Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort–Chukchi Seas


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Received 27 August 2002; accepted 5 December 2002

“Capsule”: The trophic status and biomagnification of persistent OCs within the near-shore Beaufort–Chukchi Seas food web from Barrow, AK is discussed.

Abstract
Stable isotope values ($\delta^{13}C$, $\delta^{15}N$) and concentrations of persistent organochlorine contaminants (OCs) were determined to evaluate the near-shore marine trophic status of biota and biomagnification of OCs from the southern Beaufort–Chukchi Seas (1999–2000) near Barrow, AK. The biota examined included zooplankton (Calanus spp.), fish species such as arctic cod (Boreogadus saida), arctic char (Salvelinus alpinus), pink salmon (Oncorhynchus gorbuscha), and fourhorn sculpin (Myoxocephalus quadricornis), along with marine mammals, including bowhead whales (Balaena mysticetus), beluga whales (Delphinapterus leucas), ringed seals (Phoca hispida) and bearded seals (Erignathus barbatus). The isotopically derived trophic position of biota from the Beaufort–Chukchi Seas marine food web, avian fauna excluded, is similar to other coastal food webs in the Arctic. Concentrations of OCs in marine mammals were significantly greater than in fish and corresponded with determined trophic level. In general, OCs with the greatest food web magnification factors (FWMFs) were those either formed due to biotransformation (e.g. $p,p'$-DDE, oxychlorodane) or considered recalcitrant (e.g. $\beta$-HCH, 2,4,5-Cl substituted PCBs) in most biota, whereas concentrations of OCs that are considered to be readily eliminated (e.g. $\gamma$-HCH) did not correlate with trophic level. Differences in physical–chemical properties of OCs, feeding strategy and possible biotransformation were reflected in the variable biomagnification between fish and marine mammals. The FWMFs in the Beaufort–Chukchi Seas region were consistent with reported values in the Canadian Arctic and temperate food webs, but were statistically different than FWMFs from the Barents and White Seas, indicating that the spatial variability of OC contamination in top-level marine Arctic predators is attributed to differences in regional sources of contamination rather than trophic position.

Keywords: Biomagnification; Cetaceans; Fish; Bioaccumulation; Organochlorines; Pinnipeds

1. Introduction
The Arctic has become a sink for various anthropogenic compounds, such as persistent organochlorine contaminants (OCs), that have originated from limited use within this region and release from temperate environments with subsequent long-range transport and deposition via atmospheric and oceanic currents (Macdonald et al., 2000). Due to their persistence and hydrophobicity, OCs can accumulate directly from water into lower trophic level organisms and biomagnify with increasing trophic position due to dietary exposure (Thomann, 1989). This is particularly evident in the Arctic marine environment where the biomagnification of OCs is influenced by the efficient transfer of lipids within food webs (Falk-Petersen et al., 1990), as well as the nutritional demands of higher trophic level mammalian predators present in this system (Fisk et al., 2001).
The accumulation, biotransformation and excretion of OCs in lower trophic level biota influences the concentration of contaminants in higher trophic level species (Fisk et al., 2001; Moisey et al., 2001). Therefore, determining the trophic transfer of OCs is necessary to better address exposure and risk to human and wildlife populations.

Stable nitrogen (N) isotopes have been used to assess the relative trophic level of aquatic biota and provide a quantitative, continuous variable for studying the biomagnification of OCs within complex food webs (Fisk et al., 2001; Hop et al., 2002; Ruus et al., 2002). The use of stable N isotopes to assess trophic position has numerous advantages over traditional methods such as analysis of gut contents as it averages dietary assimilation over a longer period of time (DeNiro and Epstein, 1981); however, it does not allow for specific identification of prey and relative proportion of ingestion. In contrast, the ratios of stable carbon isotopes \( \frac{^{13}C}{^{12}C} \) in biota can help to elucidate trophic interactions by establishing the relative contribution of marine (or pelagic) versus coastal (or benthic) carbon sources (France and Peters, 1997). As a result, the quantification of both stable C and N isotope ratios can provide valuable information into the feeding ecology of biota and its potential influence on the trophic enrichment of OCs (Fisk et al., 2001; Hobson et al., 2002).

Studies investigating OCs in the Alaskan Arctic marine environment have focused on low trophic level biota (Hoekstra et al., 2002c), top predators (Krahn et al., 2000; Kucklick et al., 2002), and species of local significance (Hoekstra et al., 2002b; O’Hara et al., 1999). However, the biomagnification of OCs and the influence of trophic position on contaminant profiles within a quantified marine food web have not been addressed this region. The objectives of this study were to evaluate the basic structure of the marine food web (excluding seabirds) in the southern Beaufort–Chukchi Seas region and quantify OC accumulation in various species of local interest (e.g. traditional subsistence dietary items). Results were compared to other Arctic marine studies (Fisk et al., 2001; Hop et al., 2002; Muir et al., 2003) to assess whether the spatial trends of OC concentrations observed in top marine predators, such as ringed seals (Muir et al., 2000), were statistically significant and due to differences in trophic status or regional contamination.

2. Methodology

2.1. Field sampling

Samples were collected from 1999 to 2000 at Barrow, AK \( (71^\circ 17'\ N, 156^\circ 45'\ W) \) and Pt. Lay, AK \( (69^\circ 43'\ N, 163^\circ 00'\ W) \) through the North Slope Borough Department of Wildlife Management (DWM) and the Alaska Department of Fish and Game (ADFG; Fig. 1). Zooplankton samples \( (Calanus\ spp.) \) were collected using 100 µm plankton tow \( (1\ m^2\ opening) \) and stored in pre-cleaned glass jars (Hoekstra et al., 2002c). Arctic cod \( (Boreogadus\ saida) \), arctic char \( (Salvelinus\ alpinus) \), pink salmon \( (Oncorhynchus\ gorbuscha) \), and fourhorn sculpin \( (Myoxocephalus\ quadricornis) \) samples were obtained from Native (Inuit) subsistence fishers at Elson Lagoon (Barrow, AK).

Muscle and blubber samples from ringed seals \( (Phoca\ hispida) \) and bearded seals \( (Erignathus\ barbatus) \) were collected from Inuit subsistence harvests. Bowhead \( (Balaena\ mysticetus) \) and beluga whale \( (Delphinapterus\ leucas) \) blubber and muscle samples were obtained from Inuit subsistence whalers with the permission of the Alaska Eskimo Whaling Commission. Field sampling techniques of mammalian tissues have been previously described (Hoekstra et al., 2002a) and samples were transported to the National Water Research Institute (Environment Canada, Burlington, ON, Canada) and stored at \(-20^\circ\ C\). Tissues were homogenized prior to stable isotope analysis and OC extraction. Ages of selected specimens were determined from arctic cod otoliths (Gjosaeter and Ajiad, 1994) and bearded seal, ringed seal, and beluga whale teeth rings (Stewart et al., 1996; Hohn et al., 1989). Age estimates for other biota were not quantified due to the lack of available otoliths or tissues.

2.2. OC extraction and analysis

Biota samples were extracted and analyzed using previously described techniques with only minor modification (Hoekstra et al., 2002b,c). In brief, zooplankton samples were pooled for OC extraction and stable isotope analysis. Whole fish and full thickness blubber cores from marine mammals were homogenized prior to OC extraction. Tissue subsamples were mixed with sodium sulfate and spiked with two polychlorinated biphenyl (PCB) internal standards (CB-30 and CB-204) to monitor analyte recovery. Fish and zooplankton
samples were extracted with dichloromethane (DCM) via Soxhlet and concentrated. Blubber samples were extracted with DCM by using a Polytron® homogeniser (Brinkmann, Westbury, NY). Lipids and other bioorganic materials in each sample were removed using gel permeation chromatography and the lipid percent was determined gravimetrically.

The analytes for each sample were concentrated and separated on 100%-activated silica gel into two fractions: hexane (Fraction 1) and hexane: DCM (Fraction 2). 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. The hexachlorobiphenyl CB-166 was added to volume correct (iso-octane) and concentrated to 1000 μL. The hexachlorobiphenyl CB-166 was added to volume correct (iso-octane) and concentrated to 1000 μL. The hexachlorobiphenyl CB-166 was added to volume correct (iso-octane) and concentrated to 1000 μL.

2.3. Analytical quality assurance of OC analysis

Recovery of PCB surrogate standards was considered acceptable if greater than 70% for both CB-30 and CB-204 and concentrations were adjusted accordingly. Sample quantification was performed using multiple external standards provided by the National Laboratory for Environmental Testing (Environment Canada, Burlington, ON, Canada) that were analyzed after every 10 samples. Quality assurance protocol 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 (NIST; Gaithersburg, MD, USA) and participation in international interlaboratory comparison program on PCB analysis (QUASIMEME, Aberdeen, UK).

3. Stable isotope analysis and trophic level calculations

The protocol for stable C and N isotope analysis has been described elsewhere (Hoekstra et al., 2002a). In brief, samples of whole zooplankton and muscle tissue of various fish and marine mammals were analyzed in triplicate for both stable 15N/14N and 13C/12C isotope ratios using a Micromass Optima continuous-flow isotope-ratio mass-selective detector directly coupled to a Carlo Erba NA1500 elemental analyzer (Carlo Erba, Milan, Italy). Isotope enrichment was reported using the following notation:

\[ \delta R_{\%} = \left( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 1000 \]  

where the differential notation (\( \delta R \)) represents the relative difference between isotope ratios of the sample and standard gases \((^{13}C/^{12}C\) and \(^{15}N/^{14}N\)). The standard deviation for C and N isotope values in samples was ±0.2‰.

It was assumed that the Calanus copepods collected in this study were primary herbivores and occupied a secondary trophic position \((TL = 2.0)\). Isotopically derived trophic level for all other specimens was determined using the relationship developed by (Fisk et al., 2001):

\[ TL = 2.0 + \left( \frac{\delta^{15}N_{\text{consumer}} - \delta^{15}N_{\text{Calanus}}}{3.8} \right) \]  

where TL is the trophic level, \(\delta^{15}N_{\text{consumer}}\) is the \(\delta^{15}N\) signature in the specified organism, \(\delta^{15}N_{\text{Calanus}}\) is the mean \((±1\ SE)\) \(\delta^{15}N\) value for Calanus spp. \((9.8 ± 0.2)\) and 3.8 is the trophic enrichment factor for \(\delta^{15}N\) in an Arctic marine food web (Hobson et al., 2002).

3.1. Data analysis and OC trophic transfer calculations

The concentrations of several sum group (Σ) OCs are presented in Table 1. As wet weight OC concentrations were significantly correlated with lipid content, statistical comparisons of concentrations and relative distribution among species were performed using lipid-normalized values to reduce the effect of inter-species variation of lipid (Hebert and Keenleyside, 1995). Each statistical analysis was performed using Systat®, version 8.0 (SPSS, Chicago, IL, USA) with a maximum probability of a making a type-I error (α) established at 0.05.

Lipid-normalized OC concentrations were log10- transformed to reduce the skewness and kurtosis of the raw data prior to statistical comparisons. Interspecies comparisons of OC concentrations and relative proportions were performed using Tukey’s honestly significant difference (HSD) test. The biomagnification of OCs through the entire food web was determined by calculating a food web magnification factor (FWMF) based on the relationship between \(\delta^{15}N\)-determined trophic level and log10-transformed, lipid-normalized OC concentrations (Fisk et al., 2001). It should be noted, however, the OC data from bearded seals were omitted in this analysis as the \(\delta^{13}C\) signature of these specimens were significantly heavier than other biota, indicating a benthos-oriented feeding strategy relative to the other specimens collected.

The FWMF values for selected OCs generated in this study were compared with other published reports from the Arctic (Borgå et al., 2001; Fisk et al., 2001; Hop et al., 2002; Kleivane et al., 2000; Muir et al., 2003) to test the hypothesis that the trophic transfer of OCs are consistent among Arctic food webs containing similar species. The influence of study location, trophic level (TL), and the first-order interaction on log10-transformed OC concentrations (lipid adjusted) was examined using a general linear model (GLM). The first-order interaction (location×TL), was not significant according to Type III Sums-of Squares and therefore, the GLM was reduced to the following equation:
Table 1
Mean (± 1 S.E.) biological data (range in parentheses) and concentrations (ng g⁻¹; lipid wt.) of sum (Σ) OC groups in zooplankton, whole fish and marine mammal blubber from the Beaufort–Chukchi Seas (1999–2000)

<table>
<thead>
<tr>
<th>Species</th>
<th>n</th>
<th>TL</th>
<th>Age (year)</th>
<th>Body length (cm)</th>
<th>Lipid %</th>
<th>ΣClBe</th>
<th>ΣCHLor</th>
<th>ΣDDT</th>
<th>ΣPCBb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zooplankton</td>
<td>5</td>
<td>2.0±0.1</td>
<td>–</td>
<td>–</td>
<td>27±0.9</td>
<td>11±2.4</td>
<td>19±3.6</td>
<td>2.9±0.7</td>
<td>4.3±1.7</td>
</tr>
<tr>
<td>Pink salmon</td>
<td>7</td>
<td>2.3±0.1</td>
<td>–</td>
<td>–</td>
<td>6.3±0.1</td>
<td>23±3.1</td>
<td>22±3.5</td>
<td>21±1.6</td>
<td>29±1.6</td>
</tr>
<tr>
<td>Arctic char</td>
<td>3</td>
<td>3.1±0.1</td>
<td>–</td>
<td>–</td>
<td>4.5±0.8</td>
<td>32±5.2</td>
<td>40±8.7</td>
<td>30±11</td>
<td>36±3.8</td>
</tr>
<tr>
<td>Arctic cod</td>
<td>12</td>
<td>3.3±0.1</td>
<td>(1–1–2)</td>
<td>12 (10–19)</td>
<td>3.7±0.4</td>
<td>54±11</td>
<td>40±3.2</td>
<td>76±11</td>
<td>70±14</td>
</tr>
<tr>
<td>Fourhorn sculpin</td>
<td>7</td>
<td>3.5±0.2</td>
<td>–</td>
<td>15 (13–17)</td>
<td>4.4±0.5</td>
<td>36±14</td>
<td>28±6.4</td>
<td>49±15</td>
<td>48±17</td>
</tr>
<tr>
<td>Bearded seal</td>
<td>7</td>
<td>3.8±0.1</td>
<td>7.2 (1–20)</td>
<td>212 (153–251)</td>
<td>80±3.9</td>
<td>57±9.5</td>
<td>65±11</td>
<td>236±19</td>
<td>180±21</td>
</tr>
<tr>
<td>Ringed seal</td>
<td>20</td>
<td>4.1±0.2</td>
<td>6.2 (1–20)</td>
<td>108 (64–151)</td>
<td>84±3.6</td>
<td>48±7.8</td>
<td>190±50</td>
<td>487±85</td>
<td>326±37</td>
</tr>
<tr>
<td>Bowhead whale</td>
<td>20</td>
<td>3.8±0.1</td>
<td>–</td>
<td>1090 (775–1540)</td>
<td>70±2.6</td>
<td>196±20</td>
<td>282±25</td>
<td>260±28</td>
<td>437±40</td>
</tr>
<tr>
<td>Beluga whale</td>
<td>25</td>
<td>3.8±0.2</td>
<td>23 (9–40)</td>
<td>380 (304–435)</td>
<td>85±1.8</td>
<td>330±30</td>
<td>224±44</td>
<td>1320±134</td>
<td>1979±231</td>
</tr>
</tbody>
</table>

n = sample size; TL = trophic level [Eq. (2)].

log₁₀[OC] = μ + location + TL + ε  

where ε is the error vector and μ is a constant. An a-posteriori Scheffé’s method was selected to assess the relative biomagnification of selected OC analytes among locations. Separate FWMF were not determined for the limited number of species available.

A second method was employed to investigate the biomagnification factors (BMFs) of OCs between species:

BMF = [(OCp_predator/OCp_prey)]/[(TLp_predator/TLP_rey)]  

where OCp_predator and OCp_prey represent the lipid normalized concentrations of a specific OC or ΣOC group in the predator and prey species, respectively. The BMF values were adjusted for trophic level based on δ¹³C values for the predator and prey species (TLp_predator and TLP_rey), respectively (Fisk et al., 2001). Those OCs with FWMF and BMF values found to be statistically greater than unity (via the Z-test) were considered to accumulate from prey to predator (for BMFs) and within the selected food web (for FWMF). Arctic cod was used as a model prey species for quantifying the bioaccumulation of OCs by ringing seals from fish (Bradstreet and Cross, 1982). The BMFs for beluga whales and bearded seals were determined using mean OC concentrations in all fish as the relative dietary contribution of individual species were not available.

Relationships between FWMFs, BMFs for homeotherms and poikilotherms and octanol–water partition coefficients (Kow) for major OC analytes were investigated using Model-I, first-order linear regression. Coefficient values were derived from Hawker and Connell (1988) and Mackay et al. (1992).

4. Results

4.1. Trophic level status

Stable δ¹³C isotope signatures ranged from mean values of −21.2 to −17.7‰ in the biota sampled (Fig. 2) from the near shore marine food web of the southern Beaufort–Chukchi Seas (Hoeckstra et al., 2002a). Bearded seals had the greatest ¹³C enrichment of any homeotherm (mean δ¹³C value of −17.5‰), which is indicative of a link to the benthic food web. There was a wide range of δ¹³C values in the Beaufort–Chukchi Seas biota, with mean values ranging from 9.8 to 16.9‰ (Hoeckstra et al., 2002a) and followed a general enrichment of 3.8‰ with each trophic level (Table 1; Fig. 2). As a strict herbivore, calanoid copepods were the most ¹⁴N depleted.

4.2. OC concentrations and biomagnification in Beaufort–Chukchi Seas biota

The lipid-adjusted, mean ΣOC concentrations in calanoid copepods, Calanus spp., ranged from 4.3 to 46
The mean concentrations of ΣOC classes quantified in all fish species ranged from 21 to 76 ng g⁻¹ (lipid weight). The lipid-normalized concentrations in fishes were significantly greater than in zooplankton (ANOVA, \( P < 0.05 \) for all comparisons). The lipid-normalized concentrations of ΣOCs increased from fish to marine mammal blubber (ANOVA; \( P < 0.05 \)) for all ΣOC-species comparisons. In marine mammals, lipid-normalized ΣPCB concentrations in blubber exceed those of other quantified ΣOC groups (Tukey’s HSD, \( P < 0.05 \) for all comparisons).

The lipid-adjusted concentrations of ΣPCB, ΣDDT, ΣCHLOR, and ΣClBz in beluga whale blubber were significantly greater than in other marine mammals. All ΣOC concentrations determined in ringed seal blubber were significantly greater than in bearded seals. Lipid-adjusted ΣOC concentrations for all classes, except ΣHCH, were significantly lower in bowhead whales relative to beluga whales (ANOVA, Tukey’s HSD, \( P < 0.05 \)). No statistically significant difference in lipid-normalized ΣHCH blubber concentrations between bowhead and beluga whales was observed (Tukey’s HSD, \( P > 0.05 \)). Overall, the most abundant classes of OC compounds in cetaceans were ΣPCB and ΣDDT, whereas ΣPCB and ΣCHLOR were the dominant ΣOC groups in pinnipeds.

Fig. 3 illustrates the biomagnification of various OCs with trophic level in biota from the southern Beaufort–Chukchi Seas. The FWMFs values calculated in this study ranged from 0.65 to 9.30 (Table 2). Significant correlations between \( \log_{10} K_{ow} \) and FWMF [FWMF = 1.33(\( \log_{10} K_{ow} \)) − 3.95; \( P = 0.02 \); \( r^2 = 0.34 \)] for all OCs (excluding metabolites) and for PCBs alone [FWMF = 3.21(\( \log_{10} K_{ow} \)) − 16.0; \( P < 0.001 \); \( r^2 = 0.68 \)] were observed. Similar relationships were also observed for pooled BMF values of all fish (\( P < 0.03 \); \( r^2 > 0.35 \)). However, no significant correlation between \( \log_{10} K_{ow} \) and BMFs for pinnipeds (\( P > 0.20 \)) and cetaceans (\( P > 0.30 \)) were observed.

In general, FWMF values from the near-shore, southern Beaufort–Chukchi Seas food web were statistically comparable with values determined from similar species collected in the North Baffin Bay (Fisk et al., 2001). However, the FWMFs from both locations were, overall, significantly different (Scheffé’s test, \( P < 0.01 \)) from FWMFs determined from the White Sea (Muir et al., 2003), and the Barents Sea (Hop et al., 2002; Kleivane et al., 2000; Table 3).

5. Discussion

5.1. Trophic level determination

The species investigated were selected components of the pelagic and benthic marine food web of the southern Beaufort–Chukchi Seas near Barrow, AK. While
Fig. 3. The relationship between log$_{10}$-transformed, lipid-normalized concentration for selected OCs (ng g$^{-1}$) and trophic level (TL). Mean (±1 S.E.) values for each species are provided and are represented by the following symbols: zooplankton (●), fish (■), and marine mammals (▲). Statistically significant ($P < 0.05$) correlations are illustrated by the Model-I, first-order linear regression relationship. Food web magnification factors (FWMF; Table 2) are determined by $10^b$, where $b$ is the slope between log$_{10}$-transformed OC concentrations (lipid weight) and TL. Note: bearded seals and fourhorn sculpins were not included in this regression due to enriched $\delta^{13}$C values relative to other biota.
the application of a single trophic enrichment factor for $\delta^{15}$N throughout the food web is an undoubted simplification of an inherently complex system, the general placement of Calanus spp. at the second TL, Arctic cod at TL 3.3, and ringed seals at TL 4.1, is consistent with documented feeding strategies (Bradstreet and Cross, 1982). As a result, the $\delta^{15}$N and $\delta^{13}$C profiles varied among the species analyzed (Fig. 2) and were in reasonable agreement with current, but limited, knowledge concerning the diet of Arctic marine biota (Hobson et al., 1995).

It should be noted that the dependence of this study on seasonally harvested biota (especially marine mammals which were mostly collected outside the winter months) might bias these results and not reflect the overall trophic structure throughout the remainder of the year. Additionally, the $\delta^{13}$C profiles in the Beaufort-Chukchi Seas are not homogeneous. Calanoid copepods and other zooplankton from the Beaufort Sea are more $^{13}$C-depleted relative to the Chukchi Sea (Schell et al., 1998), thereby affecting the interpretation of $\delta^{13}$C values quantified in marine mammals actively feeding within both regions.

It was assumed that the zooplankton (Calanus spp.) samples collected during this study represented the TL 2.0, as this genus is known to graze on primary producers. Fish species, with the exception of pink salmon, generally represented the third trophic level. Arctic char (TL 3.1) and Arctic cod (TL 3.3) feed on zooplankton and are regarded as primary prey for various marine mammals (Bradstreet and Cross, 1982). Fourhorn sculpin (TL 3.5) prey on various fish and benthic invertebrates (Atkinson and Percy, 1992), and as a result have a more $^{13}$C enriched (benthic) signature relative to other

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>FWMF&lt;sup&gt;a&lt;/sup&gt; ($r^2$)</th>
<th>BOW/Calanus</th>
<th>Cod/Calanus</th>
<th>RS/cod</th>
<th>BS/fish</th>
<th>BLGA/fish</th>
</tr>
</thead>
<tbody>
<tr>
<td>TriClBz</td>
<td>1.91 (0.41)</td>
<td>0.3</td>
<td>0.5</td>
<td>3.4</td>
<td>2.2</td>
<td>2.7</td>
</tr>
<tr>
<td>PentaClBz</td>
<td>2.93 (0.52)</td>
<td>0.7</td>
<td>3.1</td>
<td>2.1</td>
<td>0.6</td>
<td>4.3</td>
</tr>
<tr>
<td>HCB</td>
<td>1.36 (0.03)</td>
<td>8.7</td>
<td>2.6</td>
<td>0.3</td>
<td>0.2</td>
<td>5.5</td>
</tr>
<tr>
<td>ΣClBz</td>
<td>1.53 (0.12)</td>
<td>5.9</td>
<td>2.5</td>
<td>0.6</td>
<td>0.5</td>
<td>4.3</td>
</tr>
<tr>
<td>$\alpha$-HCH</td>
<td>1.58 (0.19)</td>
<td>3.7</td>
<td>0.9</td>
<td>2.2</td>
<td>0.9</td>
<td>1.7</td>
</tr>
<tr>
<td>$\beta$-HCH</td>
<td>2.89 (0.51)</td>
<td>15.6</td>
<td>1.6</td>
<td>8.9</td>
<td>2.7</td>
<td>17.8</td>
</tr>
<tr>
<td>$\gamma$-HCH</td>
<td>0.65 (0.08)</td>
<td>4.6</td>
<td>0.7</td>
<td>0.9</td>
<td>0.2</td>
<td>3.2</td>
</tr>
<tr>
<td>ΣHCH</td>
<td>3.22 (0.54)</td>
<td>4.3</td>
<td>1.8</td>
<td>2.8</td>
<td>0.9</td>
<td>3.7</td>
</tr>
<tr>
<td>cis-Chlordane</td>
<td>0.72 (0.54)</td>
<td>3.0</td>
<td>4.6</td>
<td>0.9</td>
<td>0.7</td>
<td>5.3</td>
</tr>
<tr>
<td>trans-Chlordane</td>
<td>0.98 (0.09)</td>
<td>3.6</td>
<td>4.6</td>
<td>0.6</td>
<td>0.2</td>
<td>0.6</td>
</tr>
<tr>
<td>cis-Nonachlor</td>
<td>2.65 (0.26)</td>
<td>7.0</td>
<td>2.4</td>
<td>1.9</td>
<td>0.3</td>
<td>19.5</td>
</tr>
<tr>
<td>trans-Nonachlor</td>
<td>5.21 (0.63)</td>
<td>9.5</td>
<td>2.5</td>
<td>5.7</td>
<td>4.8</td>
<td>12.3</td>
</tr>
<tr>
<td>Oxychlor dane</td>
<td>9.30 (0.67)</td>
<td>11.9</td>
<td>1.5</td>
<td>7.5</td>
<td>3.4</td>
<td>25.6</td>
</tr>
<tr>
<td>HPEX</td>
<td>7.71 (0.57)</td>
<td>31.4</td>
<td>1.7</td>
<td>10.2</td>
<td>4.4</td>
<td>22.1</td>
</tr>
<tr>
<td>ΣCHLOR</td>
<td>4.78 (0.58)</td>
<td>12.8</td>
<td>2.6</td>
<td>4.6</td>
<td>2.2</td>
<td>13.5</td>
</tr>
<tr>
<td>$\delta$-DDE</td>
<td>5.35 (0.48)</td>
<td>23.1</td>
<td>2.5</td>
<td>4.8</td>
<td>6.2</td>
<td>46.5</td>
</tr>
<tr>
<td>$\delta$-DDT</td>
<td>1.53 (0.05)</td>
<td>27.6</td>
<td>0.6</td>
<td>2.4</td>
<td>1.3</td>
<td>20.7</td>
</tr>
<tr>
<td>ΣDDT</td>
<td>2.95 (0.31)</td>
<td>14.3</td>
<td>2.2</td>
<td>2.4</td>
<td>2.1</td>
<td>23.0</td>
</tr>
<tr>
<td>CB-28</td>
<td>1.29 (0.10)</td>
<td>4.2</td>
<td>1.4</td>
<td>1.1</td>
<td>0.3</td>
<td>1.5</td>
</tr>
<tr>
<td>CB-52</td>
<td>2.62 (0.34)</td>
<td>6.7</td>
<td>1.1</td>
<td>3.3</td>
<td>1.2</td>
<td>4.3</td>
</tr>
<tr>
<td>CB-95/66</td>
<td>3.75 (0.41)</td>
<td>19.1</td>
<td>2.3</td>
<td>1.4</td>
<td>0.9</td>
<td>10.8</td>
</tr>
<tr>
<td>CB-99</td>
<td>5.94 (0.57)</td>
<td>17.6</td>
<td>4.5</td>
<td>20.5</td>
<td>10.1</td>
<td>25.8</td>
</tr>
<tr>
<td>CB-101</td>
<td>3.89 (0.44)</td>
<td>10.6</td>
<td>1.7</td>
<td>8.1</td>
<td>4.0</td>
<td>13.9</td>
</tr>
<tr>
<td>CB-105</td>
<td>5.79 (0.66)</td>
<td>8.5</td>
<td>2.5</td>
<td>1.1</td>
<td>0.7</td>
<td>5.7</td>
</tr>
<tr>
<td>CB-118</td>
<td>3.77 (0.44)</td>
<td>7.1</td>
<td>2.9</td>
<td>3.3</td>
<td>2.3</td>
<td>11.6</td>
</tr>
<tr>
<td>CB-138</td>
<td>4.72 (0.61)</td>
<td>18.2</td>
<td>1.9</td>
<td>9.4</td>
<td>6.2</td>
<td>11.6</td>
</tr>
<tr>
<td>CB-153</td>
<td>6.69 (0.64)</td>
<td>14.7</td>
<td>2.8</td>
<td>10.4</td>
<td>8.2</td>
<td>20.2</td>
</tr>
<tr>
<td>CB-180</td>
<td>6.52 (0.60)</td>
<td>8.6</td>
<td>2.2</td>
<td>6.8</td>
<td>4.5</td>
<td>16.4</td>
</tr>
<tr>
<td>ΣPCB</td>
<td>3.26 (0.47)</td>
<td>10.9</td>
<td>1.5</td>
<td>6.8</td>
<td>4.0</td>
<td>24.4</td>
</tr>
</tbody>
</table>

<sup>a</sup> FWMF calculated by the equation: FWMF = $10^b$, where $b$ was the slope of the OC concentration—TL relationship; the correlation coefficient ($r^2$) is provided in parentheses. OC concentrations—TL relationships that are not statistically significant ($P \geq 0.05$) are annotated by italicized FWMF values.

<sup>b</sup> BMF for predator–prey comparison were corrected for trophic level [Eq. (4)].
Table 3
Food web magnification factors (FWMFs, ±1 S.E.) for selected OCs in the southern Beaufort–Chukchi Seas food web compared with other Arctic marine locations

<table>
<thead>
<tr>
<th>Region analyte</th>
<th>Beaufort–Chukchi Sea (BC)</th>
<th>North Baffin Bay (NB)</th>
<th>Barents Sea (BA)</th>
<th>White Sea (WH)</th>
<th>Site difference (P &lt; 0.05)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HCB</td>
<td>1.36 ± 0.17</td>
<td>1.75 ± 0.11</td>
<td>1.55 ± 0.10</td>
<td>2.27 ± 0.37</td>
<td>No sig. difference</td>
</tr>
<tr>
<td>α-HCH</td>
<td>1.58 ± 0.25</td>
<td>2.19 ± 0.15</td>
<td>1.50 ± 0.14</td>
<td>1.91 ± 0.23</td>
<td>No sig. difference</td>
</tr>
<tr>
<td>β-HCH</td>
<td>2.89 ± 0.30</td>
<td>4.21 ± 0.55</td>
<td>1.51 ± 0.36</td>
<td>0.61 ± 0.16</td>
<td>BC ≠ BA, WH; NB ≠ BA, WH</td>
</tr>
<tr>
<td>γ-HCH</td>
<td>0.65 ± 0.07</td>
<td>1.39 ± 0.54</td>
<td>1.68 ± 0.07</td>
<td>0.62 ± 0.12</td>
<td>No sig. difference</td>
</tr>
<tr>
<td>cis-Chlordane</td>
<td>0.72 ± 0.07</td>
<td>1.72 ± 0.54</td>
<td>NQ</td>
<td>0.66 ± 0.20</td>
<td>No sig. difference</td>
</tr>
<tr>
<td>trans–Nonachlor</td>
<td>5.21 ± 0.09</td>
<td>5.13 ± 0.37</td>
<td>10.4 ± 1.41</td>
<td>4.83 ± 1.01</td>
<td>BA ≠ BC, NB, WH</td>
</tr>
<tr>
<td>p,p′-DDE</td>
<td>5.35 ± 0.15</td>
<td>7.39 ± 0.10</td>
<td>14.5 ± 1.44</td>
<td>3.62 ± 0.66</td>
<td>WH ≠ BC, NB, BA; BC ≠ BA</td>
</tr>
<tr>
<td>p,p′-DDT</td>
<td>1.53 ± 0.10</td>
<td>2.78 ± 0.15</td>
<td>NQ</td>
<td>1.31 ± 0.45</td>
<td>WH ≠ BC, NB</td>
</tr>
<tr>
<td>CB-28</td>
<td>1.29 ± 0.06</td>
<td>1.28 ± 0.29</td>
<td>3.31 ± 0.06</td>
<td>1.31 ± 0.30</td>
<td>BA ≠ BC, NB, WH</td>
</tr>
<tr>
<td>CB-99</td>
<td>5.94 ± 0.14</td>
<td>5.52 ± 0.17</td>
<td>11.9 ± 1.37</td>
<td>2.61 ± 0.85</td>
<td>BC ≠ BA, NB, WH</td>
</tr>
<tr>
<td>CB-138</td>
<td>4.72 ± 0.18</td>
<td>5.57 ± 0.31</td>
<td>17.0 ± 1.42</td>
<td>3.44 ± 0.66</td>
<td>BA ≠ BC, NB, WH</td>
</tr>
<tr>
<td>CB-153</td>
<td>6.69 ± 0.14</td>
<td>5.87 ± 0.11</td>
<td>18.8 ± 1.44</td>
<td>2.93 ± 0.59</td>
<td>BA ≠ BC, NB, WH; WH ≠ BC, NB</td>
</tr>
<tr>
<td>ΣPCB</td>
<td>3.26 ± 0.12</td>
<td>2.90 ± 0.09</td>
<td>NQ</td>
<td>1.59 ± 0.22</td>
<td>WH ≠ BC, NB</td>
</tr>
</tbody>
</table>

Not statistically different (Scheffe’s test, \( P > 0.05 \) for all comparisons); NQ = not quantified due to limited available data.

a This study (Table 2).

b From Fisk et al. (2001), excluding avian data.

c Derived from Borga et al. (2001), Hop et al. (2002), and Kleivane et al. (2000), excluding avian data and benthic-oriented species [sculpin and spider crab \( (H. araneus) \)].

d From Muir et al. (2002),

Fish species. This species had slightly higher \( ^{15}N \) signatures compared to other fishes due to predation on other fish species and (or) feeding on benthos, which are typically more \( ^{15}N \) enriched relative to pelagic biota (Hobson et al., 2002), and therefore was excluded from FWMF calculations (see below). The stable isotope profile of pink salmon (TL 2.3) indicates that these specimens were feeding on pelagic amphipods or primary producers, which is consistent with the dietary profile of this species (Healey, 1991).

The marine mammals sampled, with the exception of the bowhead whale, occupied the highest trophic levels. The isotope profile of the bowhead whale is consistent with known feeding behaviour as this species feeds mainly on plankton (Lowry, 1993), and subsequently occupies a lower-trophic level position (TL 2.9) relative to other marine mammals (TL ≈ 4) from this study area (Hoekstra et al., 2002a). Beluga whales occupy a similar trophic level (TL 3.8) as ringed seals. The diet of this odontocete is assumed to consist mainly of fish, including arctic cod, as well as a variety of benthic and pelagic invertebrates (Heide-Jorgensen and Teilmann, 1994), which is consistent with the TL calculated herein.

Arctic cod are believed to be the main prey item for ringed seals (TL 4.1) during the late autumn and early spring, whereas pelagic and benthic invertebrates become increasingly important food sources during the summer (Bradstreet and Cross, 1982; Weslawski et al., 1994). The isotopic data generated reflects the overall integration of arctic cod and benthos into the diet of ringed seals. However, the collection of ringed seals in summer may underestimate the contribution of arctic cod to the isotopically determined TL of this species.

The diet of bearded seals (TL 3.8) consists mainly of pelagic and epibenthic fish, as well as a variety of benthic invertebrates (Antonelis et al., 1994; Finley and Evans, 1983). While the foraging behaviour of bearded seals changes seasonally with prey availability and habitat conditions, the benthic feeding strategy of this species is consistent with the quantified \( ^{13}C \) signature (Fig. 2).

5.2. Organochlorine concentrations

The \( \Sigma OC \) concentrations in marine biota from the southern Beaufort–Chukchi Seas food web increased from zooplankton, to fish, to the various marine mammals analyzed in this study and are similar to those reported in other studies from the Alaskan and western Canadian Arctic. The concentration of PCBs and other chlorinated pesticides in zooplankton collected near Barrow, AK were previously reported as part of a larger comparison within the North American Arctic (Hoekstra et al., 2002c) and are consistent with published reports on OCs in Arctic marine zooplankton from this region (Bidleman et al., 1989; Hargrave et al., 2000). The increased OC concentration and modification of analyte profiles in fish relative to zooplankton is reflective of the feeding ecology and subsequent dietary exposure as well as contaminant bioaccumulation and possible biotransformation (Bidleman et al., 1993; Borga et al., 2001; Fisk et al., 2001; Ruus et al., 1999).

The OC concentrations in the two pinniped species, ringed seals and bearded seals, were comparable to other investigations from arctic Alaska (Krahn et al., 1997; Kucklick et al., 2002). In these studies, the mean
lipid-normalized \( \Sigma \text{PCB} \) concentrations (±1 SE) in bearded and ringed seal blubber were 200±52 and 900±110 ng g\(^{-1} \), respectively. However, concentrations of \( \Sigma \text{PCB} \), \( \Sigma \text{CHLOR} \), and \( \Sigma \text{DDT} \) were significantly lower than those reported from the eastern Canadian and European Arctic (Bang et al., 2001; Fisk et al., 2002; Kleivane et al., 2000), and in agreement with the circumpolar trends of OCs observed in higher-trophic level biota, such as ringed seals (Muir et al., 2000), and seabirds (de March et al., 1998).

The magnitude of OC accumulation in bowhead whales is relatively low compared to other cetaceans (Aono et al., 1997; Weisbrod et al., 2000). This reflects the lower dietary exposure to OCs to this bowhead whale stock (TL 2.9) due to feeding on lower trophic level invertebrates and primary producers (Hoekstra et al., 2002a) as well as the remoteness of this habitat from major areas of industrialization. In contrast, beluga whales had the highest OC concentrations (except \( \Sigma \text{HCH} \)) of any marine mammal analyzed in this study, likely due to the age of the animals sampled (Table 1), dietary exposure from higher trophic level prey, and other factors influencing the bioaccumulation of lipophilic contaminants. Our results are similar to those reported in Alaskan Arctic beluga populations (Krahm et al., 2000) but significantly lower than concentrations in belugas from the eastern Canadian Arctic (Muir et al., 1999).

5.3. Interspecies differences in OC accumulation

Fig. 3, for example, illustrates the biomagnification of structurally diverse OCs in marine biota from the southern Beaufort–Chukchi Seas coastal food web. Of the chlorinated pesticide-related compounds, FWMF values ranged from 0.65 for \( \gamma\text{-HCH} \) to 9.3 for \( \text{OXY} \) and from 1.3 to 6.7 for the most abundant PCB congeners. It has been suggested that FWMF determined by a simple, first-order linear model overestimate OC transfer for poikilotherms and underestimate values for homeotherms (Fisk et al., 2001; Hop et al., 2002; Ruus et al., 2002). Hop et al. (2002) showed that FWMF are generally higher in homeotherms relative to poikilotherms due to higher energy requirements, greater biomass of lipid reservoirs, and duration and route of exposure. While the limited number of species available to this study prevents the creation of FWMF for both classes of biota, the BMF values reported herein are consistent with the previous reports in which OC accumulation were lower for fish and higher for marine mammals, compared to the overall FWMF for individual OC analytes.

Interspecies differences in accumulated OCs may be explained by the feeding strategy of an organism and its ability to biotransform OCs, which is dependent on the structure of the compound and the presence and (or) capacity of enzymatic systems to be induced and metabolize xenobiotics (Boon et al., 1992; Bucheli and Fent, 1995). As well, the different chemical structure and (or) degree of chlorination will affect the physiochemical properties of OCs, such as partition coefficients, and will therefore influence passive uptake into biota. Fisk et al. (2001) reported significant correlation between FWMFs and log\(_{10}\)\(K_{\text{ow}}\) of various OCs and suggested that variability may be attributed to metabolism of these compounds and (or) inaccuracy of reported \(K_{\text{ow}}\) values. A significant correlation between BMF-log\(_{10}\)\(K_{\text{ow}}\) was observed for fish \((P = 0.004; r^2 = 0.35)\), but not for marine mammals \((P = 0.30; \text{data not shown})\). The metabolism of OCs in poikilotherms appears to be a minor factor affecting accumulation compared to uptake and partitioning from the environment, whereas biotransformation clearly influences the bioaccumulation profiles of OCs in homeotherms (Table 2). However, this interpretation should be regarded with caution as the analysis of different tissues from fish (whole body) and marine mammals (blubber) in this study may confound these interspecies differences.

The FWMF for PCB congeners in the Beaufort–Chukchi Seas marine food web ranged from 1.29 for CB-28 to 6.69 for CB-153. Overall, the highest FWMFs were found for the hexa- and hepta-Cl substituted PCB homologues. The BMFs generated for fish ranged from 1.1 for CB-52 to 5.4 for CB-99, and were generally lower than BMFs for cetaceans (1.5–25.8) and pinnipeds (0.3–20.5), respectively (Table 2). The biotransformation of PCB congeners is typically mediated...
through the induction of cytochrome P-450 (CYP) iso-
zymes, including members of the CYP1A and 2B sub-
families and is dependent upon the Cl substitution patterns (Okey, 1990). Congeners with 2,4,5-substitution (CB-99, 138, 153, 180), which are typically recalci-
trant in most biota (Boon et al., 1994), had the highest FWMF compared to all other congeners. The relatively high biomagnification of these congeners in home-
otherms is similar to other marine food chain studies (Hop et al., 2002; Muir et al., 1988; Ruus et al., 1999; other marine food web studies (Fisk et al., 2001; Hop et al., 2002; Norstrom et al., 1988; Ruus et al., 1999). As p,p'-DDT may be formed and (or) accumulated by plankton and algae (Patil et al., 1972), the accumulation of ΣDDDE in fish may also be the result of dietary uptake and (or) DDT metabolism and subsequent DDE formation (Addison and Willis, 1978). The average BMF value for DDE in all fish (BMF = 3.5) is biased towards four-
horn sculpin (BMF = 4.9) compared to other teleosts (BMF = 2.7, 2.5, and 2.5 for arctic char, cod and salmon, respectively). The relative contribution of ΣDDDE to ΣDDT in sculpin is significantly different from BMFs for other fish (P < 0.001) but similar to those observed in marine mammals. Recent investigations have suggested that Myoxocephalus spp. possesses a unique ability to bio-
transform PCB congeners that are recalcitrant in other fish (Stapleton et al., 2001). In the Great Lakes, ΣDDDE was the most strongly biomagnified compound class in deepwater sculpin relative to other quantified OCs (Evans et al., 1991), suggesting that that sculpin (or their prey) have an increased capacity to dehydrochlorinate o,p'- and p,p'-DDT to their respective DDE metabolites compared to other fish collected in this study.

5.4. OC biomagnification in Arctic marine food webs

The concentrations of OCs in top marine predators, such as ringed seals and beluga whales, vary geo-
graphically within the Arctic (Muir et al., 1999, 2000).

Through the induction of cytochrome P-450 (CYP) iso-
zymes, including members of the CYP1A and 2B sub-
families and is dependent upon the Cl substitution patterns (Okey, 1990). Congeners with 2,4,5-substitution (CB-99, 138, 153, 180), which are typically recalci-
trant in most biota (Boon et al., 1994), had the highest FWMF compared to all other congeners. The relatively high biomagnification of these congeners in home-
otherms is similar to other marine food chain studies (Hop et al., 2002; Muir et al., 1988; Ruus et al., 1999; other marine food web studies (Fisk et al., 2001; Hop et al., 2002; Norstrom et al., 1988; Ruus et al., 1999). As p,p'-DDT may be formed and (or) accumulated by plankton and algae (Patil et al., 1972), the accumulation of ΣDDDE in fish may also be the result of dietary uptake and (or) DDT metabolism and subsequent DDE formation (Addison and Willis, 1978). The average BMF value for DDE in all fish (BMF = 3.5) is biased towards four-
horn sculpin (BMF = 4.9) compared to other teleosts (BMF = 2.7, 2.5, and 2.5 for arctic char, cod and salmon, respectively). The relative contribution of ΣDDDE to ΣDDT in sculpin is significantly different from BMFs for other fish (P < 0.001) but similar to those observed in marine mammals. Recent investigations have suggested that Myoxocephalus spp. possesses a unique ability to bio-
transform PCB congeners that are recalcitrant in other fish (Stapleton et al., 2001). In the Great Lakes, ΣDDDE was the most strongly biomagnified compound class in deepwater sculpin relative to other quantified OCs (Evans et al., 1991), suggesting that that sculpin (or their prey) have an increased capacity to dehydrochlorinate o,p'- and p,p'-DDT to their respective DDE metabolites compared to other fish collected in this study.
Isotopic analysis revealed a general pattern of δ¹⁵N enrichment with trophic status that was consistent with other marine investigations in Arctic and temperate locations. The mean δ¹⁵N ranges for *Calanus* spp. to ringed seals (9.8–16.9‰) in the Beaufort–Chukchi Seas food web was similar to the same portion of the marine food webs in the eastern Canadian Arctic (9–17.3‰; Jarman et al., 1996). However, direct comparison North Baffin Bay and this study (Table 3). Muir et al. greater and lower, respectively, than values from the Barents and White Seas (excluding avian data) were still significantly greater than FWMFs from the southern Beaufort–Chukchi Seas, North Baffin Bay and the White Sea (P > 0.25 for all comparisons). As the trophic status of the investigated species are similar among locations, the significantly greater FWMF and BMF (fish to pinnipeds) values generated for Barents Sea food web suggest that the top marine predators from this location feed on unidentified species containing higher concentrations relative to those quantified in the aforementioned studies.

### 6. Conclusions

The trophic status of marine biota from the southern Beaufort–Chukchi Seas region, as determined by δ¹³C and δ¹⁵N, is similar to other Arctic marine systems. OC concentrations were correlated with δ¹⁵N and generally increased from zooplankton to fish to marine mammals. As a result, this study further supports the application of stable isotopes to not only elucidate the trophic position of biota, but also to quantify the biomagnification of OCs within food webs.

The OCs with the greatest FWMFs were biotransformation products (e.g. p,p'-DDE, OXY) or those considered recalcitrant in most biota (e.g. β-HCH, *ortho-meta* or *meta-para* Cl substituted PCB congeners). The distribution of OCs in fish and marine mammals reflects the differences in contaminant biomagnification due to an organism’s feeding strategy, exposure, detoxification capacity, and the physical–chemical properties of the compound. The concentrations of OCs in fish were significantly correlated with log*K*<sub>ow</sub>, suggesting that physical partitioning of a chemical and the limited biotransformation capacity influences OC profiles in lower trophic level organisms. However, the ability to biotransform and excrete OCs is an important factor governing the accumulation profile in marine mammals compared to lower trophic level prey.

The OC concentrations, with the exception of HCH isomers, were relatively lower in biota from the southern Beaufort–Chukchi Seas region compared other
Arctic locations. As isotopically derived trophic position of similar marine species are consistent among Arctic locations, the geographical variability of OC concentrations in top marine predators, such as ringed seals and beluga whales, is likely attributed to differences in OC contaminant exposure rather than in gross differences in feeding ecology.

Acknowledgements

The Co-operative Institute for Arctic Research, located at the University of Alaska Fairbanks (UAF, Fairbanks, AK), provided financial support for this project. Data generation from the North Baffin Bay, Barents Sea, and White Sea was supported by the Natural Science and Engineering Research Council, the Norwegian Research Council, and Pew Fellow Program in Marine Conservation, respectively. The analyses of OCs in biota from the Barents Seas were performed by the Environmental Toxicology Laboratory (Norwegian School of Veterinary Science, Oslo, Norway). The authors are very grateful for the provision of samples by the Inuit hunters at Barrow and Pt. Lay, AK. Benny Akootckok, C.D.N. Brower, H. Brower Jr., L.A. Dehn, J.C. George, C. Rosa, R. Suydam, V. Woshner, G. Sheffield and many others provided additional samples. Beluga and seal age estimates were provided by R. Suydam (DWM) and L.A. Dehn (UAF). Age determination of arctic cod otoliths was performed by J. Cassetsman (Ontario Ministry of Natural Resources, Picton, ON, Canada). This project would not have been possible without the support of the Barrow Arctic Science Consortium and DWM, including T. Albert, D. Norton, L. Dela Rosa, A. Brower, D. Vinas, and C. Willetto.

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