was stored at −40°C in solvent-cleaned glass jars with Teflon lid liners prior to analysis.

Biopsy samples were taken from bears tranquilized for other research projects in R1, R3, R5, R12, and R16 (Figure 1). A small patch of skin was exposed on the rump just under the tail using a scalpel blade. A sample of approximately 100–200 mg of skin and subcutaneous adipose tissue was taken with a standard 3-mm veterinary biopsy punch. The samples were placed in cleaned 5-ml glass vials with Teflon lids and frozen prior to shipment to NWRC. The wound was treated with a cream antiseptic.

A premolar vestigial tooth was pulled for age determination at the time of sampling. The date and location of capture, sex, and age were obtained for all bears used in the final analysis. Age was established by histological sectioning of teeth (Stirling and Archibald 1977).

Extraction

Accurately weighed (2 g) necropsy fat samples were ground with 10 g anhydrous sodium sulphate, wet-packed with hexane into a glass column (1 cm ID) and extracted with 80 ml dichloromethane/hexane (1:1). Solvents were evaporated using a Rotavapor RE111 rotary evaporator (Büchi Instruments) to a about 5 ml final volume and adjusted to a volume of exactly 10 ml. An aliquot equivalent to approximately 100 mg fat was spiked with 10 µl CB-112 (2 ng/µl) internal standard, and the volume adjusted to 1 ml for cleanup by gel permeation chromatography (GPC). Another aliquot of the lipid extract approximately equivalent to 200 mg fat was used to determine lipid content gravimetrically.

Biopsy samples were removed from the freezer just prior to lipid extraction. In the initial procedure, skin was separated from the fat plug by using a sharp scalpel while the biopsy was still frozen. The fat plug was placed into a 20-ml tared scintillation vial and the oil was expressed with a small spatula. Any pieces of connective tissue were removed and the scintillation vial was weighed again to determine the weight of lipid. The weighed oil was dissolved in 1 ml dichloromethane, spiked with 10 µl CB-112 (2 ng/µl) internal standard, then 1 ml hexane was added prior to GPC cleanup. The 45 biopsy samples from Wrangel Island (R1) and the Arctic Ocean (R3) were extracted by a different procedure. The whole biopsy was weighed into a 25-ml scintillation vial and ground with approximately 1 g sodium sulphate using a spatula. The vial and mixture was weighed, and the lipid extracted by swirling with 4 × 5 ml rinses of 1:1 dichloromethane:

Table 1. Regions, dates, and relationship to Canadian Polar Bear Management Zones (Taylor and Lee 1995) for sampling of polar bear adipose tissue

Region	Month/Year	Zone	Geographical Location
R1	4/93	—	Wrangel Island (Russia), Chukchi Sea
R2	3/88–3/90	SB (part)	Bering Sea and Bering Strait south of the Arctic Circle, Chukchi Sea, and Goodhope Bay, Alaska Coast to 155°W
R3	4/89–5/93	NB	McLure Strait and the adjacent Arctic Ocean
R4	12/89–5/90	NB/SB	Amundsen Gulf and Beaufort Sea to 135°W
R5	4/89–5/90	VM/PC	Viscount Melville Sound west of 100°W
R6	12/89–5/90	MC	Queen Maud Gulf and Larsen Sound
R7	1/90–5/90	PC	Barrow Strait and Cornwallis Island
R8	12/89–5/90	GB	Gulf of Boothia
R9	4/89–6/90	PC/BB	Baffin Bay north of 72°N, Lancaster Sound, Jones Sound, Kane Basin, Thule, and Ellesmere Island
R10	12/89–1/90	BB	Southern Baffin Bay and Northern Davis Strait (between the Arctic Circle and 72°N)
R11	10/89–4/90	FB	Foxe Basin and Hudson Strait west of 72.5°W
R12	8/89–9/90	WH	Western Hudson Bay (Cape Churchill area)
R13	1/90–4/91	SH	Eastern Hudson Bay (Belcher Islands)
R14	12/89–3/91	DS	Davis Strait (below the Arctic circle) and Hudson Strait east of 72.5°W
R15	1/90–7/90	—	East coast of Greenland near Scoresby Sound
R16	3/90–4/90	—	Svalbard Islands (Norway)

hexane, which were removed by Pasteur pipet. The remaining sample plus sodium sulphate was air-dried overnight, heated to 100°C for 30 min to remove the solvent, and reweighed. The weight of extracted lipid was determined by subtracting this weight from the original sample plus sodium sulphate weight. The weight of lipid extracted from the biopsy was comparable in the two methods.

Cleanup and Separation

The sample extract spiked with CB-112 internal standard was injected quantitatively into one loop of an Autoprep 1002A (ABC Laboratories) gel permeation chromatograph (GPC). The GPC column was eluted with dichloromethane:hexane (1:1) at a flow rate of 5 ml/min. The first 140 ml (lipids) was dumped and 160 ml (CHCs) was collected.

The sample from the GPC was evaporated to about 2 ml on a Rotavapor and quantitatively transferred with hexane (a total volume of 10 ml), onto the top of a 10-g alumina column (1 cm ID × 12 cm long, Fisher Scientific, basic alumina, activated at 300°C for 2 h) for final cleanup. The column was eluted with 65 ml dichloromethane:hexane (1:1). The eluate was evaporated to 5–6 ml on a Rotavapor, then transferred into a 15-ml centrifuge tube that contained 50 µl toluene as a keeper. Further concentration of the sample was achieved under a gentle stream of nitrogen, reducing the sample volume to 1–2 ml. The sample was transferred quantitatively into a 4-ml Reacti Vial that was precalibrated to 70 µl and contained 10 µl CB-154 (5 ng/µl) performance internal standard. The sample volume was reduced under a gentle nitrogen flow to a 70-µl final volume and transferred into the glass insert (100 µl) of a Hewlett-Packard autosampler vial for analysis by gas chromatography-mass selective detector (GC/MSD). For the biopsy samples from R1 and R3, the volume was reduced to 70 µl using a 10-ml, pear-shaped flask and a Rotovapor.

GC-MSD Analysis

The samples were analyzed with a Hewlett Packard 5970 MSD equipped with a 5890 GC, a 7673A automatic injector, and HP 59940 MS Chem Station (HP-UX series). The column was DB-5, 30 m × 0.25 mm ID, thin phase (Supelco). The GC conditions were: injector temperature 250°C, initial oven temperature 100°C, hold 3 min, injector temperature 250°C, first ramp 100°C to 180°C at 20°C/min, second ramp 180°C to 320°C at 5°C/min, volume injected 2 µl. The MSD conditions were: EI ionization at 70 eV in SIM mode, interface temperature 300°C, He carrier gas at 5 PSI (1–2 ml/min).

Quantitation was performed using CB-112 as the internal standard. Response factors of individual CHCs relative to the internal standard were determined daily by injection of a secondary polar bear quantitation (PBQ) standard. PBQ was made from a 150-g polar bear fat sample that was prepared similarly to the samples. The dissolved fat was injected into 54 loops of the GPC (two batches). The eluates were pooled, then fractionated into three fractions on a 35-g Florisil column deactivated by addition of 1.2% water: F1, 145 ml hexane; F2, 145 ml dichloromethane:hexane (15:85); F3, 200 ml dichloromethane:hexane (1:1). The fractions were reduced to 2.2 ml final volume prior to compound identification and determination of CHC concentrations.

Full-scan GC-MSD (m/z 50 to m/z 650) was used to confirm the identity of peaks in all three fractions of PBQ. A sample size of 2 µl was injected into the GC-MSD from F1, F2, and F3 of the concentrated PBQ secondary standard (about 70 mg fat equivalent per µl toluene). Identities of most major peaks were confirmed by comparison of full spectra and retention times obtained from standards. Molecular formulas of ''unknown'' chlordane-related compounds (Muir *et al.* 1988; Norstrom *et al.* 1988) were established from the molecular ion and mass spectral characteristics of the unknown compounds. There were no significant interferences from biogenic or nonchlorinated compounds in any of the fractions.

The concentration of CHCs in F1, F2, and F3 of PBQ in most cases were determined by comparison with authentic standards. The concentrations of some chlordane compounds were determined by gas chromatography–flame ionization detection (GC-FID). GC-FID has the advantage that its response is relatively constant among compound classes on a molar carbon basis (Yieru *et al.* 1990). This allows FID response factors for compounds to be calculated with good accuracy based on molecular formulas, and molar carbon response factors determined from known-concentrations standards. With this procedure the exact structure need not be known. Chlorine does not contribute to FID response. The three fractions of PBQ were injected (2 µl) sequentially into a Hewlett Packard 5890 GC, operated in FID mode. GC conditions were as described for MSD detection, above. Integrated peak areas for contaminants in PBQ were compared directly to integrated areas obtained from injecting authentic quantitation standards of a mixture of 18 CHCs, and mixtures A, B, C, and D of individual PCB congeners (CLB-1, National Research Council of Canada, Marine Analytical Chemistry Standards Program). Because of coelution in F1, PCB-99, and MC-6 (nonachlor-III) concentrations were estimated by a combination of full-scan and SIM GC-MSD using PCB-99 and t-nonachlor standards.

After determination of the concentrations of CHCs, the PBQ fractions were recombined and diluted 20-fold with hexane to a

Table 2. List of analytes in the two CHC compound classes and mean percent contribution to these classes across all 16 regions

	Percent in ΣPCB					Percent in ΣCHL			
Congener	Mean	SE	Min.	Max.	Congener/Metabolite	Mean	SE	Min.	Max.
47/48	0.4	0.01	0.01	2.02	Compound "C"	3.2	0.16	0.37	15.1
56/60	0.1	0.01	0	0.73	Photoheptachlor	3.4	0.20	1.11	50.2
99	8.3	0.16	2.05	16.4	Heptachlor epoxide	7.7	0.11	2.28	11.8
149	0.1	0.01	0	1.21	Oxychlordane	46.8	0.67	16.5	63.6
118	1.0	0.04	0.11	4.41	U-4	8.6	0.35	1.67	33.2
146	0.8	0.02	0	1.94	C-5	1.5	0.05	0	6.19
153	46.0	0.65	21.6	89.5	C-3	2.2	0.06	0.04	5.56
137	0.6	0.02	0	2.64	C-4	2.7	0.09	0.66	18.5
138	8.5	0.17	1.10	15.6	Nonachlor III (MC6)	10.3	0.19	0.65	25.5
182	0.5	0.02	0	2.10	*trans*-Nonachlor	9.6	0.33	1.47	53.4
183	0.8	0.02	0	2.59	U-2	3.9	0.11	0	11.2
156	1.1	0.02	0.09	2.43					
157	0.7	0.02	0.11	4.42					
180	18.5	0.32	2.71	34.0					
170/190	8.6	0.21	1.15	22.4					
194	3.8	0.25	0.23	51.9					

ΣPCB = sum of polychlorinated biphenyls, ΣCHL = sum of chlordane-related compounds. ΣCHL contains metabolites (*e.g.*, oxychlordane) and environmental degradation products (photoheptachlor and unknown compounds, "U") as well as unidentified components of technical chlordane ("C" compounds)

suitable concentration for daily GC-MSD calibrations. Aliquots (50×0.7 ml) of PBQ were sealed in 1-ml ampules, which were opened and used as needed. For determination of CHCs using PBQ, selected single ions were monitored over four chromatographic windows. The external standard was prepared daily by mixing 50 µl PBQ (20-fold diluted), 10 µl CB-112 (2 ng/µl) internal standard and 10 µl CB-154 (5 ng/µl) performance standard; 2 µl of this standard was injected after every third sample. Responses of the analytes in standard and sample runs were normalized to the performance standard (CB-154) prior to internal standard calculation of concentrations based on CB-112 in the sample relative to the standard. The nominal detection limit was 10 µg/kg (ppb).

Recoveries of the CB-112 internal standard were calculated. For an analysis to be acceptable for inclusion in the data set, the recoveries were required to be in the range 85–110%. The laboratory consistently performed well in the Canadian Northern Contaminants Program quality assurance program.

Results

It is beyond the scope of this paper to consider the congener makeup of the major compound classes in detail. A list of the analytes and the average percent contribution to the ΣPCB and ΣCHL compound classes across all eighteen regions is given in Table 2. Some geographical differences have been noted in PCB congener makeup (Letcher *et al.* 1995), but these variations are minor compared to variations in concentration of ΣPCB among areas, and are therefore not a major factor in analysis of overall geographical distribution.

All statistical analyses were performed on the sum of the compounds for CHLs and PCBs. Data were expressed as µg/kg lipid weight prior to analysis. Bernhoft *et al.* (1997) found significantly higher levels of CHLs in adults than subadults and cubs. Therefore, the data set was divided into adults (5 years of age and older) and cub/subadults (0–4 years old). There were 320 bears in the adult data set and 241 bears in the subadult data set. Adult bears were put into three sex categories, male (M), solitary females (F), and females with cubs (FC). The data for adults and cubs/subadults were analyzed separately and then tested for similarity. The significance level for all statistical analyses was $\alpha = 0.05$.

The analyses were performed using the General Linear Model (GLM) procedure of the SAS 6.03 statistical software package. An analysis of covariance between the adult and subadult data was done, using the classification factors (1) sex with three levels (M, F, FC); (2) region with 16 levels; (3) their interactions; (4) the covariate age (years); and (5) its interaction with sex and with region.

Analyses of covariance showed that ΣCHL was the only parameter that had significant differences between adults and subadults (the latter had higher concentrations). The situation for ΣPCB was somewhat cloudy. The analysis of covariance failed to identify any significant differences between adults and cubs/subadults, although the mean subadult ΣPCB concentrations were higher.

Because the purpose of the study was to use biomonitoring data to compare CHC contaminant burdens among regions in arctic marine environments, the data set was restricted to bears >4 years old to minimize the effect of age in the statistical comparisons. The data for bears <4 years old and detailed analysis of the effect of age and sex will be reported elsewhere. The age and sex (M, F, FC) distribution of the adult bears from each region used in the analysis are described in detail in Table 3. Preliminary analyses using the normality tests of D'Agostino *et al.* (1990) showed that the data were not normally distributed. The ΣCHL, DDE, and ΣPCB data were $\log_e$-transformed, which restored normality. Therefore, geometric mean concentrations were used rather than arithmetic means. Although logarithmic transformation was not able to normalize the DIEL data due to the large number of nondetects, geometric means were assumed to be the best representation for DIEL also. The geometric mean concentrations and ranges for CHCs in adult bears are given in Table 4 for each region and sex category.

A summary of geometric mean concentrations by sex category is given in Table 5. The geometric mean concentration of

Table 3. Age (min, max, median) and sex distribution of sampled adult polar bears in each region

Region	Total n	Female			Female with Cub			Male			Median Age (years)	Sex Ratio (F:FC:M)
		n	Min	Max	n	Min	Max	n	Min	Max		
R1	17				16	6	25	1	10	10	9	0:16:1
R2	9	4	6	15	2	16	17	3	5	17	15	4:2:3
R3	25	14	12	31				11	7	31	20	14:0:11
R4	12	8	5	20	1	25	25	3	7	11	8	8:1:3
R5	21	12	6	22				9	5	14	10	12:0:9
R6	13	5	7	13	4	8	14	4	6	9	8	5:4:4
R7	10	2	9	26				8	5	35	9	2:0:8
R8	10	4	5	8	1	21	21	5	5	26	10	4:1:5
R9	18	6	6	15	3	5	20	9	6	19	7.5	6:3:9
R10	5	1	8	8				4	6	25	8	1:0:4
R11	29	8	5	16	3	9	20	18	5	20	12	8:3:18
R12	95	31	5	25	31	6	24	36	5	25	11	31:31:36
R13	12	3	6	11				9	5	24	7	3:0:9
R14	9	4	6	22	2	8	22	3	6	14	9	4:2:3
R15	18	6	7	20	3	7	15	9	5	19	10	6:3:9
R16	14	4	7	32	4	9	14	6	5	10	9.5	4:4:6
Total	320	112			70			138			10	81:39:135

ΣCHL in adult males was significantly lower (29%) than concentrations in both solitary females and females with cubs. There was no significant difference in ΣCHL concentrations between solitary females and females with cubs. The pattern for ΣPCB was the opposite to that for ΣCHL. Males had significantly higher (45%) concentrations than solitary females and females with cubs, and the two female groups were not significantly different from each other. Females with cubs had significantly higher (22%) geometric mean concentrations of DDE than solitary females and males, which were not significantly different from each other.

The effect of sex on residue concentrations was the same at all ages and locations. There were significant differences among regions in overall concentration of residues but there were no interactions between region and sex for ΣCHL, DDE, and ΣPCB or between region and age for ΣCHL and ΣPCB in adults. That is, the effect of location on residue concentrations in adults was the same at all ages and for all sex groupings for ΣCHL and ΣPCB, while it was the same only for sex groupings for DDE. There was a significant interaction between region and age (different rates of change with age in different locations) for DDE in adults. The resulting model for all data sets related the CHC concentration to region and to the age and sex of the bear, having factored out the effect of interactive terms.

Because the sex ratios of the samples varied considerably among regions (Table 3), it was necessary to correct for sex before comparing concentrations among regions. The effect of sex on $\log_e$-transformed concentrations of ΣCHL, DDE, and ΣPCB was independent of region and age, therefore the data for solitary females (F) and females with cubs (FC) were first standardized to adult males by multiplying the untransformed data by a standardization coefficient. The standardization coefficients for ΣCHL, DDE, and ΣPCB in the F and FC classes are given in Table 6. Significance of differences among regions was compared using the Tukey-Kramer multiple comparison option of the SAS GLM procedure. This is the most appropriate multiple comparison to use when the number of comparisons is large relative to the number of means being compared and there are unbalanced sample sizes.

The geometric mean CHC residue concentrations in adipose tissue lipid of adults in each region (Table 4), standardized to adult males using the coefficients in Table 6 to minimize the effect of sex, are presented in Figure 2. The ΣCHL results have also been age-standardized to the median age of 10 using the GLM model intercept for each region, and a slope with age of −0.021. All but one of the samples were female in R1, therefore the sex corrections were greatest in this region. Because of the large set of males in R12 (n = 33), only the male data are plotted. In all cases except ΣCHL in R11 and R12, the uncorrected geometric means were within the confidence intervals of the male-standardized geometric means, although marginally so for both ΣCHL and ΣPCB in a number of cases.

The geographical distribution of ΣPCB is notable for the peaks occurring in R3, R15 and R16 (Figures 1 and 2). ΣPCB concentrations in this group were significantly higher than in the other regions except R13. ΣPCB also tended to be higher than average in R1, R5, and R13 but there were fewer significant differences from other areas. The distribution of ΣCHL (Figure 2) did not have the same degree of variation, nor the same distribution pattern as ΣPCB. While ΣPCB was relatively high in R1, the lowest concentrations of ΣCHL were found in this region. ΣCHL concentrations were significantly higher in R13 than the nine lowest regions. ΣCHL concentrations in R1 were significantly lower than in all regions except R2, R4, and R8. There were few other significant differences among regions. DIEL concentrations had a pronounced tendency to increase from west to east (Figure 2). Concentrations in R1 and R2 were significantly lower than in most other regions. DIEL concentrations were significantly higher in the four highest regions in the east (R11, R13, R15, and R16) than in the four lowest regions in the west (R1, R2, R4, and R5). DDE was distributed in a similar fashion to ΣPCBs (Figure 2), except that there were more pronounced peaks in R1, R10, and R13. Concentrations in R13 were significantly higher than in

eight of lowest regions, and concentrations in R6 and R8 were significantly lower than in the highest eight regions.

Discussion

Biological Influences on CHC Concentrations

We have been able to determine strong relationships between contaminant concentrations, sex, and age and correct for these in the geographical distribution analysis, but there is still a substantial amount of variability in the data set that is unexplained by these factors. Thus, many of the trends and differences among areas apparent in the data are not statistically significant, in some cases because of small sample sizes.

Some variability is almost certainly due to seasonal dietary and physical condition effects. Differences in seasonal fasting behavior may affect CHC concentrations. Pregnant polar bears may go more than half the year without eating. Polischuk *et al.* (1995) showed that ΣCHL and ΣPCB concentrations approximately double in adipose tissue of pregnant females fasting over a 5–6-month period from September to mid-February. However, ΣPCB body burdens (mg/bear) probably do not change as dramatically (Polischuk personal communication). It is possible that clearance of CHCs only occurs during active feeding. Therefore, calculating body burdens from measurements of body composition, when this can be done, may significantly reduce variability. The higher concentrations of ΣPCB and DDE in Wrangel Island bears (R1) compared to the same stock in the Bering and Chukchi Seas (R2) (Figure 2), may be due to fact that all but one of the Wrangel Island bears were females emerging from dens after a long fast, rather than differences in exposure between the two regions. However, it is surprising that ΣCHL concentrations are not similarly affected.

Several factors may affect fasting in males. Over most of their range, adult males do not den except during inclement weather in winter and spring. Depending on prey availability, they may not fast at all. In Hudson Bay, Scoresby Sound, and Svalbard, lack of ice forces the bears onto land in the warmer months (Derocher and Stirling 1990). During this period both males and females fast, making little use of terrestrial food sources (Ramsay *et al.* 1991). In other regions, bears remain associated with ice, and there is no evidence of fasting for predictable periods although food may be in short supply in summer (Schliebe personal communication). It is unclear what effect these differences in habit will have on concentrations of CHCs. Further studies are required.

ΣCHL and ΣPCB concentrations decreased in cubs from approximately two times the concentrations in their mothers at birth to adult concentrations around age 4–5. ΣCHL was significantly lower in adult males than females, and ΣPCB was significantly higher. Bernhoft *et al.* (1997) also found lower levels of chlordane in males from Svalbard, and a more pronounced decrease with age than for other CHC residues. The probable explanation for the lower ΣPCBs in females is that they have an additional mechanism for PCB clearance via milk (Polischuk *et al.* 1995; Bernhoft *et al.* 1997). Lower ΣCHL concentrations in males is difficult to explain. Induction of CYP2B enzyme protein concentration in liver is proportional to both ΣPCB and ΣCHL concentrations (Letcher *et al.* 1996), therefore higher ΣPCB levels could contribute to faster ΣCHL clearance through metabolism. However, females have ΣPCB levels that overlap those of younger males and should also be induced. It is possible that a constitutive CYP enzyme specific to males is responsible for metabolizing chlordanes. Another reason may be differences in dietary exposure of males and females, *e.g.*, differences in pup vs. adult seal predation.

The biomagnification factor of DDE in polar bear from ringed seal is <1, much lower than for ΣCHL and ΣPCB (Muir *et al.* 1988). Therefore DDE is excreted by polar bears faster than ΣCHL and ΣPCB. Faster excretion is likely to be the reason for the lack of difference in DDE and DIEL concentrations in cubs/subadults and adults in the present study. DDE and DIEL concentrations in adipose tissue may therefore be more affected than ΣCHL and ΣPCB by the length of previous periods of fasting or eating at the time the sample was taken, potentially introducing more variability to the data.

Individual dietary preferences, regional differences in prey species availability, and food-chain structure will also contribute to variability in the data. In the present study, we discarded data for three bears from Cape Dorset in R13 because of exceptionally high concentrations of all CHCs and unusual PCB and DDT residue patterns. These bears had probably consumed some beluga, in addition to ringed seal. Walrus and baleen whale carcasses are potential food for polar bear in some of the areas represented in this study in summer, especially in the Bering and Chukchi Seas (Schliebe personal communication). Walrus (except when eating seals) (Muir *et al.* 1995) and baleen whales feed at a lower trophic level than ringed seal and are therefore expected to be less contaminated by CHCs. Consumption of whale carcasses and walrus may have contributed to the lower concentrations of many CHCs in polar bears from the Bering, Chukchi, and Beaufort Seas. Sex-based differences in the diet of polar bears may occur. From data collected throughout the Canadian Arctic, 93% (14/15) of bearded seal kills that were found while still in the possession of a polar bear were being consumed by adult males (Stirling and Derocher 1990). Age and seasonal variations in the proportions of arctic cod and amphipods in ringed seal diet (Hobson and Welch 1992) may also influence CHC uptake.

Geographical Distribution and Temporal Trends of CHCs in Polar Bears

ΣPCB: A survey of CHC contaminants in polar bear liver and composite adipose tissue in 1982–84 found somewhat elevated concentrations of ΣPCB in fat and liver of polar bears from M'Clure Strait (R3, formerly Zone G, now part of Zone MB, Table 1) (Norstrom *et al.* 1988). ΣPCB concentrations in M'Clure Strait were approximately two times higher than those in adjacent Amundsen Gulf and Viscount Melville Sound (R4 and R5, formerly Zones H1 and H2, now part of Zone NB and Zone VM). From Figure 2 it can be seen that R3 had three to eight times higher concentrations of ΣPCB than R4 and R5 in 1989–93. The bulk of the R5 samples taken in 1993 were biopsies from the Arctic Ocean off Prince Patrick Island. The earlier samples in R5 were necropsy samples taken south of Melville Island in M'Clure Strait (Figure 1 in Norstrom *et al.* 1988). Given the steep gradient in ΣPCB concentrations moving from the Arctic Ocean down M'Clure Strait and Viscount Melville Sound to Barrow Strait (R3, R5, R7, Figure

Table 4. Geometric means, 95% confidence limits, and ranges of CHC concentrations in adult polar bear adipose tissue, μg kg^{-1} lipid wt., by region and sex

Region	Sex	n		ΣCHL	DDE	ΣPCB	DIEL
R1	F	16	Geomean	1,016	298	5,535	31
			95% Confidence Limit	668–1,546	169–526	3,488–8,785	21–47
			Range	294–7,673	46–2,911	1,001–28,723	7–217
	M	1		923	120	2,849	36
R2	F	6	Geomean	1,396	91	2,058	42
			95% Confidence Limit	1,095–1,781	73–113	1,706–2,482	26–68
			Range	839–1,877	66–144	1,478–2,883	21–114
	M	3	Geomean	1,053	90	2,380	51
			95% Confidence Limit	279–3,982	54–151	1,999–2,834	20–128
			Range	393–5,495	53–162	2,104–2,958	16–107
R3	F	13	Geomean	4,709	266	13,818	157
			95% Confidence Limit	3,671–6,041	181–391	9,875–19,335	132–187
			Range	2,029–10,270	63–1,027	4,281–40,843	85–277
	M	11	Geomean	2,689	435	22,391	130
			95% Confidence Limit	1,718–4,210	342–553	16,927–29,618	89–192
			Range	871–15,013	249–1,043	12,608–53,175	31–426
R4	F	9	Geomean	2,146	93	3,772	80
			95% Confidence Limit	1,646–2,798	71–122	2,826–5,034	57–112
			Range	1,164–4,369	52–191	2,062–7,693	34–201
	M	3	Geomean	1,364	71	4,732	103
			95% Confidence Limit	770–2,417	51–101	2,969–7,542	61–174
			Range	814–2,711	51–106	2,645–6,494	62–190
R5	F	13	Geomean	2,613	116	5,193	105
			95% Confidence Limit	2,172–3,143	84–161	4,007–6,731	91–123
			Range	1,090–4,227	49–285	2,004–9,811	59–153
	M	9	Geomean	2,245	166	11,357	19
		(DIEL 6)	95% Confidence Limit	1,728–2,917	125–221	9,533–13,529	4–107
			Range	3,922–2,158	116–523	6,151–16,005	43–173
R6	F	9	Geomean	2,981	71	3,372	67
		(DIEL 8)	95% Confidence Limit	2,527–3,515	48–105	2,686–4,233	22–209
			Range	2,158–4,515	32–262	2,169–6,128	19–218
	M	4	Geomean	2,293	38	3,853	Not measured
			95% Confidence Limit	1,319–3,986	27–52	2,472–6,006	
			Range	1,194–4,743	24–61	2,344–7,779	
R7	F	2	Geomean	2,753	197	3,087	122
			95% Confidence Limit	1,889–4,011	110–353	2,656–3,589	110–135
			Range	2,098–3,612	129–300	2,769–3,441	113–131
	M	8	Geomean	1,734	106	4,266	167
			95% Confidence Limit	1,328–2,264	68–163	2,907–6,259	141–198
			Range	1,151–4,119	46–287	1,229–8,493	119–246
R8	F	5	Geomean	2,679	97	2,245	216
			95% Confidence Limit	1,680–4,271	74–127	1,306–3,861	127–370
			Range	1,805–7,662	61–144	1,502–7,558	126–696
	M	5	Geomean	1,211	67	2,919	93
			95% Confidence Limit	636–2,306	49–91	1,857–4,587	52–166
			Range	505–3,235	42–98	1,440–5,798	49–289
R9	F	9	Geomean	2,339	278	3,244	138
			95% Confidence Limit	1,917–2,854	205–377	2,679–3,928	93–204
			Range	1,368–3,666	122–481	2,011–4,880	38–290
	M	9	Geomean	2,428	147	7,633	249
			95% Confidence Limit	1,575–3,741	89–242	6,304–9,242	164–379
			Range	670–5,502	46–744	4,671–12,041	80–617
R10	F	1		3,492	319	6,836	Not measured
	M	4	Geomean	4,618	432	6,258	17
		(DIEL 3)	95% Confidence Limit	2,572–8,291	277–675	3,100–12,631	3–113
			Range	2,260–11,862	233–755	2,266–12,607	40–50
R11	F	11	Geomean	2,942	246	3,774	73
		(DIEL 9)	95% Confidence Limit	2,261–3,828	156–388	2,669–5,336	18–290
			Range	1,572–7,278	74–1,031	1,490–8,861	44–446
	M	18	Geomean	2,093	224	5,648	191
		(DIEL 17)	95% Confidence Limit	1,641–2,669	172–291	4,454–7,162	96–382
			Range	785–6,632	62–659	2,225–17,274	60–751

Table 4. (*Continued*)

Region	Sex	n		ΣCHL	DDE	ΣPCB	DIEL
R12	F	62	Geomean	2,738	302	3,965	107
		(DIEL 57)	95% Confidence Limit	2,499–3,000	272–336	3,616–4,348	74–156
			Range	515–6,707	80–689	1,551–11,970	48–363
	M	36	Geomean	1,765	253	6,022	108
		(DIEL 33)	95% Confidence Limit	1,508–2,064	205–313	5,176–7,006	65–181
			Range	544–4,723	71–1,100	2,160–19,783	61–659
R13	F	3	Geomean	7,937	867	10,939	400
			95% Confidence Limit	5,723–11,009	482–1,560	6,976–17,155	277–578
			Range	5,364–10,682	417–1,313	6,795–17,984	253–512
	M	9	Geomean	4,380	528	9,685	316
			95% Confidence Limit	3,448–5,564	417–670	8,067–11,628	247–404
			Range	2,355–7,040	312–1,014	5,985–14,056	188–574
R14	F	6	Geomean	2,825	268	4,564	174
			95% Confidence Limit	2,120–3,763	140–513	3,473–5,997	108–281
			Range	1,853–5,850	55–592	2,805–8,717	53–383
	M	3	Geomean	2,315	213	8,037	227
			95% Confidence Limit	1,172–4,573	144–316	6,988–9,243	108–480
			Range	1,404–5,396	131–283	7,003–9,450	131–576
R15	F	9	Geomean	5,044	278	22,419	384
			95% Confidence Limit	3,862–6,586	214–360	17,047–29,485	290–508
			Range	2,876–8,220	147–578	12,297–45,996	175–835
	M	9	Geomean	2,729	305	18,232	122
		(DIEL 8)	95% Confidence Limit	1,796–4,149	200–465	12,605–26,372	35–429
			Range	946–7,899	132–1,009	7,841–39,444	79–524
R16	F	8	Geomean	3,162	253	12,775	189
			95% Confidence Limit	2,219–4,504	186–345	9,275–17,595	136–264
			Range	1,713–10,092	118–573	6,244–32,699	119–601
	M	6	Geomean	5,616	413	29,409	344
			95% Confidence Limit	4,347–7,254	273–624	19,427–44,521	264–448
			Range	2,904–7,573	203–995	10,353–47,219	206–578

Table 5. Differences in concentrations of CHCs among the three sex categories in adult polar bears (FC = female with cubs, F = solitary female, M = male)[a]

	FC	F	M
ΣCHL			
Mean	2,756*	2,793*	2,126
Range	1,368–7,995	515–10,681	393–15,013
95% CL	2,536–2,996	2,524–3,091	1,899–2,379
n	56	110	137
DDE			
Mean	258	191*	210*
Range	58–578	33–1,313	24–1,100
95% CL	221–300	165–222	183–241
n	56	110	137
ΣPCB			
Mean	4,557*	4,812*	7,246
Range	1,595–45,997	1,478–40,843	1,229–53,175
95% CL	3,911–5,311	4,146–5,586	6,381–8,229
n	56	110	137
DIEL			
Mean	101	118	104
Range	25–429	19–835	16–751
95% CL	87–118	104–134	91–118
n	52	105	124

[a] Geometric mean concentrations (μg kg^{-1} lipid wt.), ranges, 95% confidence limits, and sample sizes are reported. An asterisk indicates that the means are not significantly different from each other. R1 (Wrangel Island) bears were not included in the analysis because they were mainly females that had been fasting for months and had depleted fat reserves

1), there may well be a gradient in ΣPCB concentrations in polar bears within R5 itself that could explain the differences between the 1982–84 and 1989–93 data.

Norheim *et al.* (1992) analyzed polar bear fat and liver collected from Svalbard during the period 1978–1989. They found ΣPCB concentrations in adipose tissue of adults to be 31,000 ± 30,000 μg/kg. Bernhoft *et al.* (1997) found geometric mean ΣPCB concentrations in adult males and females from Svalbard in 1990–1994, age 7–15 years, to be 28,100 μg/kg and 15,700 μg/kg, respectively. These results are very close to the geometric mean concentrations in the present study for six adult male and eight adult female polar bears from the same area and period, 29,409 μg/kg and 12,775 μg/kg. The geometric means

Table 6. Standardization coefficients for conversion of untransformed CHC residue concentrations in subcutaneous adipose lipid of polar bear solitary females (F) and females with cubs (FC) to equivalent concentrations in males (M)

Sex	S-PCB	S-CHL	DDE
F	1.406	0.705	0.969
FC	1.531	0.69	0.766

[a] The factors are derived from the antilogarithm of the differences between the LSMEANS (SAS GLM Procedure) of the $\log_e$-transformed concentrations of the three sex categories. There were no differences in concentrations of DIEL among categories. Wrangel Island bears (R1) were omitted from this analysis

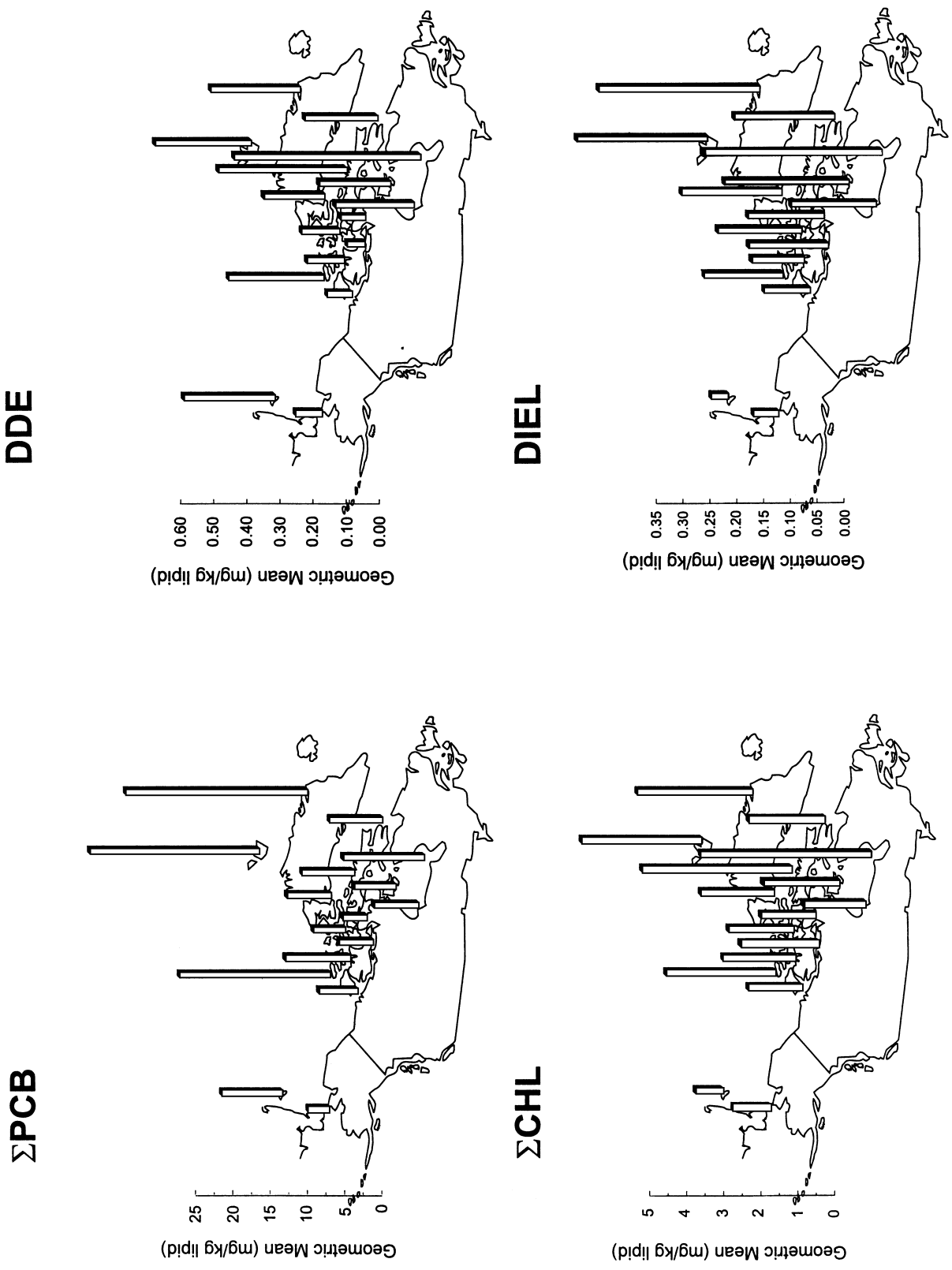

Fig. 2. Hemispheric distribution of concentrations of CHCs in adipose tissue of adult polar bears. Data are geometric mean concentrations (µg/kg lipid weight) for males plus females standardized to males to remove influence of sex on the comparison among regions. ΣCHL data are also standardized to age 10 years. Bars are centered in regions as defined by Figure 1 and Table 1. Numbers of individuals in each region are given by Total n in Table 3, except in western Hudson Bay, where only data for males (n = 33) are plotted

of the male-standardized data from the present study were slightly lower, 22,700 μg/kg than males alone.

The geographical distribution of ΣPCB in polar bears (Figure 2) was in good agreement with that in their ringed seal diet to the extent that the sampling areas overlap. Within the Canadian Arctic, there were no significant differences in ΣPCB concentrations in ringed seal among areas that encompass most of the present data set for polar bears, although the patterns of PCB congeners varied with latitude (Weis and Muir 1997). There are no ΣPCB concentration data for ringed seal in R3 where the anomalously high ΣPCB concentrations in polar bears occurred (Figure 2). ΣPCB concentrations in beluga from the Mackenzie River estuary, Baffin Bay, Davis Strait and Hudson Bay also did not demonstrate a significant geographical variation (Muir *et al.* 1990; Stern *et al.* 1994). ΣPCB concentrations in Alaska ringed seal tended to be lower (Schantz *et al.* 1993), whereas ΣPCB concentrations in Svalbard were higher (Oehme *et al.* 1988).

Temporal trends in ΣPCB concentrations in polar bears cannot be determined with any precision from the present data sets. In our earlier analyses of composite polar bear adipose tissue, the samples had mixed sex and age composition. Concentrations of ΣPCB in composite polar bear adipose tissue from R10, R11, and R12 were two times higher in 1983–84 than in 1969 (Norstrom *et al.* 1988). This may not be the true temporal trend because these samples were not controlled for sex and age (Norstrom *et al.* 1988), which significantly increases variability in the data. Other studies used packed-column analyses (Bowes and Jonkel 1975; Norheim *et al.* 1992), which probably overestimated ΣPCB concentrations. A comparison of ΣPCB concentrations in R3–R12 in the present study with the analysis of composite adipose tissue in equivalent Management Zones in 1982–84 (Norstrom *et al.* 1988) indicated no consistent pattern. Concentrations were higher in four areas, lower in five areas, and the same in three areas for the period 1982–84 than in 1989–93. The mean (±SD) ΣPCB among the 11 Management Zones in 1982–84 was 5,570 ± 1,880 μg/kg. The mean concentration in the corresponding regions in 1989–93 was 5,830 ± 1,860 μg/kg. Overall, there does not appear to be a well-defined trend up or down in ΣPCB concentrations in polar bears in the Canadian Arctic in the 1980s. Gregor *et al.* (1995) were unable to detect any consistent trend in PCB concentrations in ice cores from the Agassiz Ice Cap on Ellesmere Island (R9).

Muir (1994) found no significant change in ΣPCB concentrations in ringed seal from R14 between 1986 and 1992. It is therefore possible that ΣPCB concentrations were already approaching a steady state in the 1980s in the Davis Strait area. This has been occurring in more contaminated areas at similar latitudes, *e.g.*, in Baltic Sea herring (*Clupea harengus*) and guillemots (*Uria algae*) eggs (Bignert *et al.* 1993). Davis Strait is closely connected to the North Atlantic Ocean, so it is not clear whether the ΣPCB trend in seals from this area is representative of other arctic/subarctic areas.

Σ*CHL:* There was a narrower range of ΣCHL than ΣPCB concentrations in polar bear adipose tissue (corrected for sex and age), and only a modest tendency for a west to east increase (Figure 2). Mean ΣCHL concentration in Svalbard male bears was two times higher than in females (Table 4), which is counter to the overall trend for females to have higher concentrations of ΣCHL (Table 6). ΣCHL concentration in Svalbard male bears was three times higher than that reported in Svalbard males by Bernhoft *et al.* (1997), although ΣCHL concentrations in females were similar in the two studies. In a previous study in 1982–84, ΣCHL concentrations were determined in liver and composite adipose tissue from a more limited study area (R4 to R12 and R14 (Norstrom *et al.* 1988). The geographical distribution of concentrations in polar bear liver and adipose tissue in 1982–84 had a more distinct west to east increase than in the present study. However, the previous samples were not controlled for sex and age and had a higher proportion of females and cubs/subadults (which have higher concentrations than adult males).

The geographical distribution of ΣCHL concentrations in polar bears (Figure 2) was in reasonably good agreement with that in ringed seals in the Canadian Arctic. Weis and Muir (1997) found no significant regional differences in ΣCHL concentrations in ringed seal blubber in Canada after removing effects of age and sex. There are no comparable data for ringed seal from other areas of the Arctic.

Mean ΣCHL concentration in polar bear adipose tissue from R4–R12, R14, composited without regard to sex and age, was higher in 1982–84 (3,730 ± 1,720 μg/kg) (Norstrom *et al.* 1988) than the present results from the same areas in 1989–91 (2,100 ± 320 μg/kg, Table 4). The most notable differences between the two time periods were threefold lower concentrations of ΣCHL in northern and western Hudson Bay (R11 and R12) in 1989–90 than in 1982–84. R13 in southeastern Hudson Bay was not sampled in 1982–84. In 1990–91, concentrations in R13 were higher than in other regions in Hudson Bay, but still not as high as in R11 and R12 in 1982–84. In spite of the difficulties in comparing the two data sets because of age and sex differences in sample composition, ΣCHL concentrations appear to have declined throughout the Arctic and Subarctic in the 1980s, especially in Hudson Bay. Muir (1994) did not find any significant changes in ΣCHL concentrations in ringed seal from R14 during the 1986 to 1992 period, but there are no other temporal trend data for ΣCHL with which to make comparisons.

In our earlier study, concentrations of ΣCHL not controlled for sex or age in composite polar bear adipose tissue from R10, R11 and R12 were four times higher in 1982–84 than in 1969 (Norstrom *et al.* 1988). Although concentrations of other CHCs were also higher in 1982–84 than 1969, the increase was much greater for ΣCHL. The factor of four increase may be an overestimate because the sex and age composition of the two composites sampled was different. Nevertheless, there is an indication that peak ΣCHL concentrations in the arctic marine ecosystem occurred sometime in the late 1970s or early 1980s. This is consistent with the increase in chlordane concentrations in Baltic Sea northern pike, which occurred between 1971 and 1982 during a period of rapid decline of ΣPCB and DDE (Moilanen *et al.* 1982).

DDE and DIEL: Geographical distribution of DDE concentrations was more similar to that of ΣPCB than ΣCHL, that is, the highest concentrations in the western part of the study area were in R1 and R3. However, DDE concentrations had a stronger increasing trend from west to east that ΣPCB. DDE concentrations were more than two times higher in regions R12 to R18 in the east than in regions further west, excluding R1, R5 and R7. A similar distribution of DDE was found in 1982–84 (Norstrom

et al. 1988) and interpreted as indicating additional atmospheric load in the eastern areas from North America. ΣDDT concentrations in ringed seal, after removing effects of sex, were uniform throughout the Canadian Arctic, however DDE concentrations were highest in Hudson Bay (Weis and Muir 1997), as was the case for polar bears.

Oehme *et al.* (1988) found two to three times higher concentrations of DDE in Svalbard ringed seal than ringed seal in the Canadian Arctic (Weis and Muir 1997). However, we found low concentrations of DDE in Svalbard bears, similar to those in the eastern Canadian Arctic (Table 4), in good agreement with those obtained by Bernhoft *et al.* (1997) from Svalbard bears during 1990–94. This suggests that the data of Oehme *et al.* (1988) are not comparable to the other ringed seal data because of different age and sex composition, or poor condition.

Region-by-region comparison of the present results with those obtained in composite samples in 1982–84 (Norstrom *et al.* 1988) showed no notable differences in DDE concentrations between the two time periods except in R5 and R7, which had concentrations approximately 1.5 times higher in 1989–93 than in 1982–84. Not much importance can be attached to the latter finding, because the older composite samples covered a narrower geographical span within these two regions and had a much smaller sample size (14 vs. 53). Mean concentrations in composite adipose tissue in R4–R12, R14 were 0.40 ± 0.34 μg/kg in 1982–84 (Norstrom *et al.* 1988) and 0.26 ± 0.21 μg/kg in 1989–93. Only females with cubs have significantly lower (25%) DDE concentrations than males and solitary females (Table 5), and there is no effect of age. Therefore comparison of trends between the two sampling periods is less affected by biological factors than is the case for ΣPCB and ΣCHL. The data suggest that DDE continues to decline in the Arctic.

Geographical and temporal trends of DIEL were much more erratic than those of the other three CHCs treated in the present study. DIEL concentrations in polar bears may be more strongly influenced by factors such as clearance rate from the bear, the interaction of this with fasting and reproductive status, and environmental factors such as precipitation. Many analyses were also at or below the detection limit with the methods used in this study, which adds significantly to the variance. The polar bear is therefore not the best biomonitor for this CHC. Nevertheless, there was nearly a tenfold decrease in mean DIEL concentrations across the Canadian Arctic between 1982–84 (960 ± 260 μg/kg) (Norstrom *et al.* 1988) and 1989–93 (98 ± 59 μg/kg). This is probably a genuine indication that there is a downward trend in DIEL in the arctic ecosystem. DIEL concentrations in ringed seal showed no obvious regional or temporal variations, but the data set is very small (Muir *et al.* 1988; Muir 1994).

Significance of Geographical Distribution of CHCs in Polar Bear

One of the most important findings was the relatively uniform distribution of CHC concentrations over much of the study area. This finding is supported by ringed seal data (Weis and Muir 1997). Thus, polar bear and ringed seal data provide an indication of extensive transport and deposition of CHCs to all areas of the Arctic and Subarctic. ΣCHL was the most uniformly distributed of the CHCs measured in polar bears in the present study. This result is in accord with the finding of lower geographical variation of ΣCHL in air and seawater in the northern hemisphere than in tropical areas (Iwata *et al.* 1993). The lower ΣCHL concentrations in polar bears from R1 and R2 indicate that ΣCHL loading is less in the Chukchi, Bering, and western Beaufort Seas than in the rest of the Arctic. In 1989–91, ΣCHL was undersaturated in the Bering and Chukchi Seas, showing the potential for increases in water concentrations in these areas (Iwata *et al.* 1993). By contrast, temperate latitude Pacific and Atlantic Ocean waters appeared to be roughly at equilibrium with the atmosphere. ΣCHL concentrations now may be close to global equilibrium, or at least entering a phase of uniform distribution and a slower rate of decline.

Wania and Mackay (1993) suggested that the degree of transport of CHCs to the Arctic is influenced mainly by volatility. The least volatile compounds, such as DDT and highly chlorinated PCBs, will tend to have a higher particulate/gaseous phase distribution ratio, and therefore will be more readily scavenged by precipitation and dry deposition close to the sources of emission than more volatile compounds such as chlordane. This would create a negative gradient in deposition from sources at mid-latitudes to remote areas in the Arctic. The more volatile compounds, such as HCH and ΣCHL, will tend to have a more uniform distribution from mid-latitudes to the Arctic. Polar bear and other biomonitoring data are in general agreement with this theory. For example, ΣCHL concentrations were much closer in Arctic and St. Lawrence River beluga (*Delphinapterus leucas*) than were DDE and ΣPCB concentrations in the two areas (Muir *et al.* 1990). Concentrations of ΣPCB and DDE were much higher in seals and seabirds from Scandinavia than from Svalbard, whereas the more volatile polychlorinated camphenes were more evenly distributed (Andersson *et al.* 1988).

The polar bear data provide evidence of distinct regional irregularities in concentrations of CHC contamination of the arctic marine ecosystem. The high concentrations of most CHCs, especially ΣPCB, in polar bears from Scoresby Sound in East Greenland and Svalbard relative to most of the North American Arctic may reflect transport from European as well as North American sources to the Greenland Sea and Barents Sea. Weis and Muir (1997) also attributed the high concentrations of CHCs in seals from Svalbard to sources in Europe that were close to the water bodies from which the seas were collected. Bletchly (1984) estimated that 70% of the world production of PCBs was in the US and Europe. The prevailing airstream movement across eastern North America is north or east, therefore the main deposition of PCBs and other CHCs volatilized from soils and lakes in North America is to the North Atlantic Ocean, where ocean currents may transport them to the Norwegian and Greenland Seas (Barrie *et al.* 1992). Similarly, transport of CHCs from Europe to northern seas may occur at any time of the year (Pacyna 1995) but is especially common in winter (Barrie 1986). The more complicated patterns and higher concentrations of some polychlorinated dibenzo-p-dioxins and dibenzofurans in ringed seal from Svalbard than the Canadian Arctic have been attributed to the influence of European sources at Svalbard (Norstrom *et al.* 1990). Therefore, the net effect of air and water circulation patterns in the northern western hemisphere may be to focus transport of CHCs from source areas in North America and Europe to the seas between Greenland and Europe.

Another important long-range transport mechanism may be ice movement. Pfirman *et al.* (1995) showed that there is consider-

able entrainment of sediments in ice forming on Siberian shelves. The ice moves from Siberia across the Arctic Ocean and exists through Fram Strait over about 3 years. Particulate matter focused on the surface of the ice during seasonal melts may concentrate contaminants from the air. When the ice melts in the Greenland Sea, particulate matter and associated contaminants become available to the marine food chain. This is an alternative explanation for the higher concentrations of CHCs in polar bears and seals from East Greenland and Svalbard.

Concentrations of ΣPCB, ΣCHL, DDE, and DIEL in polar bears from the Bering, Chukchi, and western Beaufort Seas tended to be among the lowest in the study area. This is consistent with atmospheric circulation in this area, which is dominated by eastward flow from Asia and the north Pacific Ocean. Ocean currents flow the same direction from the Bering Sea into the Chukchi Sea (Barrie *et al.* 1992). Sources of CHCs in the Bering, Chukchi, and western Beaufort Seas are therefore more likely to be in eastern Asia. PCB use was much less used in Asia, except Japan, than in North America and Europe (Bletchly 1984). Iwata *et al.* (1993) showed lower concentrations of ΣPCB in surface waters of the north Pacific, Bering, and Chukchi seas than in the north Atlantic Ocean. ΣCHL concentrations in surface waters appeared to be more evenly distributed in northern areas. Unlike PCBs, southeast Asia and India are probably the most significant current global sources of DDT (and therefore DDE), but most of the atmospheric burden is deposited to oceans close to the source, resulting in a very steep decrease in concentration between tropical and arctic waters in the Pacific (Iwata *et al.* 1993, 1994). DDE concentrations in arctic air and seawater are so low that they were seldom detected until recently (Barrie 1994).

The most intriguing finding in CHC distribution in polar bears was the similarity in concentrations in bears from the Arctic Ocean near Prince Patrick Island (R3) in the Canadian Archipelago to those in eastern Greenland and Svalbard (R15 and R16). Regions near R3 in the Beaufort Sea, Amundsen Gulf, and Barrow Strait had much lower ΣPCB concentrations than in R3, while concentrations in adjacent Viscount Melville Sound were intermediate. This suggests a steep gradient between R3 and neighboring regions to the south and east. The median age for bears from R3 was 20, significantly higher than for other regions (Table 3), however, age composition of the sample is unlikely to have resulted in anomalously high concentrations. ΣPCB concentrations in adult females are not dependent on age, and more than half of the samples were females. ΣPCB concentrations in R3 females alone was also much higher than in females from surrounding regions (Table 4). It is tempting to conclude that the similarity in CHC concentrations in R5 and R17/R18 is due to ocean current transport from the Greenland Sea to R3. However, there is no direct surface water flow between the two areas along the northern coast of Greenland and Ellesmere Island. Instead, surface waters in R3 are more likely to be directly influenced by flow from the east Siberian Sea via the Beaufort Gyre (Barrie *et al.* 1992). It is also unlikely that there are regional differences in atmospheric deposition of CHCs that could account for such a steep gradient.

There may be an ecological explanations for the high ΣPCB concentrations in R3 and R5. Welch *et al.* (1992) noted that energy flow at the lowest trophic concentrations in Lancaster sound was dominated by phytoplankton and pelagic-feeding copepods. They suggested that ice algae and under-ice amphipods were more important in areas where there was multiyear ice, such as R3. The relative importance of ice algae and amphipods versus phytoplankton and copepods in the food chain may influence bioconcentration of CHCs (and heavy metals). If ice algae dominate, concentrations of contaminants in the top few meters of water under the ice, including snow and ice melt water, are probably more important to bioconcentration than those in deeper waters. Given that most of the bears in R3 were sampled on permanent ice, it is possible that the exaggerated concentrations of ΣPCB and other CHCs compared to surrounding regions are due to a more under-ice–based feeding ecology of ringed seals. There is little information on regional variability in ringed seal feeding ecology, but higher concentrations of cadmium in ringed seal and polar bear from the western Arctic (R5–R8) have been attributed to a greater proportion of hyperiid amphipods in the ringed seal diet (Macdonald and Sprague 1986; Braune *et al.* 1991). Polar bears from M'Clure Strait in R5 also had exceptionally high concentrations of mercury in liver compared to nearby regions (Braune *et al.* 1991). If the source of mercury in R5 was biogeochemical rather than long-range transport, this finding supports the idea that polar bear or ringed seal feeding ecology in this region may differ from other areas in some respect.

Acknowledgments. Many individuals have contributed to this work besides the principal investigators in the list of authors. Foremost among these are the hunters who provided many of the samples; and Michael Mulvihill, Jiping Zhu, and Mary Simon, who were in charge of the chemical analysis. Jason Duffe, Garry Donaldson, and Leonard Bastien assisted in data management and analysis. Wendy Calvert, Andy Derocher, Dennis Andriashek, John Lee, and Aiko Sutherland were involved in obtaining and summarizing biological and geographical information for the Canadian samples. Thomas Evans is thanked for his thorough review of the manuscript. Funding for chemical and statistical analysis was obtained through the Northern Contaminants Program, Arctic Environmental Strategy, Indian and Northern Affairs Canada. Collection of samples in Greenland was paid by a grant from the Aage V. Jensens Foundation.

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