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Bioaccumulation of Organochlorine Contaminants in Bowhead Whales (*Balaena mysticetus*) from Barrow, Alaska

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Abstract. Bowhead whale (*Balaena mysticetus*) blubber (n = 72) and liver (n = 23) samples were collected during seven consecutive subsistence harvests (1997–2000) at Barrow, Alaska, to investigate the bioaccumulation of organochlorine contaminants (OCs) by this long-lived mysticete. The rank order of OC group concentrations (geometric mean, wet weight) in bowhead blubber samples were toxaphene (TOX; 455 ng/g) > polychlorinated biphenyls (ΣPCBs; 410 ng/g) > dichlorodiphenyltrichloroethane-related compounds (ΣDDT; 331 ng/g) ≥ hexachlorocyclohexane isomers (ΣHCHs; 203 ng/g) ≥ chlordanes and related isomers (ΣCHLOR; 183 ng/g) > chlorobenzenes (ΣCIBz; 106 ng/g). In liver, ΣHCH (9.5 ng/g; wet weight) was the most abundant ΣOC group, followed by ΣPCBs (9.1 ng/g) ≥ TOX (8.8 ng/g) > ΣCHLOR (5.5 ng/g) > ΣCIBz (4.2 ng/g) ≥ ΣDDT (3.7 ng/g). The dominant analyte in blubber and liver was *p,p′*-DDE and α-HCH, respectively. Total TOX, ΣPCBs, ΣDDT, and ΣCHLOR concentrations in blubber generally increased with age of male whales (as interpreted by body length), but this relationship was not significant for adult female whales. Biomagnification factor (BMF) values (0.1–45.5) for OCs from zooplankton (*Calanus* sp.) to bowhead whale were consistent with findings for other mysticetes. Tissue-specific differences in OC patterns in blubber and liver may be attributed to variation of tissue composition and the relatively low capacity of this species to biotransform various OCs. Principal component analysis of contaminants levels in bowhead blubber samples suggest that proportions of OCs, such as β-HCH, fluctuate with seasonal migration of this species between the Bering, Chukchi, and Beaufort Seas.

Persistent organochlorine contaminants (OCs), a structurally diverse group of agricultural and industrial compounds (or by-products), are present in virtually every compartment in the Arctic (de March *et al.* 1998). The transport of these compounds to cold, remote regions is primarily due to long-range atmospheric (Mackay and Wania 1995) and oceanic transport (Macdonald *et al.* 2000; Li *et al.* 2001). The persistence, toxicity, and bioaccumulation potential of OCs is significant in the Arctic marine environment, where many species have greater lipid content than their temperate counterparts (de March *et al.* 1998). As a result, OCs have reached relatively high levels in top predatory marine species, such as beluga whales (*Delphinapterus leucas*) and polar bears (*Ursus maritimus*) (Andersen *et al.* 2001; Norstrom *et al.* 1998).

The bowhead whale (*Balaena mysticetus*) is a large, filter-feeding, baleen whale (mysticete) found in Arctic waters. The largest population, the Bering-Chukchi-Beaufort Seas stock, migrates annually between the eastern Beaufort Sea–Amundsen Gulf in summer and the Bering Sea in the winter (Lowry 1993; Moore and Reeves 1993; Schell *et al.* 1989). In 1993, the population of this stock was estimated to be 8,200 (7,200–9,400, 95% confidence interval) and is gradually recovering from commercial exploitation in the late 1800s and early 1900s at an annual rate of population increase of 3.2% (1.4–4.7%, 95% confidence interval) (Raftery and Zeh 1998). This species is of cultural and nutritional importance to the native communities in northern Alaska, Canada, and Chukotka, Russia, and the native subsistence harvest is carefully regulated (O'Hara *et al.* 1999a).

The bowhead whale occupies a lower trophic position relative to other arctic marine mammals (Hoekstra *et al.* 2002a) as this species almost exclusively feeds on low-trophic-level prey items, such as plankton and euphausiids. As a result, the bowhead may accumulate higher proportions of some OCs relative to other higher-trophic-level biota. The information available on OC concentrations in water from the Bering, Chukchi, and Beaufort Seas suggests that contaminant levels vary along the migratory range of the bowhead whale (Iwata *et al.* 1993; Li *et al.* 2001; Macdonald *et al.* 2000). Previous investigations have documented OC concentrations in a limited number of blubber samples from bowhead whales (McFall *et al.* 1986; Mossner and Ballschmiter 1997; O'Hara *et al.* 1999b), but the biomagnification and potential biotransformation of OCs and polychlorinated bornanes (toxaphene) from lower trophic prey items has not been addressed. Blubber and liver tissue from native harvested bowhead whales were ob-

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tained to investigate the hypothesis that OC concentrations vary by gender and age and may fluctuate seasonally (fall versus spring) with migration between the Beaufort and Bering Seas.

Materials and Methods

Field Sampling

Field sampling of the bowhead whale has been previously described (O'Hara *et al.* 1999b). Complete blubber cores and liver samples from bowhead whales were provided by native subsistence hunters in Barrow and Kaktovik, AK (Figure 1). Samples were collected by staff at the Department of Wildlife Management and Alaskan Department of Fish and Game with the approval of the Alaskan Eskimo Whaling Commission (Barrow, AK). Blubber cores from approximately the same location on each whale (dorsal midline, 1 m caudal to the blowhole) were collected. Life history information was recorded from each whale harvested (*i.e.*, body length, baleen length, sex, etc.). Body length classification (length cohort) was based on lengths at sexual maturation and known age characteristics (juveniles: 6–8.9 m; subadults: 9–12.9 m; adults: $>$ 13 m) (George *et al.* 1999).

Bowhead whale blubber (n = 72) was collected from seven consecutive harvests; fall 1997 (n = 19), spring 1998 (n = 9), fall 1998 (n = 14), spring 1999 (n = 13), fall 1999 (n = 9), spring 2000 (n = 4), and fall 2000 (n = 4). Samples were temporarily stored at −20°C at the Arctic Research Facility (Barrow, AK), and temperature was maintained during transport to analytical laboratories at the National Water Research Institute (Environment Canada, Burlington, Ontario). Blubber cores and liver subsamples were trimmed under clean laboratory conditions, homogenized, and stored at −20°C in prefired glass containers.

Chemicals and Standards

All solvents (pesticide grade) were obtained from Caledon Laboratories (Georgetown, Ontario). Tracepur® ACS-grade granular sodium sulfate (Na_2SO_4) was obtained from EM Science (Gibbstown, NJ). Pesticide-grade dry silica (60–200 mesh) was obtained from ACP (Montréal, Quebec). SX-3 Biobeads (200–400 mesh) used in gel permeation chromatography (GPC) columns were purchased from Bio-Rad Laboratories (Hercules, CA).

Biota Sample Extraction and Clean-Up

Blubber and liver samples were homogenized with Na_2SO_4 and spiked with two polychlorinated biphenyl (PCB) internal standards (CB-30 and CB-204) to monitor the efficiency of the extraction protocol. Liver samples were extracted with dichloromethane (DCM) using Soxhlet extraction for 16 h and subsequently passed through an Allihn funnel containing Na_2SO_4 and concentrated. Blubber samples were transferred to a 250-ml steel beaker with DCM and extracted using a Polytron® homogenizer (Brinkmann, NY) and treated in the same manner as described above. Lipids and other bioorganic materials in each sample were removed using GPC. Lipid percent was determined gravimetrically.

The analyte sample was concentrated and OCs were separated on 8 g of 100% activated silica gel into two fractions: 65 ml of 100% hexane (F1) and 90 ml of 50% hexane:50% dichloromethane (F2). Endrin ketone and 1,3-dibromobenzene (1,3-DBB) were added as laboratory spiking surrogates to determine fractionation performance. Samples were transferred to 2,2′,4-trimethylpentane (iso-octane), roto-evaporated, and concentrated under a gentle stream of high-purity nitrogen gas to 1,000 μl. CB-166 was added to volume correct sample analysis.

PCB/OC Pesticide Analysis

The PCB (100 congeners) and OC pesticide analysis on both fractions from blubber and liver samples were performed using a Hewlett-Packard 5890 gas chromatograph (GC) with a ^{63}Ni-electron capture detector (ECD; Wilmington, DE). Injections of 1 μl were performed by a HP 7673 autosampler with a splitless time of 2.0 min (injector temperature: 220°C). Compound separation was completed using a 60 m × 0.25 mm (ID) DB-5 column (internal film thickness 0.25 μm; J&W Scientific, Folsom, CA) with H_2 carrier gas (constant flow rate 0.91 ml/min). Nitrogen was used as the makeup gas for the ECD (detector temperature: 325°C). The oven temperature program was initiated at 80°C, held for 2 min, ramped to 150°C at 10°C/min, then 2°C/min to 280°C (10-min hold time), and maintained until the completion of the 90-min run. Sample quantification was performed using multiple external standards provided by the National Laboratory for Environmental Testing (Environment Canada, Burlington, Ontario).

Toxaphene Analysis

Toxaphene analysis was performed using a HP Model 6890 GC with HP Model 5973 mass selective detector (MS) in electron capture negative ion mode. Compound separation was completed using a 30 m × 0.25 mm (ID) HP5-MS column (internal film thickness 0.25 μm) The quantification methods and GC-MS operating parameters used have been previously described (Rose *et al.* 2001). The second fraction (F2) was analyzed for total toxaphene (TOX) and homologs using a "Hercules" technical toxaphene standard. Fraction 2 was also analyzed for individual congeners (Parlar 11/12, Parlar 15, Parlar-25, Parlar-31, B6-923, B7-515, B7-1001, B8-531, B8-789, B8-806/809, B8-1413, B8-1414/1945, B8-2229, B9-715, B9-1025, B9-1046, B9-1049, B9-1679, and B10-1110), which were quantified by a series of authentic external standards from Dr. Ehrenstorfer (Augsburg, Germany) or Promochem (Wesel, Germany). Due to fractionation patterns, F1 was quantified for B8-1413 and B9-1679. Polychlorinated bornanes were assigned the appropriate nomenclature as described elsewhere (Andrews and Vetter 1995). In brief, the letter *B* refers to chlorobornane and the number indicates Cl substitution. The four-digit numeric code describes the arrangement of Cl substitution on the bornane carbon skeleton. The Parlar numeric system was used to classify congeners with undetermined structure.

Analytical Quality Assurance

Percent recoveries for PCB and OC pesticide internal standards (CB-30, CB-204, 1,3-DBB, and endrin ketone) were determined for all blubber and liver samples. Samples with extraction recoveries of $<$ 70% for CB-30 and CB-204 were reprocessed and analyzed. All results were blank corrected by subtracting the mean blank concentration from each batch of processed samples. Detection limits were approximately 0.1 ng g^{-1} for all OC compounds. Calibration standards were run every 10 samples. Quality assurance protocol for organochlorine analysis included the use of two standard reference materials (SRM1588 cod liver oil and SRM1945 whale blubber homogenate) from the National Institute of Standards and Technology (Gaithersburg, MD) and participation in an international interlaboratory comparison program on toxaphene analysis (Quasimeme, Aberdeen, UK).

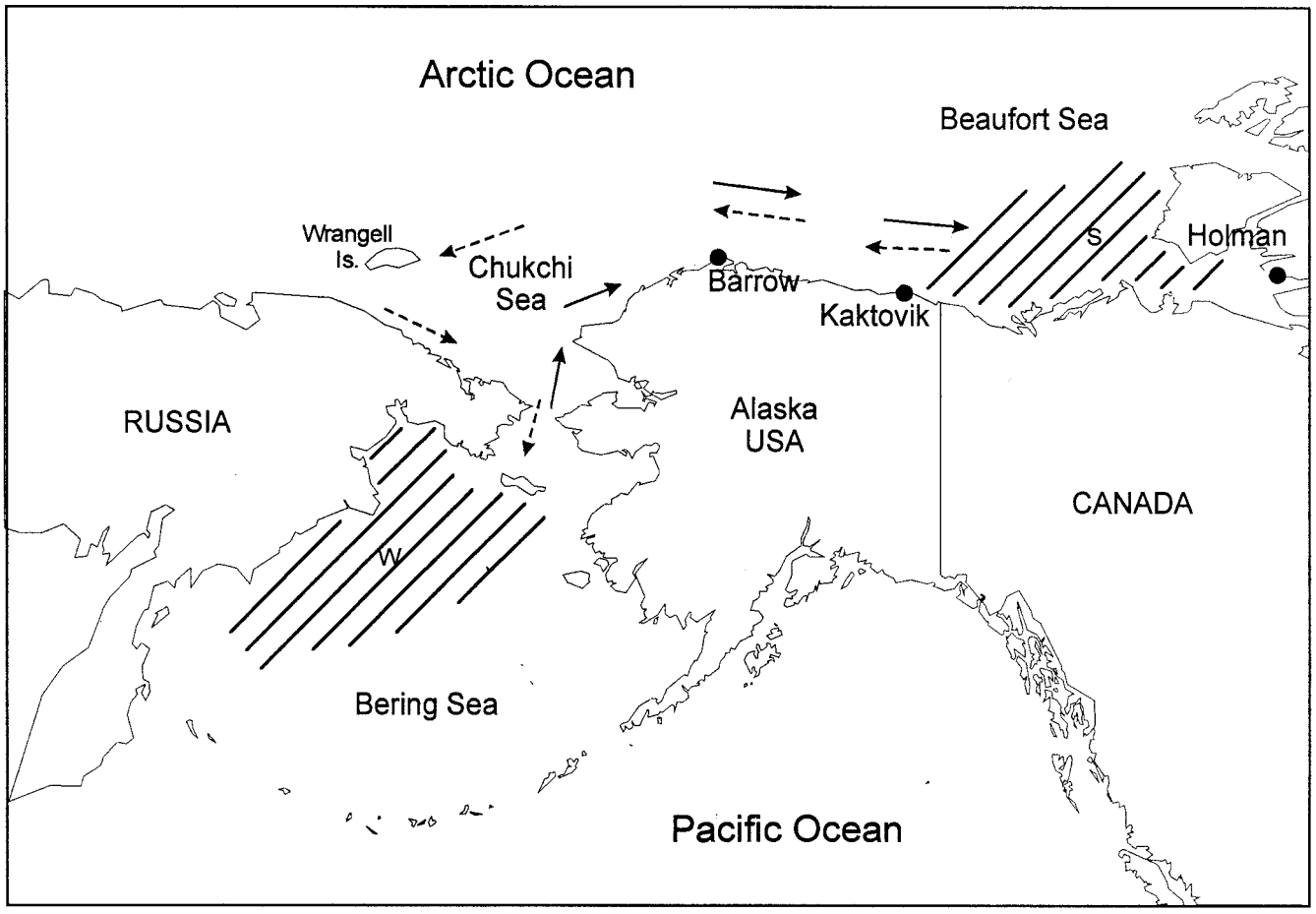

Fig. 1. Annual migration route of the *Balaena mysticetus* (BCBS stock) between wintering (W) and summering (S) grounds. Solid and dashed lines indicate spring and fall migrations, respectively. Biota sampling locations for this study are indicated with black dots

Statistical Analysis

Variation in concentrations of major OC chemical classes (ΣCHLOR, ΣCIBz, ΣDDT, ΣHCHs, ΣPCBs, and TOX) was examined using an analysis of covariance (ANCOVA) using Systat® for Windows, version 8.0 (SPSS, Chicago, IL). Table 1 provides a more detailed account of the composition of each sum OC group. The OC data and length for each whale was log-transformed to reduce the skewness and kurtosis of the data for statistical analysis.

The influence of gender, length, and season of harvest and all first-order interaction effects on ΣOC concentrations (and major individual compounds) in bowhead blubber samples were examined using an ANCOVA model for all covariates and first-order interaction effects was:

$$\text{Log}_{10}\,\Sigma\text{OC} = \mu + \text{sex} + \log_{10}(\text{length}) + \text{season} + [\text{sex} \times \log_{10}(\text{length})] + [\text{sex} \times \text{season}] + [\log_{10}(\text{length}) \times \text{season}] + \varepsilon \quad (1)$$

where ε is the error vector and μ is a constant value. This model was reduced to the following model as factors not significant according to Type III sums of squares test were removed ($\alpha = 0.05$):

$$\text{Log}_{10}\,\Sigma\text{OC} = \mu + \text{sex} + \log_{10}(\text{length}) + \text{season} + [\text{sex} \times \log_{10}(\text{length})] + \varepsilon \quad (2)$$

Differences among group means were analyzed using the Tukey-Kramer HSD test ($\alpha = 0.05$). Correlations between concentration and age, as interpreted from whale length, were determined by Model I linear regression.

Biomagnification factors (BMFs) for OC analytes and sum groups between the bowhead whale (blubber and liver samples) and zooplankton samples were calculated (Table 1). The BMF values were defined as the ratio of the lipid-normalized concentrations using the following equation:

$$\text{BMF} = [\text{Bowhead tissue; ng g}^{-1}\text{ lipid}]/[\textit{Calanus}\text{ spp.; ng g}^{-1}\text{ lipid}] \quad (3)$$

The OC concentrations for zooplankton samples used in the calculation were derived from Hoekstra *et al.* (2002b).

Multivariate statistical analysis on lipid-normalized, untransformed concentration data was conducted using principal components analysis (PCA) to investigate potential patterns in OC data associated with seasonal differences (fall versus spring) in analyzed bowhead blubber samples. Prior to PCA, OC data for individual analytes were normalized to the sum of total OCs. To reduce the number of variables, only those analytes that were above limits of analytical detection in all samples were used. Individual OCs used for PCA are listed in Table 1. PCA was performed on the correlation matrix using Sirius, version 6.0 (Pattern Recognition Associates, Bergen, Norway).

Table 1. Geometric mean concentration (± 1 SE) and biomagnification factors (BMFs) of major sum-OC (ΣOC) groups in bowhead whale blubber and liver samples (1997–2000)

OC Compound	ng g^{-1}, Wet Weight		BMF (Lipid Weight Basis)[a]	
	Liver	Blubber	Liver/*Calanus*	Blubber/*Calanus*
# Samples	23	71		
% Lipid	6.6 ± 0.4	75.8 ± 1.6		
TriCIBz	0.7 ± 0.01	1.2 ± 0.2	0.3	0.5
PentaCIBz	0.3 ± 0.01	0.8 ± 0.1	0.4	1.0
HCB	3.1 ± 0.3	100 ± 7.0	11.6	12.6
ΣCIBz	4.2 ± 0.4	106 ± 7.5	7.6	8.6
α-HCH	6.2 ± 0.4	114 ± 11	3.4	5.4
β-HCH	2.3 ± 0.2	57 ± 2.9	3.9	8.4
γ-HCH	0.8 ± 0.1	29 ± 2.6	2.1	6.7
ΣHCH	9.5 ± 0.5	203 ± 13	3.4	6.3
cis-Chlordane	1.0 ± 0.1	23 ± 3.0	9.4	18.9
trans-Chlordane	0.8 ± 0.2	27 ± 2.5	6.7	19.7
cis-Nonachlor	0.3 ± 0.03	16 ± 1.2	2.2	10.1
trans-Nonachlor	1.2 ± 0.1	34 ± 1.8	5.5	13.7
Oxychlordane	0.5 ± 0.02	12.6 ± 2.0	7.9	17.3
cis-HPEX	1.6 ± 0.2	40 ± 4.6	20.9	45.5
ΣCHLOR	5.4 ± 0.5	152 ± 9.2	2.8	18.5
p,p'-DDE	1.7 ± 0.2	141 ± 19	4.6	33.5
p,p'-DDT	0.2 ± 0.1	31 ± 5.1	0.3	4.0
ΣDDT	3.7 ± 0.3	331 ± 55	2.7	20.8
CB-28	0.2 ± 0.02	8.3 ± 0.8	1.7	6.1
CB-52	0.2 ± 0.02	17 ± 1.8	1.3	9.8
CB-95/66	0.3 ± 0.07	21 ± 2.5	4.5	27.7
CB-99	0.2 ± 0.02	13 ± 1.4	4.5	25.5
CB-101	0.4 ± 0.04	14 ± 1.3	5.1	15.4
CB-105	0.1 ± 0.01	2.8 ± 0.2	5.1	12.3
CB-118	0.2 ± 0.02	14 ± 1.2	1.7	10.3
CB-138	0.1 ± 0.01	12 ± 0.7	2.5	26.4
CB-153	0.3 ± 0.03	13 ± 1.5	5.7	21.4
CB-180	0.1 ± 0.01	3.8 ± 0.9	3.8	12.5
ΣPCB	9.1 ± 0.9	410 ± 29	4.0	15.9
B6-923	0.2 ± 0.01	0.03 ± 0.01	1.3	0.1
B7-1001	1.2 ± 0.2	2.0 ± 0.1	8.0	1.4
B8-1413	0.1 ± 0.03	15 ± 1.4	1.6	21.2
B8-2229	0.2 ± 0.04	14 ± 1.0	3.0	18.3
B9-1679	0.8 ± 0.3	21 ± 1.4	7.4	16.9
TOX	8.8 ± 5	455 ± 45	2.6	8.5
Dieldrin	3.0 ± 0.3	84 ± 4.3	4.5	11.1

[a] Derived from Hoekstra *et al.* 2002b; ΣCIBz = sum of 1,2-diCIBz, 1,4-diCIBz, 1,2,3-triCIBz, 1,2,4-triCIBz, 1,3,5-triCIBz, pentaCIBz, and HCB; ΣHCH = sum of α-HCH, β-HCH, and γ-HCH; ΣCHLOR = sum of *cis*-chlordane, *trans*-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor, heptachlor, and *cis*-heptachlor epoxide (HPEX); ΣDDT = sum of *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDT, and *p,p'*-DDT; ΣPCB = sum of congeners 4/10, 7/9, 6, 8/5, 19, 12/13, 18, 15/17, 24/27, 16, 32, 54/29, 26, 25, 50, 28, 31, 33/21/53, 51, 22, 45, 46, 52, 49, 43, 47/48, 44, 59, 42, 64, 41/71, 40, 100, 63, 74, 76/98, 70, 95/66, 91, 55, 56/60, 92/84, 101, 99, 119, 83, 97, 87, 81, 85, 136, 110, 82, 151, 135, 144, 107/147, 149/133, 118, 114, 143, 141, 145, 153, 132, 105, 141/179, 137, 176/130, 163, 138, 158, 129/178, 175, 187, 182, 183, 128, 167, 185, 174, 177, 171, 156, 202/173, 172, 197, 180, 193, 191, 199, 170/190, 198, 201, 176/203, 189, 206, 195, 207, 194, 205, 208, and 209; TOX = sum of all toxaphene components.

Results and Discussion

OC Levels in Bowhead Whales

The rank order for ΣOC groups in bowhead whale blubber was TOX > ΣPCBs > ΣDDTs ≥ ΣHCHs ≥ ΣCHLOR > ΣCIBz. In liver, ΣHCH was the most abundant ΣOC group, followed by ΣPCBs ≥ TOX > ΣCHLOR > ΣCIBz ≥ ΣDDT (Table 1). The most abundant analytes (from highest to lowest) extracted from bowhead whale blubber samples were *p,p'*-DDE, α-HCH, hexachlorobenzene (HCB), dieldrin, *cis*-heptachlor epoxide (HPEX), *trans*-nonachlor, and *p,p'*-DDT. In liver, the major OC compounds were α-HCH, HCB, dieldrin, β-HCH, *p,p'*-DDE, HPEX, *trans*-nonachlor, B7-1001, and *cis*-chlordane, respectively. Although this study reports toxaphene for the first time in bowhead whales, concentrations of other major OC groups (regardless of gender) were similar to previously reported values (McFall *et al.* 1986; Mossner and Ballschmiter 1997; O'Hara *et al.* 1999b). The rank order of OC concentrations in calanoid copepods (*Calanus* sp.) from several locations in the Beaufort Sea were TOX ≥ ΣPCB > ΣHCH > ΣDDT >

ΣCHLOR > ΣCIBz (Hoekstra *et al.* 2002b). The rank order of major OC groups and individual analytes in these potential prey items were generally reflected in blubber tissue of the bowhead whale.

The OC concentrations in the bowhead are relatively low compared to other balaenopterid cetaceans. The ΣPCB, ΣDDT, and ΣCHLOR concentrations were an order of magnitude greater in the northwest Atlantic right whale (*Eubalaena glacialis*), an endangered baleen whale that migrates along the eastern seaboard of Canada and United States (Weisbrod *et al.* 2000). The proximity of the right whale habitat to major industrialized areas along the eastern seaboard of the United States and Canada is likely responsible for the higher concentrations compared to the bowhead whale.

The concentrations of OCs in the northeast Atlantic and north Pacific populations of minke whale (*Balaenoptera acutorostrata*) were higher than those recorded in bowhead whale blubber (Aono *et al.* 1997; Kleivane and Skaare 1998). Though ΣPCB and ΣDDT concentrations in minke whales from the North Pacific are greater than levels found in the bowhead whale, these OC groups were present in similar proportions of the total OC concentrations in both the north Pacific minkes and the bowheads (Aono *et al.* 1997). The relative contribution of ΣHCH and ΣCIBz to total OC burden was greater in bowheads than in the minke whale. The difference in OC profiles between these species may be attributed to the remoteness of the bowhead whale habitat from major areas of industrialization, possible excretion and biotransformation of CIBz and HCH in fish and other prey items of the minke whale, and the high proportion of ΣHCH in Arctic waters compared to more temperate marine environments (Li 1999; Macdonald *et al.* 2000; Wania and Mackay 1996).

Influence of Gender, Age on OC Concentration

No significant effects of gender and body length were observed for ΣHCH and ΣCIBz concentrations ($p > 0.05$ for both sum OC groups). This is consistent with previous investigations that found negligible relationship between age (as interpreted by length) and ΣHCH and ΣCIBz accumulation for a smaller sample size of bowheads (O'Hara *et al.* 1999b). These OC groups are generally more polar and therefore do not accumulate to the same degree as other more recalcitrant, hydrophobic compounds (Mackay *et al.* 1992). Some HCH isomers are regarded as being easily metabolized by marine mammals and potentially by lower trophic organisms (Hoekstra *et al.* 2002b; Moisey *et al.* 2001; Willett *et al.* 1998). As ΣHCH and ΣCIBz concentrations in bowhead blubber were independent of age and gender, these groups are less likely to accumulate in this species relative to other OC compounds.

The ΣPCB, ΣCHLOR, TOX, and ΣDDT levels were significantly affected by both gender and length. The sex-length interaction term was significant for TOX, ΣPCB, ΣCHLOR, and ΣDDT groups ($p = 0.028$, 0.014, 0.026, and 0.018, respectively). The OC concentrations significantly increased with body length in males for ΣPCB (ΣPCB = 99.3[length] − 600; $r^2 = 0.56$, $p < 0.001$), ΣCHLOR (ΣCHLOR = 38.6[length] − 116; $r^2 = 0.38$, $p = 0.018$), TOX (TOX = 83.7[length] − 345; $r^2 = 0.44$, $p < 0.001$), and ΣDDT (ΣDDT = 86.7[length] − 466; $r^2 = 0.45$, $p = 0.001$). The relation between increasing OC group concentration and body length of male bowhead whales suggests that ΣPCB, ΣCHLOR, TOX, and ΣDDT concentrations increase with age. The ΣPCB, ΣCHLOR, TOX, and ΣDDT concentrations were not significantly correlated with body length of female whales ($p > 0.05$) (Figure 2). In females, ΣPCB, ΣCHLOR, TOX, and ΣDDT levels increased with length (*i.e.*, age) until approximately 13 m ($p < 0.05$). Adult female whales (body length > 13 m) had generally lower ΣPCB, ΣCHLOR, TOX, and ΣDDT concentrations than juvenile and subadult whales ($p > 0.08$).

In general, OC concentrations equally increase in juvenile male and female marine mammals. However, significant differences between gender may be detected after sexual maturity and periods of reproductive activity (Boon *et al.* 1992; O'Shea 1999). Higher OC concentrations in juvenile whales and lower concentrations in mature females (relative to mature males) are likely attributable to transfer of organochlorines from females to young during gestation and lactation. Among cetaceans, it has been estimated that 60–90% of the total ΣPCB and ΣDDT burden is transferred to the nursing calf either through transplacental exchange and/or lactation (Borrell *et al.* 1995; Tanabe *et al.* 1982). The relation with OC concentration and age (by length) further confirms the age classifications previously established for the bowhead whale and suggests that the decrease in OC concentrations in females may be due to lactation and/or *in utero* maternal transfer.

Biomagnification of OCs

The biomagnification of OCs from prey species (*Calanus* sp.) to bowhead whale liver and blubber is summarized in Table 1. The higher BMF values for most OCs for blubber demonstrate the significant bioaccumulation of OCs and metabolites (*i.e.*, HPEX, *p,p′*-DDE) within this species. Though tissue differences in the accumulation of OCs from food items in other cetaceans remains largely unexplored, the lower BMF values for bowhead liver as compared to blubber are most likely due to the partitioning of lipophilic OC compounds to the varying lipid reservoirs of the bowhead whale and/or modification of lipid dynamics and feeding ecology (*i.e.*, fasting) during migration (Lowry 1993). Previous studies of marine mammals have demonstrated that liver concentrations more accurately describe short-term accumulation from recently consumed prey items, and blubber typically represents long-term storage or reservoirs for OC compounds (Kajiwara *et al.* 2001; Wiberg *et al.* 2000).

The BMF values from bowhead whale blubber are similar for ΣPCB, ΣHCH, ΣDDT, and ΣCHLOR found in the northwest Atlantic right whale (*E. glacialis*) despite greater concentration and sample variability in the latter species (Weisbrod *et al.* 2000). The biomagnification of OCs in bowhead and right whales would be expected to be similar as both cetaceans are closely related baleen whales and have common feeding behavior (Lowry 1993; Mayo and Marx 1990).

The biomagnification of ΣHCH, ΣCIBz, ΣCHLOR, and ΣDDT between *Calanus* sp. and bowhead whale (blubber) was greater than values previously reported for each compartment

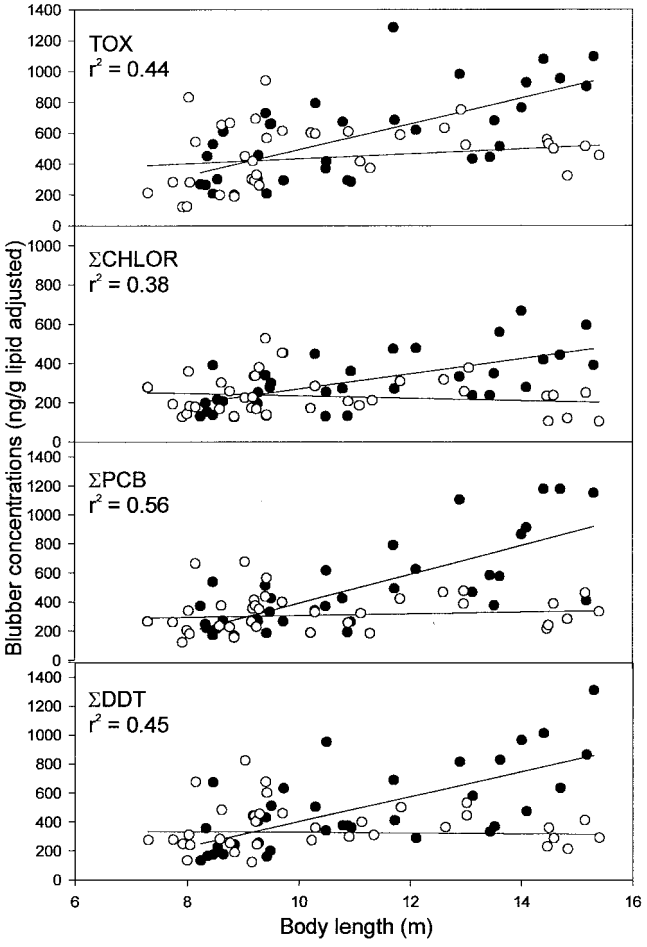

Fig. 2. Body length (in m) versus ΣOC concentrations (ng g^{-1}, lipid-adjusted) in blubber samples from male (black dots) and female (open circles) bowhead whales. Significant relations between increasing body length and ΣOC concentrations ($p < 0.05$ for all comparisons) were found for male bowhead whales

of the arctic cod (*Boreogadus saida*)–ringed seal (*Phoca hispida*)–polar bear food chain (Muir *et al.* 1988). The biomagnification of ΣPCB between Arctic cod and ringed seals from the Northwater polynya in the Canadian Arctic was calculated as 5.5 (Fisk *et al.* 2001a); lower than BMF values found in the *Calanus* sp.–bowhead whale food chain. It has been suggested that the capacity of ceteaceans to biotransform OCs is low relative to most marine mammals as the induction of cytochrome P-450 2B family of isozymes associated with OC exposure may be limited (Boon *et al.* 1994, 1997; Muir *et al.* 1995).

The BMF values of OC compounds from zooplankton by bowhead liver and blubber were significantly correlated with $\log K_{ow}$ values ($p < 0.05$; data not shown). However, the strength of the correlation ($r^2 = 0.14$) in the bowhead whale blubber tissue was very weak, suggesting that factors other than physical partitioning are influencing the accumulation of OCs in the bowhead whale. P-450 characterization and levels of enzymatic induction in the bowhead whale are not currently available, but comparisons of BMF values for individual and sum group OCs allows inferences to be made about OC metabolism (as described below).

The abundance of ΣHCH in the Bering-Chukchi-Beaufort Seas stock of the bowhead whale reflects the long-range atmospheric and oceanic transport of these chemicals and geographic proximity to areas of recent application in Asia (Li 1999; Li *et al.* 2001). The β-isomer of HCH is more prevalent in the northwest Pacific Ocean relative to the Arctic and possibly reflects the application of technical HCH in China up to the 1980s (Li 1999). The relatively high accumulation of HCB and β-HCH (blubber BMFs: 32.6 and 8.4, respectively) suggests that the metabolism of these compounds by the bowhead whale is lower compared to other chlorinated benzenes (ΣCIBz BMF: 16.8; ΣHCH BMF: 6.3) and HCH isomers. In general, β-HCH accumulates in mammals because of its equatorial Cl-substituents, which limits metabolism relative to other HCH isomers (Willett *et al.* 1998).

The BMF profile in the bowhead whale blubber suggests that this species is capable of degrading components of technical chlordane and DDT. Other studies on marine biota have found *trans*-nonachlor as the major chlordane compound and have reported similar patterns of chlordane isomer accumulation (Fisk *et al.* 2001b; Kawano *et al.* 1988). The accumulation of oxychlordane and HPEX is consistent with the overall metabolic pathways of chlordane isomers observed in mammalian systems achieved from long-term chronic exposure (Asakawa *et al.* 1996). Although the accumulation of oxychlordane and HPEX may also be due to formation of these compounds in lower trophic level organisms (Hoekstra *et al.* 2002b), the relatively high BMF values for these analytes (blubber BMFs: 17.3 and 45.5, respectively) compared to ΣCHLOR and individual chlordane isomers provides additional support of metabolism of chlordane by the bowhead whale. The DDE isomers are at a greater concentration in arctic zooplankton than the *p,p′*- and *o,p′*-DDT parent compounds (Fisk *et al.* 2001c; Hoekstra *et al.* 2002b). As a result, the difference in BMF values for *p,p′*-DDE between liver and blubber in the bowhead whale are likely due to changes in bowhead feeding (*i.e.*, fasting) and lipid dynamics, as well as metabolism of the parent DDT isomer and subsequent partitioning to lipid reservoirs.

The lower-chlorinated PCB homologs and congeners were in greater abundance than higher-chlorinated PCBs. The most abundant PCB homolog was the hexachlorobiphenyls, which accounted for 35% of the total PCB burden in bowhead blubber. Congeners belonging to the lower-chlorinated PCB homologs made up an additional 54% of total PCB concentrations. The rank order of the most abundant PCB congeners in bowhead blubber were CB-95/66 > CB-52 > CB-101 > CB-99 > CB-138 > CB-118 > CB-153 (Table 1). The relatively higher amount of the lower Cl-subsistuted PCB congeners and homologue groups reflect the PCB profile in zooplankton from this region (Hoekstra *et al.* 2002b) and supports the lower trophic feeding ecology of the bowhead whale.

The TOX homolog distribution was dominated by octachlorinated toxaphene-related compounds (45% of the mean total TOX concentrations), followed by nona- and hepta-Cl substituted TOX components (31% and 23% of total TOX, respectively). While congener-specific toxaphene data in marine biota has been previously described (Vetter *et al.* 2001), quantification of toxaphene congeners in Arctic marine mammals has been restricted to higher-trophic-level organisms, such as

ringed seals (*P. hispida*), beluga whales (*D. leucas*), and polar bears (*U. maritimus*) (Loewen *et al.* 1998; Stern *et al.* 1992; Zhu and Norstrom 1993). Current understanding of toxaphene metabolism (as inferred from relative congener distribution) in cetaceans is restricted only to odontocetes, which may have significantly different ability to metabolize toxaphene congeners than mysticetes.

In calanoid copepods from the Alaskan and western Canadian Arctic, the most dominant congener was 2-*exo*,3-*endo*,6-*exo*,8c,9b(or 8b, 9c),10a-hexachlorobornane (B6-923), followed by 2-*exo*,3-*endo*,5-*exo*,6-*endo*,8c,9b(or 8b, 9c),10a-hexachlorobornane (B7-1001) (Figure 3) (Hoekstra *et al.* 2002b). The relative abundance of B6-923 and B7-1001 in zooplankton was 21% and 17%, respectively, of the total congener-specific toxaphene concentrations (Hoekstra *et al.* 2002b). However, the dominant toxaphene congeners in bowhead whales were B9-1679, B8-2229, and B8-1413, and represent approximately 65% of the total congener-specific toxaphene concentrations.

The decreased abundance of B6-923 and B7-1001 in the bowhead whale compared to zooplankton suggests that the bowhead may degrade these congeners or that the bioaccumulation of these congeners is low relative to more hydrophobic toxaphene components. Though the possibility of OC biotransformation and distribution to other tissues cannot be eliminated, the negligible contribution of B6-923 to the total congener-specific toxaphene concentration suggests that the bowhead whale may metabolize this congener. B6-923 is believed to be more vulnerable than B7-1001 to oxidative metabolism due to the presence of two *exo*-chlorine atoms vicinal to C1 and unsubstituted C5 position on B6-923 (Vetter and Scherer 1998). The relatively high aqueous solubility of B6-923 and B7-1001 may also give rise to lower bioaccumulation of these congeners relative to more lipophilic, higher-chlorinated bornanes (Fisk *et al.* 1998).

The octa- and nonachlorobornanes congeners, B8-1413 and B9-1679, are the major toxaphene congeners in several marine mammals, representing as much as 80% of total toxaphene levels (Vetter *et al.* 1997, 2001; Bidleman *et al.* 1993; Stern *et al.* 1992). The recalcitrance of these two congeners in mammalian systems and subsequent bioaccumulation has been attributed to an alternating *endo-exo-endo-exo* Cl conformation on the six-member bornane ring and lower polarity relative to other toxaphene congeners (Krock *et al.* 1996). The greater concentration of these congeners relative to other polychlorinated bornanes suggests that bowheads accumulate B8-1413 and B9-1679 very effectively (Figure 3) and that the inability of bowhead whales to biotransform these congeners may be similar to odontocetes.

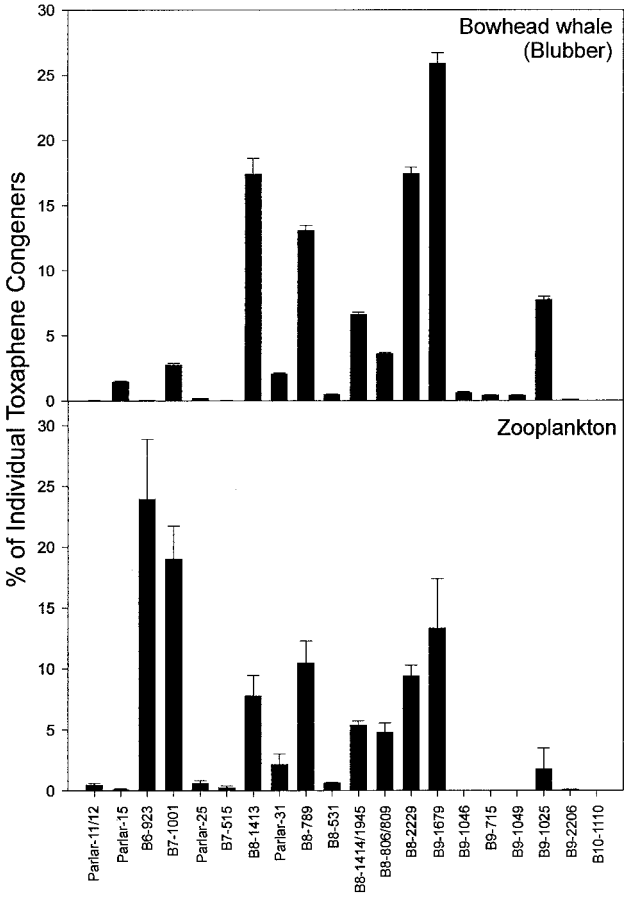

Fig. 3. Relative distribution of toxaphene congeners (average percent of total congener-specific analysis) in zooplankton (n = 8) and bowhead whale blubber samples (n = 72). Error bars represent 1 SE. Zooplankton data derived from Hoekstra *et al.* (2002b)

Temporal Variability of OC Concentrations

No significant differences in mean concentrations (by season) of the major OC groups were observed in harvested whales from 1997 to 2000. However, β-HCH, HCB, CB-153, and oxychlordane concentrations were significantly different between fall and spring harvest whales ($p < 0.05$). The patterns of OCs bioaccumulated in bowhead whale blubber appear to change seasonally (Figure 4). The season of harvest was related to the first principal component (PC1) in the analysis of OC analytes in all bowhead whale blubber samples. In the PCA, scores for fall (F97, F98, F99, F00) harvested whales were clearly separated in PC1 from those specimens collected during spring harvests (S98, S99, S00). This separation is driven largely by the positive loading of the less chlorinated PCBs, toxaphene, and chlorobenzene congeners and the negative loading of the more recalcitrant PCBs (CB-153, CB-180), *p,p′*-DDE, HCB, and β-HCH on PC1 (Figure 4; loading plot).

PCA has been previously used to give a better understanding of OC profiles in cetaceans due to potential regional and seasonal differences in accumulation (Hobbs *et al.* 2001; Krahn *et al.* 2000; Weisbrod *et al.* 2000). Bioaccumulation pattern of OCs in blubber samples from the northwest Atlantic right whale have been suggested to change seasonally due to regional differences in OC abundance in prey items and potential release of OCs from lipid reservoirs during migration (Weisbrod *et al.* 2000). This rationale may also explain the trends observed for the bowhead whale.

Geographic and seasonal differences in feeding on lower-trophic-level organisms in both summer and winter grounds may introduce significant variability in OC exposure to the bowhead whale and is likely reflected in contaminant profiles.

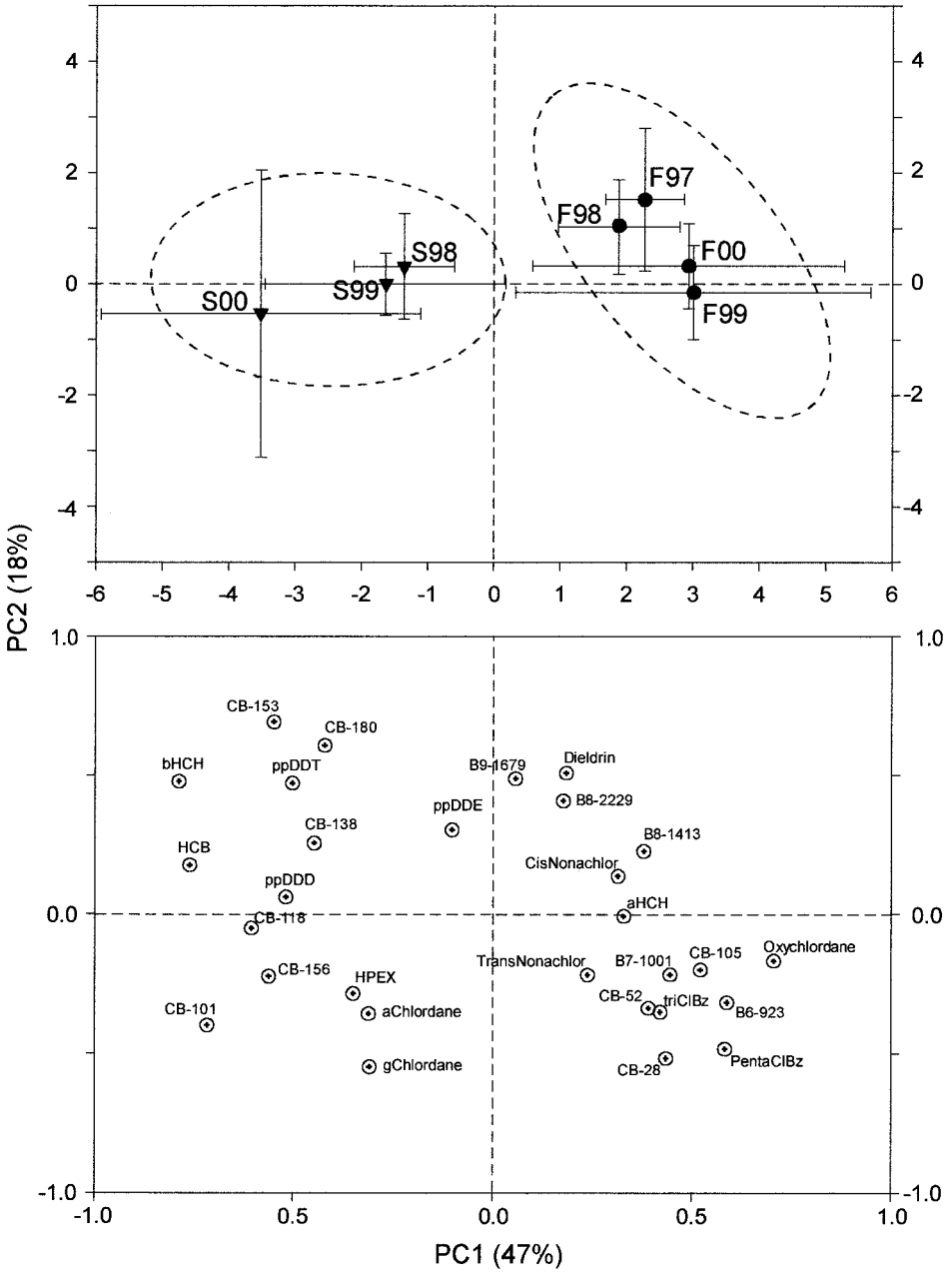

Fig. 4. Principal components analysis plots for OCs in blubber from 72 bowhead whales grouped by season (F = fall, S = spring) and year (97 = 1997, 98 = 1998, 99 = 1999, 00 = 2000) of harvest. Concentrations were normalized to total OC concentrations. The loading plots (lower panel) show the contributions of the individual compounds associated with the principal component eigenvector (scores plot; upper panel). Markers represent mean loading values (± 95% confidence interval) for each group. Ellipses are the bivariate normal 95% confidence intervals for fall- and spring-harvested whales

The invertebrate communities of the Bering and Beaufort Seas have significantly different stable carbon isotope ratios (Schell *et al.* 1998). The dietary intake of lipid-rich zooplankton and marine invertebrates by the bowhead is sufficient to cause seasonal differences in stable carbon isotope ratios in baleen and muscle tissue (Hoekstra *et al.* 2002a; Schell *et al.* 1989). As well, bowhead may undergo periods of fasting during migration (George *et al.* 1999), which may influence lipid dynamics and OC profiles.

Previous reports have suggested that the Beaufort Sea may not be a significant feeding ground, or its importance diminished, compared to the Bering Sea for bowhead whales (Hobson and Schell 1998; Schell *et al.* 1989). However, the non-significant differences in mean concentrations between spring- and fall-harvested whales over several consecutive seasons imply that the feeding ecology of the bowhead whale is similar in both regions. The observed loadings of OCs in bowhead whale blubber on PC1 also generally reflect the differences in levels of persistent OCs in the surface waters of the Bering and Beaufort Seas (Iwata *et al.* 1993; Macdonald *et al.* 2000).

An unusual feature of the PCA loadings plots is the separation of α- and β-HCH by PC1. The relative distribution of α- and β-HCH isomers in surface water is different between the Bering and Beaufort Seas (Li *et al.* 2001). The atmospheric transport of α-HCH and subsequent deposition into the cold Arctic waters has caused α-HCH to be the most dominant HCH isomer in the Beaufort Sea (Li *et al.* 2001). The relatively low Henry's law constants for β-HCH (0.0028 Pa m^3 mol^{-1} at 273°K) relative to α-HCH (0.073 Pa m^3 mol^{-1} at 273°K) (Li *et al.* 2001) is responsible for β-HCH to be less subjected to

direct atmospheric loading into the high Arctic. As a result, β-HCH partitions into the North Pacific surface water (Bering Sea), where it is found in higher concentrations than α-HCH (Li *et al.* 2001).

The partitioning of HCH isomers between the lower- and higher-latitude marine environments (*i.e.*, the north Pacific verses Arctic Oceans) has been observed in biota. The relative abundance of β-HCH was significantly greater than the α-HCH isomer in pinnipeds from 40°N–60°N in the western Pacific Ocean (Mossner *et al.* 1992; Mossner and Ballschmiter 1997; Tanabe *et al.* 1994). As well, β-HCH is the most dominant HCH isomer in blubber tissues from minke whales from the north Pacific (Aono *et al.* 1997). However, α-HCH is the dominant isomer in ringed seals and low trophic level biota from the high Canadian Arctic (Moisey *et al.* 2001). The bowhead whales harvested during the spring migration were recently occupying waters with higher β-HCH relative to the Beaufort Sea (Li *et al.* 2001) which may explain the PC1 results.

Another interesting feature of the PCA was the separation of more recalcitrant compounds by PC2. This components had higher loadings of CB-153, CB-180, toxaphene congeners, and *p,p′*-DDE relative to less-chlorinated chlorobenzene isomers and PCBs. These higher-chlorinated compounds biomagnify within aquatic food webs, and the higher proportions in spring migrating whales might suggest that a different trophic position for the bowhead whale while in the Bering Sea feeding grounds. However, trophic position of the bowheads determined the $\delta^{15}N$ did not vary for the Bering-Chukchi-Beaufort Sea stock of bowhead whales during the study period (Hoekstra *et al.* 2002a). Therefore, the loading profiles described by PC2 may be due to numerous variables affecting seasonal bioaccumulation of OCs (*i.e.*, lipid storage, lipid physiology, physiological status of the bowhead whale, feeding at ice edges) that have not been accounted for in this study. Also, these OCs may be higher in sea water and zooplankton in the Bering Sea waters than in the Beaufort (Macdonald *et al.* 2000; R Macdonald personal communication).

Conclusions

Bowhead whales are exposed to variety of OCs from lower-trophic-level prey items. Immature male and female bowhead whales accumulate OCs in the same manner. However, OC concentrations in sexually mature females are significantly lower than male whales from the same cohort due to maternal transfer to the fetus and neonate. The bioaccumulation profiles of OCs in the bowhead whale suggest that the biotransformational ability of this species is similar to other mysticetes. Though OC concentrations in fall and spring harvested whales were not significantly different, the influence of season was clearly identified by PCA using the proportions of the OCs in bowhead whale blubber. Results suggest that OC exposure to the bowhead whale changes with the annual migration between the Beaufort and Bering Seas and that spatial difference in OC concentrations in sea water and food webs within the Bering-Chukchi-Beaufort region exist.

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