

Dietary biomagnification of organochlorine contaminants in Alaskan polar bears

T.W. Bentzen, E.H. Follmann, S.C. Amstrup, G.S. York, M.J. Wooller, D.C.G. Muir, and T.M. O'Hara

Abstract: Concentrations of organochlorine contaminants in the adipose tissue of polar bears (*Ursus maritimus* Phipps, 1774) vary throughout the Arctic. The range in concentrations has not been explained fully by bear age, sex, condition, location, or reproductive status. Dietary pathways expose polar bears to a variety of contaminant profiles and concentrations. Prey range from lower trophic level bowhead whales (*Balaena mysticetus* L., 1758), one of the least contaminated marine mammals, to highly contaminated upper trophic level ringed seals (*Phoca hispida* (Schreber, 1775)). We used $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ signatures to estimate the trophic status of 42 polar bears sampled along Alaska's Beaufort Sea coast to determine the relationship between organochlorine concentration and trophic level. The $\delta^{15}\text{N}$ values in the cellular portions of blood ranged from 18.2‰ to 20.7‰. We found strong positive relationships between concentrations of the most recalcitrant polychlorinated biphenyls (PCBs) and $\delta^{15}\text{N}$ values in models incorporating age, lipid content, and $\delta^{13}\text{C}$ value. Specifically these models accounted for 67% and 76% of the variation in PCB153 and oxychlorodane concentration in male polar bears and 85% and 93% in females, respectively. These results are strong indicators of variation in diet and biomagnification of organochlorines among polar bears related to their sex, age, and trophic position.

Résumé : Les concentrations de contaminants organochlorés dans les tissus adipeux des ours polaires (*Ursus maritimus* Phipps, 1774) varient d'un bout à l'autre de l'Arctique. L'étendue des concentrations n'a pu être complètement expliquée par l'âge des ours, leur sexe, leur condition, leur lieu d'habitation ou leur statut reproductif. Les voies alimentaires exposent les ours polaires à une variété de profils et de concentrations de contaminants. Leurs proies s'échelonnent des baleines franches boréales (*Balaena mysticetus* L., 1758) de niveau trophique bas, un des mammifères marins les moins contaminés, aux phoques marbrés (*Phoca hispida* (Schreber, 1775)) de haut niveau trophique et fortement contaminés. Les signatures de $\delta^{15}\text{N}$ et de $\delta^{13}\text{C}$ nous ont permis d'estimer le statut trophique de 42 ours polaires échantillonnés le long de la côte de la mer de Beaufort en Alaska afin de déterminer la relation entre les concentrations d'organochlorés et le niveau trophique. Les valeurs de $\delta^{15}\text{N}$ dans la fraction cellulaire du sang varient de 18,2 ‰ à 20,7 ‰. Il existe de fortes relations positives entre les concentrations des biphényls polychlorés (BPCs) les plus récalcitrants et les valeurs de $\delta^{15}\text{N}$ dans les modèles qui tiennent compte de l'âge, du contenu lipidique et des valeurs de $\delta^{13}\text{C}$. En particulier, ces modèles expliquent respectivement 67 % et 76 % de la variation des concentrations de BPC153 et d'oxychlorodane chez les ours polaires mâles de même que 85 % et 93 % chez les femelles. Ces résultats indiquent de façon claire que la variation du régime alimentaire et la bioamplification des organochlorés chez les ours polaires sont reliées au sexe, à l'âge et à la position trophique.

[Traduit par la Rédaction]

Introduction

Studies of recalcitrant organochlorine (OC) contaminants in the adipose tissues of polar bears (*Ursus maritimus* Phipps, 1774) report a wide range of concentrations (Muir et al. 1988; Norstrom et al. 1988; Norstrom and Muir 1994;

Bernhoft et al. 1997; Kucklick et al. 2002; Dietz et al. 2004; Verreault et al. 2005). This variation may indicate differences in feeding ecology and biomagnification of OCs among groups of bears occupying different regions and among individual polar bears (Kucklick et al. 2002). Although polar bears are top carnivores in the arctic marine ecosystem, concentrations of OCs in their adipose tissue do not always exceed those of their typical prey (Kucklick et al. 2002). Concentrations of many OCs similar to those in polar bears have been reported in some ringed seals (*Phoca hispida* (Schreber, 1775)), walrus (*Odobenus rosmarus* (L., 1758)), and beluga whales (*Delphinapterus leucas* (Pallas, 1776)) (Muir et al. 1995; Wade et al. 1997; Kucklick et al. 2002). This absence of OC biomagnification between polar bears and their prey is partly explained by the ability of polar bears to biotransform and excrete bioaccumulative compounds that many other marine mammals cannot as effectively eliminate (Letcher et al. 1996, 1998). OC concentrations can vary among polar bears with age, sex, body condition, location of capture, or lipid composition of tis-

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sues, but these factors have not fully explained the large range of concentrations among individuals. This may be due in part to differences in diet related to the availability and use of lower trophic level prey. Kucklick et al. (2002) found that OC-biomagnification factors in the food chain from ringed seal to polar bear varied by region, further suggesting variation in polar bears' diet across the Alaskan Arctic.

Their entirely carnivorous, lipid-rich diet (Stirling and McEwan 1975) exposes polar bears to high levels of lipophilic organic pollutants found throughout the arctic marine ecosystem (de Wit et al. 2004). OC compounds generally undergo many trophic transfers, from phytoplankton to zooplankton to fish to seals, before reaching polar bears. This can result in biomagnification of highly recalcitrant OCs in polar bear tissues. Ringed seals occur throughout the range of the polar bear and represent most of their annual diet (Lønø 1970; Smith 1980; Gjertz and Lydersen 1986; Derocher et al. 2002; Bentzen et al. 2007). Ringed seals are high trophic level carnivores with a diet consisting primarily of arctic cod (*Boreogadus saida* (Lepechin, 1774)) in winter and invertebrates in summer (Lowry et al. 1980; Dehn et al. 2005b). This diet exposes ringed seals to high levels of contaminants, resulting in relatively high concentrations of OCs in their tissues (Norstrom and Muir 1994; Krahn et al. 1997; Woshner et al. 2001; Hoekstra et al. 2002b).

Like other ursids, polar bears are opportunistic predators. In the southern Beaufort Sea and elsewhere in the Arctic, their diets also include bearded seals (*Erignathus barbatus* (Erxleben, 1777)), beluga whales, and walrus (Lowry et al. 1987; Calvert and Stirling 1990; Smith and Sjare 1990; Rugh and Sheldon 1993). Both bearded seals and walrus generally have lower OC concentrations than ringed seals (Muir et al. 1995; Krahn et al. 1997). Stranded bowhead whales (*Balaena mysticetus* L., 1758) and gray whales (*Eschrichtius robustus* (Lilljeborg, 1861)), and bowhead whales taken in native subsistence hunts along Alaska's Beaufort Sea coast are also scavenged by some polar bears (Miller et al. 2004; Bentzen et al. 2007). Because bowhead whales feed predominantly on zooplankton, they occupy a low trophic position and hence they contain relatively lower contaminant concentrations than other arctic marine mammals (O'Hara et al. 1999; Hoekstra et al. 2002b). This variation in trophic level among foods likely exposes polar bears in the Beaufort Sea region to varying contaminant profiles and concentrations.

Stable isotope analysis of carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) has been used as a tool for evaluating trophic relationships in the arctic marine food web (Hobson and Welch 1992; Atwell et al. 1998; Pauly et al. 1998; Hoekstra et al. 2002a; Bentzen et al. 2007). The $\delta^{15}\text{N}$ composition of an organism has been used to trace dietary history and nutritional ecology within populations or stocks as well as movements of individuals between geographically distinct ecosystems (Abend and Smith 1995; Hilderbrand et al. 1996; Pond and Gilmour 1997; Hobson and Schell 1998; Hoekstra et al. 2002a). Analyses of $\delta^{15}\text{N}$ have been used to assess transfer of OCs between trophic levels in aquatic food webs (Hobson et al. 1995; Fisk et al. 2001). In particular, the $\delta^{15}\text{N}$ signature of an organism has been used as an indicator of its relative trophic position. There is generally an increase on the

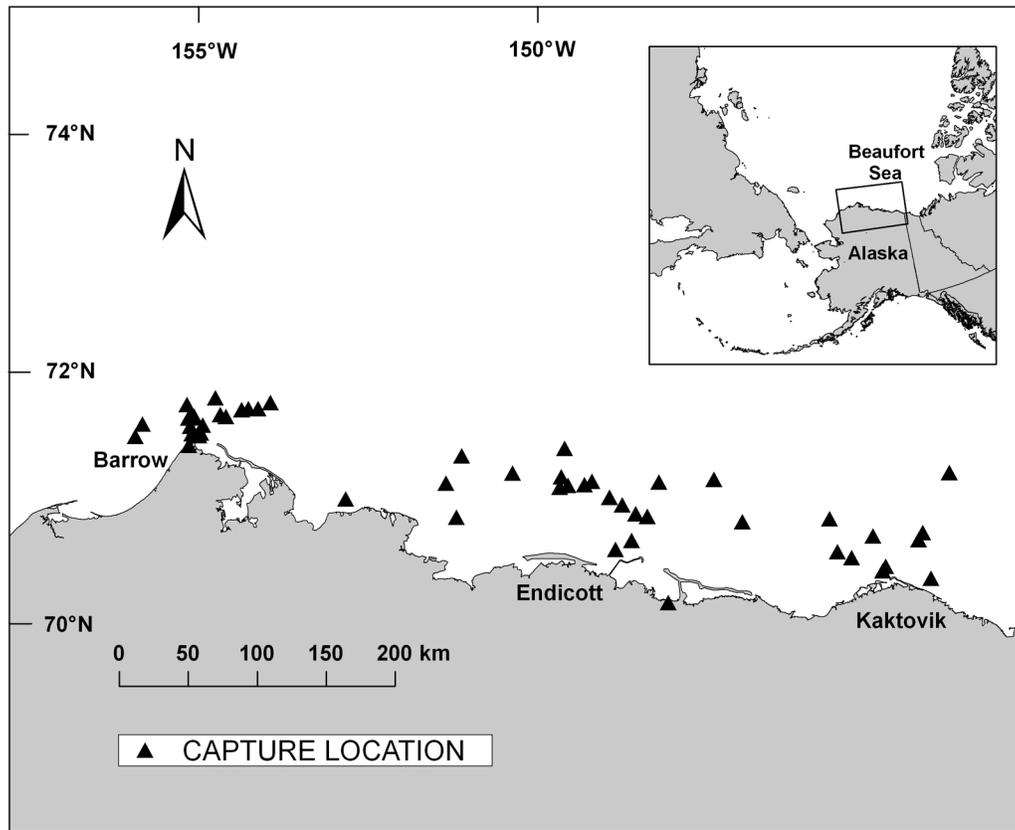
order of 3‰–5‰ in $\delta^{15}\text{N}$ values with each transfer from one trophic level to another (Hobson and Welch 1992; Kelly 2000; Kurle and Worthy 2002). This provides a relatively long-term measure of the trophic position of an organism integrated over several months, which is difficult to obtain through stomach-content analysis or short-term direct observations (Pond and Gilmour 1997; Atwell et al. 1998).

Polychlorinated biphenyls (PCBs) and chlordanes (CHL) are the two most abundant OC-contaminant groups in the adipose tissue of Alaskan polar bears (Kucklick et al. 2002). OC pesticides, including CHL, dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexane (HCH), and their by-products, are also found in tissues of polar bears throughout the Arctic (Muir et al. 1992; de Wit et al. 2004). These recalcitrant, lipophilic compounds and their metabolites tend to biomagnify in the arctic marine ecosystem, owing to the lipid-rich diets and large fat reserves of arctic marine mammals (Norstrom et al. 1988; Moisey et al. 2001; Polischuk et al. 2002; Hoekstra et al. 2003b).

OC contamination may have many adverse effects on the health of arctic marine mammals (Norheim et al. 1992; Krahn et al. 1997; Beckmen et al. 2003; Oskam et al. 2004; Lie et al. 2005). Derocher et al. (2003) found evidence for lower reproductive and cub-survival rates in polar bears associated with environmental contaminants. Altered hormone levels and bone densities, impaired immune systems, and impaired endocrine systems in polar bears have all been associated with high OC concentrations (Wiig et al. 1998; Bernhoft et al. 2000; Skaare et al. 2001; Haave et al. 2003; Oskam et al. 2003; Sonne et al. 2004). Sonne et al. (2005) found evidence of histological changes in the liver tissue of east Greenland polar bears, including chronic inflammation correlated with high concentrations of OC contaminants, but recognized that age and infectious diseases are important agents affecting polar bears' condition.

The relative importance of trophic transfer for OC exposure varies among specific organisms and chemicals. Therefore, relationships between trophic position, measured as the $\delta^{15}\text{N}$ value, and OC burdens have been used to better understand the biomagnification of OCs with each trophic transfer in the arctic marine food web and to calculate overall food web magnification factors (Fisk et al. 2001; Moisey et al. 2001; Polischuk et al. 2001; Hobson et al. 2002; Hoekstra et al. 2003a, 2003b). We hypothesize that polar bears feeding at the highest trophic level according to $\delta^{15}\text{N}$ values will contain higher contaminant burdens associated with a diet dominated by ringed seals. Polar bears utilizing measurable proportions of lower trophic level prey such as walrus, bowhead whale, or gray whale remains should have lower $\delta^{15}\text{N}$ values and lower contaminant burdens, thus reflecting this dietary difference. We examine the relationship of trophic level, measured as the $\delta^{15}\text{N}$ value in polar bear blood, with concentrations of PCBs and OC pesticides, including six individual congeners all found at high concentrations in polar bear adipose tissue and their prey. We also consider the application of a "congener profile" to help discern trophic relationships. We assess the influence of numerous biological factors and how they may explain, independently or via interactions, the variability in OC concentrations observed in the southern Beaufort Sea population of polar bears.

Fig. 1. Map of northern Alaska and the southern Beaufort Sea, showing all polar bear (*Ursus maritimus*) capture locations in spring 2003 from Barrow, Endicott, and Kaktovik.



Materials and methods

The study area and polar bear sampling were similar to those reported in Bentzen et al. (2007). All captures were conducted from Barrow (71°16'N, 156°47'W), Endicott Island (70°18'N, 147°52'W), and Kaktovik, Alaska (70°08'N, 143°34'W), and covered an area following the coastline between Barrow and Demarcation Point (69°70'N, 141°00'W) and ranging beyond 50 km from the coast (Fig. 1). Polar bears were captured from March through May 2003 by injecting intramuscular immobilizing drugs contained in projectile syringes fired from a helicopter (Amstrup et al. 2000). All capture procedures were reviewed and approved by independent animal care and use committees. Sex was determined, body mass was estimated (Durner and Amstrup 1996), and physical condition was visually assessed for all bears. Blood and subcutaneous adipose tissue were collected from 42 polar bears (18 females, 24 males) ranging in age from 1 to 22 years (Table 1). Although the ages of recaptured bears and cubs were already known, all newly captured bears were marked with a tattoo and an ear tag, and a vestigial premolar was extracted to determine age by counting cementum annuli at Matson Laboratory, Milltown, Montana (Calvert and Ramsay 1998).

Blood (5 mL) was collected from the femoral vein or artery for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ analysis as previously described in Bentzen et al. (2007). In brief, Vacutainers[®] (without anticoagulant) of whole blood were centrifuged, serum was removed, and packed blood cells were frozen at $-20\text{ }^{\circ}\text{C}$ until

analysis at the Alaska Stable Isotope Facility, University of Alaska Fairbanks. Blood samples were freeze-dried and then homogenized. Dried blood cells (0.2–0.4 mg) were then subsampled and analyzed for stable nitrogen and carbon isotope ratios using a Carlo Erba NC 2500 elemental analyzer coupled to a Finnigan Delta+ continuous-flow isotope-ratio mass spectrometer via a ConFlo III. Analysis of a peptone standard (Sigma Chemical Company) during the sample run for both $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ gave an analytical precision of $\pm 0.1\text{‰}$ and $\pm 0.2\text{‰}$, respectively.

Subcutaneous fat (~1 g) was collected from the rump using a 6 mm diameter biopsy punch, stored in Teflon[®] vials, and immediately frozen at $-20\text{ }^{\circ}\text{C}$ in the field before storage at $-80\text{ }^{\circ}\text{C}$ until analysis. All fat samples were analyzed for PCBs, CHL, HCHs, and DDTs at the National Water Research Institute in Burlington, Ontario (Environment Canada). Fat sample preparation methods were similar to those described by Hoekstra et al. (2002a). Between 0.5 and 0.6 g of the fat sample was removed from the container. Internal recovery surrogates of 1,3,5-bromobenzene, 1,2,4,5-tetra-bromobenzene, δ -HCH, endrin ketone, PCB 30, and PCB 204 were added at the extraction step. Homogenized fat tissue was mixed with precleaned sodium sulfate to form a dry powder and Soxhlet-extracted with dichloromethane (DCM). The DCM solution was reduced in volume to approximately 2 mL. The extract was applied to the top of a gel permeation column (GPC) to remove lipids using hexane:DCM (1:1) as elution solvent. Extractable lipids were determined gravimetrically on the first

Table 1. Sample information and stable isotope (C and N) composition for 24 male and 18 female polar bears (*Ursus maritimus*) captured along Alaska's Beaufort Sea coast in spring 2003.

| Polar Bear ID No. | Sex | Age (years) | Mass (kg) | DFS (km) ^a | Longitude (°W) ^b | δ ¹⁵ N | δ ¹³ C |
|-------------------|-----|-------------|-----------|-----------------------|-----------------------------|-------------------|-------------------|
| 20170 | M | 20 | 432 | 49.9 | 149.245 | 18.3 | -18.8 |
| 20184 | M | 7 | 273 | 52.5 | 148.923 | 18.4 | -19.1 |
| 20292 | M | 7 | 239 | 23.3 | 156.749 | 20.2 | -18.7 |
| 20293 | M | 22 | 545 | 33.5 | 157.621 | 19.0 | -18.0 |
| 20294 | M | 11 | 386 | 13 | 156.655 | 19.6 | -18.4 |
| 20295 | M | 6 | 250 | 13.2 | 156.601 | 20.1 | -18.2 |
| 20296 | M | 5 | 245 | 19.2 | 156.089 | 18.5 | -18.7 |
| 20298 | M | 11 | 295 | 36.7 | 155.325 | 19.7 | -17.5 |
| 20300 | M | 1 | 64 | 44.2 | 155.085 | 20.1 | -18.7 |
| 20301 | M | 1 | 32 | 44.2 | 155.085 | 19.8 | -18.8 |
| 20424 | M | 7 | 386 | 49.8 | 148.224 | 18.7 | -19.2 |
| 20437 | M | 5 | 182 | 55.3 | 149.365 | 18.2 | -19.5 |
| 20468 | M | 11 | 420 | 32.2 | 155.523 | 18.4 | -18.8 |
| 20481 | M | 14 | 409 | 56.5 | 148.775 | 20.3 | -18.5 |
| 20498 | M | 8 | 327 | 21.8 | 143.982 | 19.1 | -18.9 |
| 20553 | M | 4 | 182 | 30.2 | 144.078 | 19.5 | -19.5 |
| 20574 | M | 3 | 170 | 7.4 | 143.441 | 19.6 | -19.0 |
| 20611 | M | 3 | 205 | 72.5 | 147.476 | 20.3 | -19.0 |
| 20615 | M | 1 | 57 | 46.3 | 147.763 | 20.0 | -18.9 |
| 20625 | M | 1 | 41 | 49.2 | 148.460 | 20.0 | -19.1 |
| 20636 | M | 10 | 364 | 85.6 | 141.867 | 19.9 | -18.9 |
| 20637 | M | 8 | 273 | 85.6 | 141.867 | 20.7 | -18.3 |
| 20645 | M | 9 | 341 | 31.4 | 143.514 | 18.4 | -19.1 |
| 20649 | M | 8 | 432 | 9.3 | 142.567 | 19.8 | -19.1 |
| 6524 | F | 22 | 220 | 32.7 | 157.747 | 19.0 | -19.1 |
| 20221 | F | 4 | 114 | 6.2 | 156.424 | 20.1 | -19.4 |
| 20222 | F | 6 | 125 | 29.1 | 155.657 | 19.9 | -19.1 |
| 20223 | F | 9 | 114 | 19.6 | 155.964 | 20.0 | -18.8 |
| 20227 | F | 7 | 182 | 28.1 | 148.107 | 19.6 | -18.7 |
| 20228 | F | 6 | 216 | 47.7 | 147.978 | 19.7 | -18.5 |
| 20299 | F | 8 | 148 | 44.2 | 155.085 | 19.7 | -18.6 |
| 20413 | F | 6 | 170 | 9.1 | 143.367 | 19.3 | -18.9 |
| 20441 | F | 12 | 205 | 33.8 | 151.619 | 19.9 | -18.5 |
| 20452 | F | 17 | 170 | 18.8 | 148.424 | 19.7 | -19.3 |
| 20485 | F | 9 | 182 | 49.2 | 148.460 | 19.7 | -19.3 |
| 20502 | F | 16 | 182 | 83 | 146.400 | 18.3 | -19.3 |
| 20525 | F | 3 | 127 | 38.2 | 142.572 | 18.6 | -19.1 |
| 20528 | F | 9 | 159 | 56.7 | 151.293 | 20.3 | -18.5 |
| 20579 | F | 2 | 125 | 32.7 | 157.747 | 18.9 | -19.4 |
| 20597 | F | 9 | 216 | 28.7 | 151.439 | 20.0 | -19.1 |
| 20600 | F | 9 | 159 | 9.8 | 153.587 | 20.4 | -18.9 |
| 20635 | F | 5 | 182 | 85.6 | 141.867 | 20.4 | -19.5 |

^aCapture distance from shore.

^bAt capture location.

150 mL of GPC eluate by evaporating off the solvent. The GPC eluate was reduced to a small volume, quantitatively exchanged into hexane, and chromatographed on activated silica gel (8 g in a 1.1 cm diameter chromatographic column) to separate PCBs from other OCs. The silica gel was activated at 350 °C for a minimum of 4 h; the sodium sulfate was cleaned by ashing at 450 °C for a minimum of 4 h. Endrin ketone and 1,3-dibromobenzene (1,3-DBB) were added to determine fractionation performance. Samples were transferred to 2,2',4-trimethylpentane (iso-octane)

and concentrated to 1000 µL. Final extracts were stored at 4 °C prior to instrumental analysis.

PCB congeners and OC pesticides were determined by high-resolution capillary gas chromatograph with electron-capture detection (GC-ECD) using a Hewlett Packard 6890 GC equipped with a 30 m × 0.25 mm, 0.25 µm film thickness DB-5 column programmed at 15 °C/min to 150 °C and 3 °C/min to 265 °C. The carrier gas was H₂ (about 1 mL/min) and the make-up gas was N₂ (40 mL/min). PCB congeners and OC pesticides were quantified by GC-ECD using a

series of external standards based on a five-point calibration curve.

Detailed presentation of all contaminants analyzed and detected in polar bear subcutaneous adipose tissue is beyond the scope of this paper and will be reported separately along with concentrations in blood. This study was limited to a few contaminants and their by-products. Contaminants below detection limits, most co-eluting chemicals, and those known to have low biomagnification factors in the polar bear food web (Fisk et al. 2001; Hoekstra et al. 2003b) were not used. We examined OCs that have high food web biomagnification factors and tend to be recalcitrant in marine mammals, including contaminant groups (Σ PCB, Σ CHL, Σ DDT, and Σ HCH) and congeners (PCB 153, PCB 180, PCB194) and metabolites (oxychlordane, *p,p*-dichlorodiphenyldichlorethylene (*p,p*-DDE), and β -HCH). All of these compounds are found at high levels in polar bear adipose tissue or in their prey and may be expected to respond to differences in feeding ecology.

We used analysis of covariance (ANCOVA) to detect effects of age and sex on the stable isotope signatures independent of contaminants. Equality of variances and the assumption of normality was tested with a Shapiro–Wilk test ($P > 0.05$). We log-transformed contaminant data to meet the regression assumptions and used multiple regression with stepwise model selection ($\alpha_{\text{enter}} = 0.15$) to examine relationships between contaminant concentrations in bears and their age and location of capture (longitude and distance from shore), as well as their $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ signatures. Males and females were analyzed separately because of physiological differences in biotransformation and excretion of OCs between the sexes. Prior to model building, we limited our models to the five independent biological variables likely to affect contaminant concentrations ($\alpha_{\text{overall}} = 0.15$).

Because age and mass were correlated (Pearson's correlation, $r = 0.573$, $n = 41$, $P < 0.0001$), a multiple regression model could not be used to test for the simultaneous effects of age and mass on OC concentration. Therefore, we selected age for further comparisons because it has been clearly linked with changes in OC concentration in previous studies (Norstrom et al. 1988; Bernhoft et al. 1997). Longitude of the capture location was also excluded because of collinearity with $\delta^{13}\text{C}$ ($F_{[1,41]} = 7.10$, $P = 0.0111$). If age was significant, we reran the model without 1-year-olds to address their large effects on the relationship. Although there were only four male and no female 1-year-olds, cubs had comparatively high $\delta^{15}\text{N}$ values and correspondingly high contaminant concentrations (Polischuk et al. 2001, 2002). Eliminating 1-year-olds from models reduced variability due to maternal effects of lactation and nursing, which allowed us to better determine age-related effects in juveniles and adults.

Like Hoekstra et al. (2003b), we calculated the ratio of the concentration of OCs to that of the recalcitrant congener PCB153 (OCx/CP153) to compare contaminant profiles of polar bears with those of ringed seals and bowhead whales as an example of food items differing in trophic position. All analyses were conducted using SAS[®] version 8.0 (SAS Institute Inc. 1990) and significance levels were set at $\alpha = 0.05$.

Results

The $\delta^{15}\text{N}$ values ranged from 18.1‰ to 20.7‰ in male polar bears and from 18.3‰ to 20.4‰ in females, spanning trophic levels 4.2–4.9, as determined using methods in Hoekstra et al. (2002a). We found no significant variation in $\delta^{15}\text{N}$ values between the sexes ($F_{[1,39]} = 1.53$, $P = 0.263$), ages ($F_{[1,39]} = 2.53$, $P = 0.095$), or their interaction. The $\delta^{13}\text{C}$ values of polar bears ranged from -19.5 ‰ to -17.5 ‰ and did not differ by sex ($F_{[1,39]} = 3.63$, $P = 0.064$) or age ($F_{[1,39]} = 3.89$, $P = 0.056$). The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values were not significantly correlated in either sex ($P > 0.05$).

In both sexes, wet-mass concentrations of Σ PCB, PCB 153, PCB 180, Σ CHL, oxychlordane, Σ DDT, and *p,p*-DDE were compared with biological factors, including trophic level measured as the $\delta^{15}\text{N}$ value. Σ PCB is the total concentration of all 104 PCBs analyzed (Table 2). PCBs 153 and 180 composed 26% and 12% of Σ PCB, respectively. PCB 194 concentrations were low in these polar bears. Oxychlordane (79% of Σ CHL) was the dominant metabolite of CHL, β -HCH composed 82% of Σ HCH, and although concentrations were low, *p,p*-DDE composed 74% of Σ DDT.

The rank order of these contaminants by mean concentration, Σ PCB > PCB 153 > Σ CHL > oxychlordane > PCB 180 > Σ HCH > β -HCH > Σ DDT > PCB 194 > *p,p*-DDE, was the same for males and females. Overall, lipid content of the fat samples was the most important factor in explaining variation in most contaminant concentrations (wet mass) and was included as a significant explanatory variable for all models except *p,p*-DDE in female bears (Table 3). Lipid content alone accounted for up to 62% of the variation in contaminant burdens as measured by r^2 values. Polar bear age was strongly correlated with many OC concentrations. In males, PCB 153, Σ CHL, and oxychlordane concentrations decreased with age. However, when the four 1-year-old males were eliminated from the regression, only Σ CHL and oxychlordane decreased with age in males. Among females, all OC concentrations reported here, except PCB194, decreased significantly with age (Table 3).

In both sexes the relationship between $\delta^{15}\text{N}$ value and OC concentration was positively correlated in several contaminant groups. The concentration of all PCB congeners studied increased significantly with $\delta^{15}\text{N}$ value, and the $\delta^{15}\text{N}$ value was the most important variable in explaining the variation in PCB concentration in male bears (Table 3). In males, $\delta^{15}\text{N}$ value alone accounted for 38%, 40%, and 28% of the variation in the concentrations of PCBs 153, 180, and 194, respectively (Figs. 2A–2D). Σ CHL, the most abundant OC pesticide we found in polar bear adipose tissue, also increased in concentration with $\delta^{15}\text{N}$ value in both sexes, but the relationship was not as strong as for PCBs (Figs. 2E and 2F). Although contaminant concentrations of some bears appear as outliers from the trends, they were not consistently correlated with age, sex, or reproductive status. Weak relationships were found between $\delta^{15}\text{N}$ value and Σ DDT, *p,p*-DDE, and Σ HCH concentrations among males only. Oxychlordane concentrations were positively correlated with $\delta^{15}\text{N}$ values among females only (Table 3).

Although neither $\delta^{13}\text{C}$ value nor capture distance from shore was as important as lipid content, age, or $\delta^{15}\text{N}$ value, incorporating these factors improved the overall fit of our

Table 2. Major OC analyte concentrations (mean, minimum, maximum, and SD) in subcutaneous adipose tissue (ng/g wet mass), lipid content, and stable $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values (‰) in cellular portion of blood of polar bears (*Ursus maritimus*) from Alaska's Beaufort Sea coast collected in spring 2003.

| | ΣPCB | PCB 153 | PCB 180 | PCB 194 | $\Sigma\text{Chlordane}$ | Oxychlordane | ΣDDT | <i>p,p</i> -DDE | ΣHCH | $\beta\text{-HCH}$ | Percent lipid | $\delta^{15}\text{N}$ | $\delta^{13}\text{C}$ |
|-------------------------|--------------------|---------|---------|---------|--------------------------|--------------|--------------------|-----------------|--------------------|--------------------|---------------|-----------------------|-----------------------|
| Males (n = 24) | | | | | | | | | | | | | |
| Mean | 2777.7 | 758.8 | 284.4 | 5.4 | 379.5 | 288.5 | 55.4 | 41.8 | 180.8 | 154.6 | 29.8 | 19.4 | -18.8 |
| Maximum | 6123.2 | 2105.0 | 831.0 | 10.6 | 1601.5 | 1306.2 | 136.1 | 114.0 | 464.6 | 391.0 | 62.3 | 20.7 | -17.5 |
| Minimum | 797.1 | 208.0 | 73.9 | 0.9 | 42.2 | 23.5 | 5.8 | 3.9 | 50.3 | 43.6 | 5.9 | 18.2 | -19.5 |
| SD | 1557.3 | 471.5 | 189.5 | 2.5 | 383.7 | 322.2 | 34.9 | 27.8 | 89.2 | 76.8 | 15.6 | 0.8 | 0.5 |
| Females (n = 18) | | | | | | | | | | | | | |
| Mean | 3071.5 | 728.1 | 272.3 | 5.5 | 773.3 | 630.8 | 62.8 | 46.5 | 169.2 | 130.9 | 37.4 | 19.6 | -19.0 |
| Maximum | 7234.4 | 2004.0 | 588.0 | 9.8 | 1932.8 | 1514.0 | 181.0 | 145.0 | 443.6 | 353.0 | 74.9 | 20.4 | -18.5 |
| Minimum | 836.1 | 217.0 | 66.2 | 1.6 | 126.0 | 88.2 | 9.0 | 4.9 | 39.0 | 34.4 | 6.2 | 18.3 | -19.5 |
| SD | 1821.6 | 421.3 | 141.0 | 2.8 | 471.3 | 381.4 | 46.1 | 37.4 | 107.4 | 84.3 | 21.8 | 0.6 | 0.3 |

Note: ΣPCB is the sum of PCBs 1, 3, 6, 18, 19, 22, 25, 26, 40, 42, 43, 44, 45, 46, 49, 50, 51, 52, 53, 55, 59, 63, 66, 74, 82, 83, 84, 85, 91, 92, 95, 97, 99, 100, 101, 105, 107, 110, 114, 118, 119, 128, 129, 130, 132, 133, 136, 137, 141, 146, 147, 149, 151, 153, 156, 158, 167, 172, 173, 174, 175, 176, 177, 178, 179, 180, 183, 185, 189, 191, 193, 194, 197, 198, 205, 206, 207, 209, 4-10, 7-9, 8(5), 12(13), 134-131, 135-144, 15-17, 157-201, 163-138, 16-32, 170-190, 182-187, 200, 201, 202-171, 203-196, 208-195, 24-27, 31-28, 33-20, 47-48, 54-29, 56-60, 70-76-98, 71-41-64, and 81-87; $\Sigma\text{chlordane}$ is the sum of oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide; ΣDDT is the sum of *p,p*-DDT, *p,p*-DDD, and *p,p*-DDE; and ΣHCH is the sum of the α -, β -, and γ -hexachlorocyclohexanes.

models and they were correlated with concentrations of several contaminants. ΣPCB , ΣCHL , oxychlordane, ΣDDT , and *p,p*-DDE concentrations decreased with $\delta^{13}\text{C}$ value in female bears and PCB180 concentrations decreased slightly in males. Capture distance from shore also improved overall model fit. PCB 153, ΣCHL , oxychlordane, ΣHCH , and $\beta\text{-HCH}$ concentrations decreased slightly with capture distance from shore in female bears. In males *p,p*-DDE showed a weak negative correlation with distance from shore, which improved the fit of the overall model (Table 3).

PCB 153 was the dominant congener in polar bear adipose tissue (Fig. 3). Although some CHL, HCHs and DDTs are considered recalcitrant, they did not accumulate in polar bear fat relative to PCB 153. Ratios of ΣCHL , ΣDDT , and ΣHCH to PCB 153 were significantly lower than 1 in bears ($P < 0.05$), and ratios of ΣCHL and ΣDDT were greater than 1 in ringed seals and bowhead whales ($P < 0.05$). ΣHCH ratios were similar to PCB 153 in ringed seals, but ΣHCH to PCB 153 ratios were higher in bowhead whales ($P < 0.05$). Among the three species, ratios of ΣCHL and ΣDDT to PCB153 were significantly lower in polar bears than in their prey (ΣCHL , $F_{[3,190]} = 51.81$, $P < 0.001$; ΣDDT , $F_{[3,190]} = 142.4$, $P < 0.001$).

Discussion

The entirely carnivorous, lipid-rich diet of polar bears exposes them to an increased risk of toxic effects from organic pollutants. Although Alaskan polar bears have lower concentrations of OC contaminants than many other subpopulations (Andersen et al. 2001), we found a wide variation among individuals (ΣPCB : 797.1–6123.2 ng/g wet mass). This was consistent with previous studies from this area by Kucklick et al. (2002) and Kannan et al. (2005). The variation in ΣPCB concentrations in polar bears exceeds that reported for ringed seals and bowhead whales sampled along the north coast of Alaska (Hoekstra et al. 2002a; Kucklick et al. 2002). Because polar bear age, sex, condition, or location of capture did not explain this variation among individuals, Kucklick et al. (2002) suggested that polar bears are not exclusively feeding at the top of the arctic marine food web. Polar bears captured along Alaska's Beaufort and Chukchi sea coasts may have greater access to lower trophic level prey than many other polar bear populations (Bentzen et al. 2007). We suggest that lower values within this wide range of contaminant concentrations may be explained by the use of foods such as walrus, bearded seal, and remains of hunter-killed or stranded bowhead whales.

Using models generated with black bear (*Ursus americanus* Pallas, 1780) data, Hilderbrand et al. (1996) calculated an increase of approximately 3.2‰ in $\delta^{15}\text{N}$ signatures from prey to the cellular portions of blood in polar bears, for a diet consisting of ringed seals. Using these values, the mean $\delta^{15}\text{N}$ signatures found in polar bears were lower than expected for a diet consisting exclusively of ringed seal (Hoekstra et al. 2003b). Dietary mixing models using stable isotopes of C and N to determine the chemical feeding ecology of the bears in this study suggest a diet composed of 11%–26% lower trophic level prey in the winter of 2002–2003 (Bentzen et al. 2007). This apparent use of lower trophic level prey coincides with a particular abundance of

Table 3. Overall model results for variables used in multiple regressions of OC concentrations in Alaskan polar bear (*Ursus maritimus*) adipose tissue (wet mass) in spring 2003.

| | Percent lipid | Age | $\delta^{15}\text{N}$ | Overall model |
|--------------------------------------|----------------------|----------------------|-----------------------|---------------------------------------------|
| ΣPCB | | | | |
| Males | 1.75, 0.094, 0.08 | — | 3.51, 0.002, 0.38 | $F_{[2,21]} = 8.87, P = 0.002, r^2 = 0.46$ |
| Females | 6.11, <0.0001, 0.23 | -3.87, 0.0019, 0.28 | 4.30, 0.0009, 0.21 | $F_{[4,13]} = 14.13, P < 0.001, r^2 = 0.81$ |
| PCB153 | | | | |
| Males | 3.38, 0.0030, 0.21 | -1.95, 0.0652, 0.06 | 3.48, 0.0023, 0.39 | $F_{[3,20]} = 13.43, P < 0.001, r^2 = 0.67$ |
| Females | 8.09, 0.0117, 0.50 | -3.38, 0.0049, 0.01 | 2.93, 0.0117, 0.12 | $F_{[4,13]} = 18.31, P < 0.001, r^2 = 0.85$ |
| PCB180 | | | | |
| Males | 3.23, 0.0042, 0.17 | — | 4.34, 0.0003, 0.40 | $F_{[3,20]} = 10.69, P < 0.001, r^2 = 0.62$ |
| Females | 4.66, 0.0004, 0.37 | -2.23, 0.0428, 0.16 | 2.09, 0.0557, 0.11 | $F_{[3,14]} = 8.22, P = 0.002, r^2 = 0.64$ |
| PCB194 | | | | |
| Males | 2.31, 0.0309, 0.15 | — | 2.84, 0.0099, 0.28 | $F_{[2,21]} = 7.92, P = 0.003, r^2 = 0.43$ |
| Females | 3.31, 0.0047, 0.33 | — | 1.52, 0.1496, 0.09 | $F_{[2,17]} = 5.50, P = 0.016, r^2 = 0.42$ |
| ΣCHL | | | | |
| Males | 4.84, <0.0001, 0.30 | -4.76, 0.1357, 0.44 | 1.55, 0.1357, 0.03 | $F_{[3,20]} = 22.17, P < 0.001, r^2 = 0.77$ |
| Females | 13.52, <0.0001, 0.53 | -5.96, <0.0001, 0.20 | 4.88, 0.0004, 0.08 | $F_{[5,12]} = 43.53, P < 0.001, r^2 = 0.95$ |
| OXCHL | | | | |
| Males | 6.50, <0.0001, 0.28 | -7.43, <0.0001, 0.46 | — | $F_{[2,21]} = 48.35, P < 0.001, r^2 = 0.76$ |
| Females | 11.85, <0.0001, 0.52 | -4.97, 0.0003, 0.18 | 4.28, 0.0011, 0.08 | $F_{[5,12]} = 33.23, P < 0.001, r^2 = 0.93$ |
| ΣDDT | | | | |
| Males | 2.08, 0.0501, 0.19 | — | 1.99, 0.0592, 0.13 | $F_{[2,21]} = 4.92, P = 0.018, r^2 = 0.32$ |
| Females | 7.06, <0.0001, 0.59 | -3.21, 0.0063, 0.14 | — | $F_{[3,14]} = 19.83, P < 0.001, r^2 = 0.81$ |
| <i>p,p</i>-DDE | | | | |
| Males | — | — | 3.20, 0.0043, 0.25 | $F_{[2,21]} = 6.19, P = 0.008, r^2 = 0.37$ |
| Females | 6.29, <0.0001, 0.55 | -2.91, 0.0113, 0.13 | — | $F_{[3,14]} = 16.17, P < 0.001, r^2 = 0.78$ |
| ΣHCH | | | | |
| Males | 5.67, <0.0001, 0.61 | — | 1.50, 0.14931, 0.04 | $F_{[2,21]} = 18.98, P < 0.001, r^2 = 0.64$ |
| Females | 10.91, <0.0001, 0.62 | -5.24, 0.0001, 0.20 | — | $F_{[3,14]} = 44.43, P < 0.001, r^2 = 0.90$ |
| $\beta\text{-HCH}$ | | | | |
| Males | 5.57, <0.0001, 0.59 | — | — | $F_{[1,22]} = 31.02, P < 0.001, r^2 = 0.59$ |
| Females | 9.12, <0.0001, 0.59 | -4.48, 0.0005, 0.20 | — | $F_{[3,14]} = 31.19, P < 0.001, r^2 = 0.87$ |

Note: The order of values for all variables is *tl*, *P*, *r*²; a blank cell indicates nonsignificant variables (*P* > 0.15); $\delta^{13}\text{C}$ was a significant variable among ΣPCB (-2.54, 0.0247, 0.09), ΣCHL (-1.60, 0.1263, 0.05), oxychlorane (-4.01, 0.0017, 0.07), ΣDDT (-2.36, 0.0336, 0.08), and *p,p*-DDE (-2.32, 0.0362, 0.09) concentrations for females and PCB180 (-1.60, 0.1263, 0.05) concentrations for males; capture distance from shore was significant among PCB153 (-2.28, 0.0401, 0.06), ΣCHL (-3.69, 0.0031, 0.06), oxychlorane (-3.69, 0.0031, 0.08), ΣHCH (-3.53, 0.0033, 0.08), and $\beta\text{-HCH}$ (-2.91, 0.0113, 0.08) concentrations for females and *p,p*-DDE (-2.01, 0.0579, 0.12) concentrations for males.

bowhead whale remains associated with the subsistence hunts of Barrow, Nuiqsut, and Kaktovik, Alaska, which landed 26 whales in the fall prior to our sampling (Suydam et al. 2003). After the whales are butchered, varying amounts of hard and soft tissues are frequently available to scavengers on shore or on floating sea ice and attract large numbers of bears. In September and October 2002, Miller et al. (2004) recorded aggregations of up to 44 bears at bowhead carcasses near the village of Kaktovik and 4 bears at bowhead remains at Cross Island (70°29'N, 147°59'W), the base for the Nuiqsut whale hunt. Over 60 bears were observed at this time around the whale remains at Point Barrow, Alaska. During winter, individual bears were observed feeding on these carcasses at Barrow and other sites, and some polar bears continued to visit these carcasses through March 2003 (Bentzen et al. 2007).

Although we did not find differences in C or N signatures between the sexes during this study, like Dietz et al. (2004),

Norstrom et al. (1998), and Letcher et al. (1998), we analyzed the sexes separately because of possible dietary, behavioral, and physiological differences associated with reproductive status and lactation, which may have affected feeding ecology or OC concentrations in tissues. The $\delta^{15}\text{N}$ values of 96 polar bears collected in this area in 2004 (Bentzen et al. 2007) indicate significant differences between males and females all along the coast and support the decision to separate the sexes in this analysis.

The $\delta^{13}\text{C}$ values were negatively correlated with longitude of capture, and this is likely explained by chemical differences between the Beaufort, Chukchi, and Bering seas due to the geographic heterogeneity of stable carbon isotopes in zooplankton, fish, and mammals in the polar bear food chain (Hoekstra et al. 2003b; Dehn et al. 2005a). Our data support previous studies reporting higher $\delta^{13}\text{C}$ values among bears, seals, and bowhead whales feeding in the Chukchi Sea

Fig. 2. Concentrations of Σ PCB (A), PCB 153 (B), PCB 180 (C), PCB 194 (D), Σ DDT (E), and Σ chlordane (F) in subcutaneous adipose tissue (ng/g wet mass) in relation to $\delta^{15}\text{N}$ values (‰ in packed blood cells) in male and female polar bears (*Ursus maritimus*) sampled along the north coast of Alaska in spring 2003.

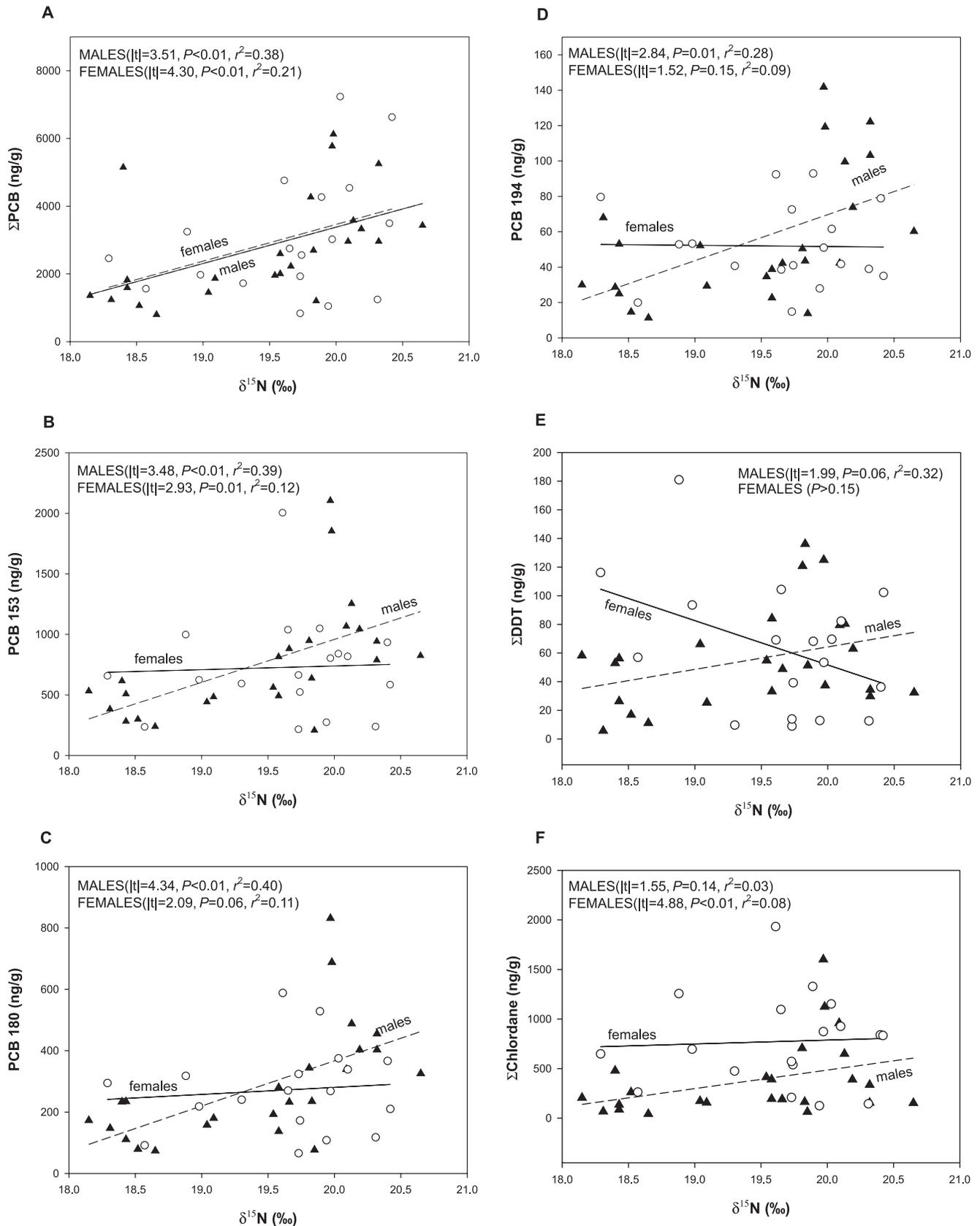
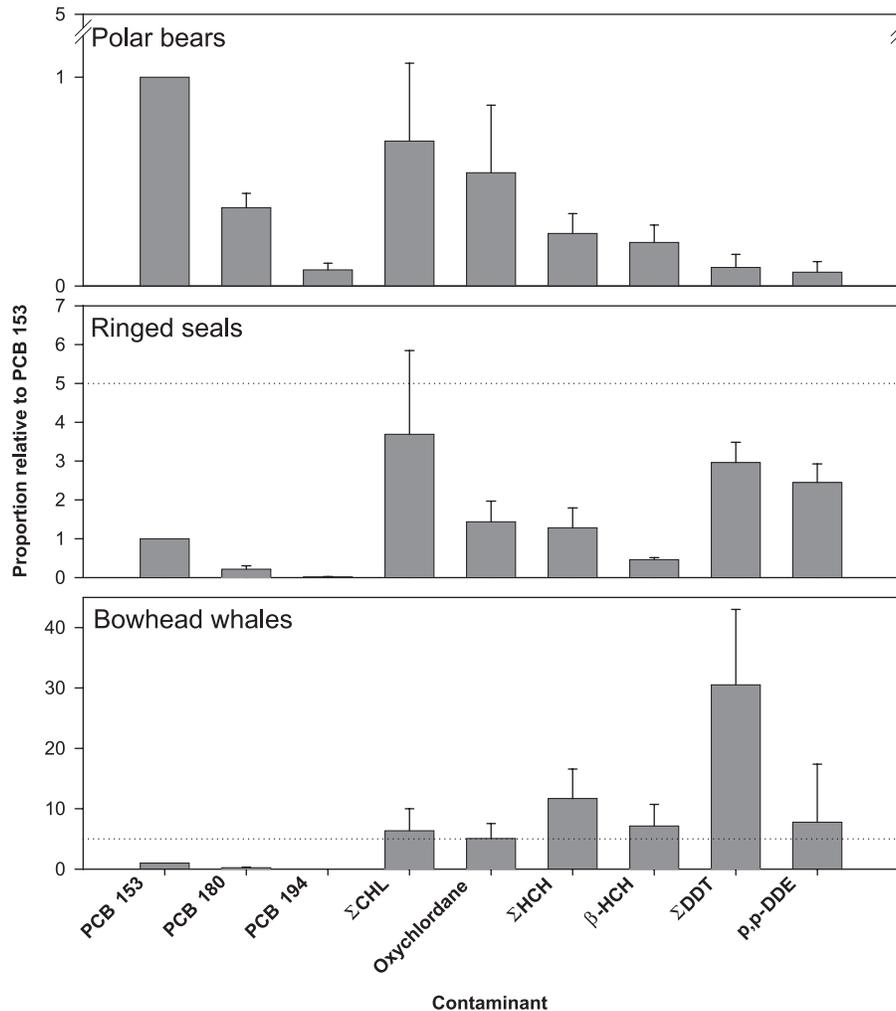


Fig. 3. Wet mass contaminant profiles (and SD for polar bears, *Ursus maritimus*, ringed seals, *Phoca hispida*, and bowhead whales, *Balaena mysticetus*) relative to PCB 153 (OCx/CB153) in polar bear adipose tissue, ringed seal blubber (Kucklick et al. 2002), and bowhead whale blubber (Hoekstra et al. 2002b) collected along the north coast of Alaska (1997–2003).



relative to those in the Beaufort Sea (Hobson and Schell 1998; Hoekstra et al. 2002a; Dehn et al. 2005a; Bentzen et al. 2007). Because of this geographic difference, the highly mobile nature of these bears (Garner et al. 1990; Amstrup et al. 2000; Ferguson et al. 2000; Durner et al. 2004), and the global distribution of these contaminants (de Wit et al. 2004), we hypothesize that the $\delta^{13}\text{C}$ value, reflecting several months' feeding, was a better indicator of the general area occupied by the bears than their specific location at capture. However, polar bears feeding farther west into the Chukchi Sea may have greater access to benthic feeding prey such as bearded seal, walrus, and gray whale. These benthic feeding species have ($\approx 2.0\text{‰}$) higher $\delta^{13}\text{C}$ signatures relative to ringed seal, which may also account for some of the differences due to longitude of polar bear capture (Hoekstra et al. 2002a; Dehn et al. 2005b; Bentzen 2006).

Geographic comparison of isotopes and OC concentrations

The $\delta^{15}\text{N}$ signatures of polar bears in this report averaged more than 1.75‰ lower than values reported for adult bears sampled in Resolute Bay ($|t|_{48} = 28.03$, $P < 0.01$; Polischuk

et al. 2001) and Lancaster Sound ($|t|_{44} = 15.87$, $P < 0.01$; Hobson and Welch 1992) in northern Canada. This difference may result from the greater availability of lower trophic level prey to polar bears along the north coast of Alaska, but may also be due to differences in fractionation factors associated with blood cells, blood plasma, and muscle tissue analyzed in those studies.

Alaskan populations of polar bears generally have lower concentrations of OC contaminants than polar bear populations throughout the Arctic (de Wit et al. 2004). We found PCB concentrations similar to those reported by Kucklick et al. (2002), as well as low ΣCHL and ΣDDT concentrations relative to populations in the Canadian, Norwegian, and Russian Arctic (Norstrom et al. 1998; Muir et al. 1999; Lie et al. 2003; Kannan et al. 2005). The exception was HCH, which tends to occur at relatively higher concentrations in tissues from Chukchi and Beaufort sea polar bear than in tissues from other populations. The higher HCH concentrations are likely a result of transport from recent applications of HCH in Asia, and are consistent with trends in other marine mammals from northern Alaska (Muir et al. 2000; Hoekstra et al. 2002b; Lie et al. 2003; de Wit et al. 2004).

Although $\delta^{13}\text{C}$ values explained little variation in contaminant concentrations, it is important to note the overlap of the southern Beaufort and Chukchi sea polar bear populations around Barrow, where much of our sampling occurred (Amstrup et al. 2004). Although access to bowhead whale carcasses may be similar from Barrow to Kaktovik, we expected that Chukchi Sea bears have a more variable diet, including greater access to walrus and other lower trophic level prey. Using $\delta^{13}\text{C}$ values as an indicator of Beaufort Sea versus Chukchi Sea prey-based feeding, we did not find strong evidence of differences in contaminant concentrations (Table 3). However, decreases in concentrations of compounds such as ΣCHL and oxychlorane among females westward toward the Chukchi Sea may support the possibility of a more diverse diet among bears in this area, including greater use of lower trophic level, carbon-enriched, benthic feeders.

We modeled capture distance from shore to examine whether feeding ecology, specifically the coastal use of lower trophic level prey, was greater in bears captured closer to shore. Our results were inconclusive, but higher concentrations of OC pesticides, but not PCBs, in nearshore bears may be an indication of bears scavenging bowhead remains. Because capture distances were small relative to movement patterns, we stress that specific capture locations are unlikely to be an accurate proxy for the measure of pelagic versus coastal habitat use.

Lipid content and age

The effects of lipid content and age on OC contaminant concentrations are well documented (Bernhoft et al. 1997; Norstrom et al. 1998; Fisk et al. 2001). Although studies indicate that most OC concentrations in polar bears are affected by age, relationships are not clear and trends vary among OC groups, likely depending on various ecological and physiological factors of biotransformation and lipid-driven toxicodistribution. Like those of Norstrom et al. (1998), our data indicate a slight decrease in ΣPCB concentration with age, but this effect remains inconclusive (Norstrom et al. 1998; Dietz et al. 2004; Kannan et al. 2005). The notable decreasing trend of ΣCHL concentration with age that we observed was also reported by Dietz et al. (2004) among east Greenland polar bears. However, Bernhoft et al. (1997) found the opposite among Svalbard polar bears.

It is understood that increased lipid content strongly affects OC concentration along with lipid-driven toxicodistribution in upper trophic level organisms. However, there is clear evidence that biomagnification occurs in addition to differences associated with the lipid content of tissues analyzed (Kidd et al. 1998; Fisk et al. 2001). As expected, the lipid content of the tissues analyzed was a significant factor in explaining the variation in concentration among all OCs analyzed. We found percent lipid to be the single most important factor in explaining variation in ΣCHL , ΣDDT , and ΣHCH concentrations, but not on a consistent basis. In some cases $\delta^{15}\text{N}$ value and (or) age were the major drivers of relationships (Table 3). For less recalcitrant $\beta\text{-HCH}$ ($|t| = 5.52$, $P < 0.01$, $r^2 = 0.59$), more than half of the observed variation in concentration among males was explained by lipid content alone. We did not lipid-normalize wet-mass concen-

trations because concentrations of the contaminants studied are not consistently correlated with percent lipid, which varyingly accounted for differences in contaminant concentrations. Because of the range of lipid relationships the addition of simple lipid-normalization factors (e.g., percent mass) would unnecessarily bias the data. By accounting for lipid content and polar bear age within the statistical models we were better able to detect the effects of trophic-level differences on OC concentrations.

Trophic-level effects

ΣPCBs , more specifically PCBs 153 and 180, dominated the OC profile of polar bear adipose tissue. The relationship between $\delta^{15}\text{N}$ value and OC concentrations has been used to explain the effect of trophic level on biomagnification of OCs in the marine food web (Hobson et al. 1995; Fisk et al. 2001; Moisey et al. 2001; Hoekstra et al. 2003b). The significant correlations of $\delta^{15}\text{N}$ value and concentrations of several of these contaminants in both male and female polar bears provide supporting evidence that PCB burdens increase with trophic level among southern Beaufort Sea polar bears (Table 3). ΣPCB , PCB 153, PCB 180, and PCB 194 concentrations all increased significantly with trophic level, measured as $\delta^{15}\text{N}$ value. Models including percent lipid and $\delta^{15}\text{N}$ values explained 46%, 67%, 62%, and 43% of the respective variation in the concentrations of these PCBs among male bears. Not surprisingly, PCB 153, the most recalcitrant of PCBs, had the highest r^2 value, and $\delta^{15}\text{N}$ alone accounted for 39% of the variation. Among females, models also yielded significant results. However age, $\delta^{13}\text{C}$ value, and capture distance from shore were more important in explaining variation in contaminant concentrations among females than among males. Differences between the sexes may be caused by the more complex contaminant pathways among females due to reproduction, extended fasting periods while denning, and differences in biotransformation and elimination of contaminants through lactation (Letcher et al. 1998; Norstrom et al. 1998).

Although blood is easily collected in the field, and generally represents the whole-body isotopic signature of an organism, the turnover rates of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in the cellular portions of blood limit dietary inferences to the period within 4 months of sampling (Hilderbrand et al. 1996; Kurle and Worthy 2002). Concentrations of some OCs in fat may represent accumulation over several years, especially among male bears, which are unable to eliminate contaminants through lactation. Although not available for this study, tissues such as bone, which have slower turnover rates of C and N, may provide additional insight into the relationships between long-term diet and contaminant concentrations.

As a group, the OC pesticides likely undergo more biotransformation and elimination by polar bears than PCB 153, and this was reflected in the differences in contaminant profiles relative to prey examples (Fig. 3). We compared contaminants relative to the wet-mass concentrations of PCB 153 in polar bear adipose tissue with blubber in ringed seal (Kucklick et al. 2002) and bowhead whale (Hoekstra et al. 2002b), assuming that the highly recalcitrant PCB 153 has the maximum possible biomagnification factor (Letcher

et al. 1998). Contaminant profiles (proportions) are more similar among individuals than actual concentrations. Therefore, normalizing the data relative to PCB 153 reduced variation in concentration differences among individuals (Letcher et al. 1995; Wiberg et al. 2000). Because Kucklick et al. (2002) and Hoekstra et al. (2002b) analyzed different numbers of congeners, reporting contaminants relative to total OC concentrations would have been inappropriate. Like Letcher et al. (1998) and Kucklick et al. (2002), we found that DDT and HCH formed a much smaller proportion of contaminants in polar bears relative to ringed seals and bowhead whales, and did not accumulate in polar bear fat relative to PCB 153 (Fig. 3). PCB 194 concentrations were also low in these polar bears despite the high biomagnification factor in many marine mammals (Bernhoft et al. 1997; Norstrom et al. 1998; Corsolini et al. 2000; Skaare et al. 2000; Braathen et al. 2004).

In multiple regressions on Σ CHL, oxychlorane, Σ HCH, β -HCH, Σ DDT, and *p,p*-DDE, we found little or no influence of $\delta^{15}\text{N}$ value within either sex, which may be explained partly by the ability of polar bears to biotransform some OCs via hepatic mixed function oxidases and excrete many potentially bioaccumulative compounds that most marine mammals cannot (Letcher et al. 1996, 1998). Although we found that our models explained the variation in Σ CHL and oxychlorane concentrations well (Σ CHL: $r^2 = 0.77$ for males and $r^2 = 0.95$ for females; oxychlorane: $r^2 = 0.76$ for males and $r^2 = 0.93$ for females), the effect of $\delta^{15}\text{N}$ was not as significant as that of the PCBs. Among males, when 1-year-olds were removed from the analysis, $\delta^{15}\text{N}$ value was no longer a significant explanatory variable. This strong decreasing trend with age is in agreement with Dietz et al. (2004). Although Σ CHL are the most abundant group of OC pesticides found in polar bear adipose tissue, the proportions relative to PCB 153 are much lower than in their prey. It is known that high concentrations of Σ PCBs may induce hepatic cytochrome P-450s, which are instrumental in the metabolizing of many organic pollutants by polar bears (Letcher et al. 1998). Therefore, activation of hepatic cytochrome P-450 isozymes, in turn, may also increase this biotransformation and clearance of CHL and other related compounds.

Although high concentrations of Σ DDT and *p,p*-DDE have been reported in ringed seals, they were not strongly correlated with $\delta^{15}\text{N}$ values in the polar bears we sampled. The relatively lower concentrations of *p,p*-DDE that we observed, and our inability to detect an increase with trophic level, are likely due to the ability of polar bears to biotransform many OC compounds through induction of hepatic mixed function oxidases (Letcher et al. 1995, 1998; Norstrom et al. 1998). Although polar bears do contain high concentrations of DDT relative to many other OC pesticides, our results support the assumption that they are not a good indicator of polar bear feeding ecology or of exposure of polar bears to DDT-related compounds via their prey. The lipid percentages in the samples explained most of the variation in HCH concentration among all bears, and no trophic-level effects were detected. HCHs vary in recalcitrance and are more water-soluble than many other OCs. Although they are found at high levels in the arctic marine ecosystem, our results support evidence

that HCHs are readily eliminated (Hoekstra et al. 2003b) and therefore less likely to undergo biomagnification with trophic level than other more lipophilic and recalcitrant compounds.

Conclusions

Strong positive relationships were found between the concentrations of several highly recalcitrant OC contaminants (wet mass) and trophic level, as determined from $\delta^{15}\text{N}$ analysis of polar bear blood cells. This emphasizes the role of polar bears' diet and the biomagnification and subsequent tissue concentrations of these compounds. The differences we observed in trophic level and OC concentrations may be influenced by the consumption of lower trophic level prey, including walrus and bowhead whale carcasses, which contain lower concentrations of most OC contaminants than ringed seal. The availability of bowhead whale carcasses to scavenging polar bears along the Beaufort Sea coast was high during this study, and these may have represented a dietary source that is lower in contaminants than ringed seals and is relatively unavailable to other polar bear populations. The winter availability of walrus and gray whale was assumed to be low in the southern Beaufort Sea, and their dietary contributions have not been fully assessed in this study. However, the use of lower trophic level prey may partly explain the lower $\delta^{15}\text{N}$ signatures and concentrations of these contaminants observed in polar bears captured along Alaska's Beaufort Sea coast relative to those of other polar bear populations.

The comparison of polar bear contaminant profiles with those of ringed seal and bowhead whale provides additional evidence that many compounds are biotransformed and eliminated in polar bears relative to PCB 153 (Wiberg et al. 2000). Because polar bears have a greater ability to biotransform and eliminate most OC pesticides than other marine mammals, the value of OCs in feeding ecology and biomagnification studies may be limited. Without a better understanding of the lipid-driven toxicodistribution, metabolism, and excretion of OCs in polar bears, studying OC biomagnification in adipose tissue may not be an adequate proxy for directly assessing dietary exposure. Although polar bears are a top carnivore in the arctic marine ecosystem, and some OCs are found at very high concentrations in their tissues, their ability to biotransform and excrete many OC pesticides, which their prey cannot, may make them poorly suited to be an indicator species of levels of these compounds in the arctic marine food web.

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