Trophic Transfer of Contaminants in a Changing Arctic Marine Food Web: Cumberland Sound, Nunavut, Canada

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Supporting Information

ABSTRACT: Contaminant dynamics in arctic marine food webs may be impacted by current climate-induced food web changes including increases in transient/subarctic species. We quantified food web organochlorine transfer in the Cumberland Sound (Nunavut, Canada) arctic marine food web in the presence of transient species using species-specific biomagnification factors (BMFs), trophic magnification factors (TMFs), and a multifactor model that included δ¹⁵N-derived trophic position and species habitat range (transient versus resident), and also considered δ¹³C-derived carbon source, thermoregulatory group, and season. Transient/subarctic species relative to residents had higher prey-to-predator BMFs of biomagnifying contaminants (1.4 to 62 for harp seal, Greenland shark, and narwhal versus 1.1 to 20 for ringed seal, arctic skate, and beluga whale, respectively). For contaminants that biomagnified in a transient-and-resident food web and a resident-only food web scenario, TMFs were higher in the former (2.3 to 10.1) versus the latter (1.7 to 4.0). Transient/subarctic species have higher tissue contaminant levels and greater BMFs likely due to higher energetic requirements associated with long-distance movements or consumption of more contaminated prey in regions outside of Cumberland Sound. These results demonstrate that, in addition to climate change-related long-range transport/deposition/revolatilization changes, increasing numbers of transient/subarctic animals may alter food web contaminant dynamics.

INTRODUCTION

Environmental contamination by persistent and bioaccumulative industrial, agricultural, and consumer-product chemicals is an ongoing global concern, including in regions far from source inputs such as the Arctic. Persistent organic pollutants (POPs) enter arctic environments through atmospheric, aquatic, and biological transport and bioaccumulate in organisms, increasing in concentration relative to the surrounding environment. Bioaccumulation can increase with each trophic step in the food web, biomagnifying contaminants to concentrations affecting arctic wildlife and human health. Yet, concentrations of at least certain legacy POPs (which include polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethanes (DDTs), chlordane (CHLs), dieldrin, chlorobenzenes (ClBzs), and hexachlorocyclohexanes (HCHs)) have been effectively reduced in arctic wildlife over the last few decades through international agreements to restrict or eliminate the use of bioaccumulative substances. Arctic food webs are rapidly changing in response to warming air and ocean temperatures and related sea ice declines. Climate changes may alter contaminant distribution, pathways, and fate through environmental changes impacting, for instance, contaminant transport, deposition, and revolatilization. Climate-related ecological changes including changes in primary production, food web structure, habitat range, migration, and diets may also influence food web contaminant dynamics. In fact, structural food web changes have the potential to impose a large influence on contaminant exposures relative to other factors, but there has only been limited empirical study of these changes to date. Cumberland Sound is a large, deep, seasonally sea ice-covered and biologically productive ocean inlet of southeast Baffin Island (Nunavut, Canada). It is home to the Inuit community of Pangnirtung, and an important feeding region for marine mammals and migratory seabirds. As elsewhere in the Arctic, significant declines in sea ice extent (9.0% per decade) have been reported for this region. Concurrently, there are reports in Cumberland Sound and various arctic
marine ecosystems of increasing numbers of pelagic, transient, and subarctic-associated species. A poleward range shift of these subarctic species and displacement of arctic species, particularly at higher trophic levels, may in fact constitute a widespread climate change impact on arctic marine ecosystems.\textsuperscript{20} Harp seal (*Phagophilius groenlandicus*), which migrate between North Atlantic and Arctic waters, have increased in abundance.\textsuperscript{21} Capelin (*Mallotus villosus*), also a migratory and subarctic-associated species, have increased and extended their range northward.\textsuperscript{22} This phenomenon of subarctic forage fish moving into arctic waters under warmer conditions is also associated with shifts from benthic- to more pelagic-type arctic marine food webs.\textsuperscript{10} Sightings of killer whale (*Orcinus Orca*), a transient, actively sea ice-avoiding predator, have also increased.\textsuperscript{16,23} Other transient species within Cumberland Sound include Greenland sharks (*Somniosus microcephalus*), ringed seal (*Phoca hispida*), and harp seal (*Phoca groenlandica*). \textsuperscript{18} Species considered year-round residents include Cumberland Sound beluga whales (*Delphinapterus leucas*), in contrast to some other beluga populations which are known to undergo long-distance migrations.\textsuperscript{25} Similarly, although subadult ringed seals (*Pusa hispida*) have shown occasional long-distance movements, there is no convincing evidence that adult ringed seals undergo long-distance seasonal migrations as do other seals like harp seals.\textsuperscript{26–29} Here, we define a transient species as one that is present only intermittently in Cumberland Sound and is known to make substantial movements or migrations outside of the Sound (>1000 km), in contrast to a resident species that is assumed to reside year-round in Cumberland Sound.

Although many current and future climate change impacts on arctic marine ecosystems remain uncertain, those that have been documented have the potential to profoundly alter contaminant exposures in individual species, as well as overall food web relative to these other factors.\textsuperscript{26–29} Here, we define a transient species as one that is present only intermittently in Cumberland Sound and is known to make substantial movements or migrations outside of the Sound (>1000 km), in contrast to a resident species that is assumed to reside year-round in Cumberland Sound.

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### EXPERIMENTAL METHODS

**Sample Collection.** Major organisms in the Cumberland Sound food web were sampled in 2007–2008 (Table S1). Sampling sites and mixed zooplankton (mainly *Calanus* spp.) sampling (n = 6) were previously described.\textsuperscript{31} Benthic scallop (*Chlamys islandica*; n = 6) and snail (*Buccinum cyanum*; n = 3) were sampled by ponar; Atlantic herring (*Clupea harengus*; n = 1), arctic char (*Salvelinus alpinus*; n = 5), and scalpin (*Myoxocephalus scorpioides*; n = 9) were sampled by gill net; capelin (n = 5) were collected by dip net, and turbot (*Reinhardtius hippoglossoides*; n = 8), arctic skate (*Amblyraja hyperborea*; n = 5), and Greenland sharks (n = 11) were collected by bottom long line. Marine mammals, harp seal (n = 5), ringed seal (n = 5), beluga whale (n = 4), and narwhal (n = 7), were captured during Inuit subsistence hunts.

**Stable Isotopes Analysis.** $\delta^{13}$N and $\delta^{15}$N were assessed in lipid-removed, powdered samples of whole zooplankton, soft tissues of scallop and snail, and muscle tissues of fish and marine mammals. Samples were combusted in an elemental carbon source using a modified version of the two-source mixing model equation

\[
\begin{align*}
\text{Carbon source} &= 1 - \frac{\delta^{13}\text{C}_{\text{zooplankton}} - \delta^{13}\text{C}_{\text{consumer}} + \Delta \delta^{13}\text{C}(\text{TP}_{\text{consumer}} - \text{TP}_{\text{zooplankton}})}{\delta^{13}\text{C}_{\text{zooplankton}} - \delta^{13}\text{C}_{\text{scallop}}} \\
&= \text{Carbon source}
\end{align*}
\]

assuming that zooplankton represent the pelagic source, scallop represent the benthic source, and a constant trophic enrichment factor ($\Delta \delta^{13}\text{C}$) of 1.3‰ for consumers analyzed using muscle tissue.\textsuperscript{36} The $\Delta \delta^{13}\text{C}(\text{TP}_{\text{consumer}} - \text{TP}_{\text{zooplankton}})$ term accounts for species occupying varying TPs higher than the sources and thus for $\delta^{13}\text{C}$-enrichment between source and consumer depending on consumer TP relative to the source.

Carbon source values closer to 1 indicate more pelagic feeding.
and values closer to 0 indicate more benthic feeding. Although the magnitude of carbon source values is influenced by the choice of Δδ13C (just as TPs are influenced by choice of Δδ15N), the relative carbon source (or TP) among individuals is not, making it a suitable variable for studying contaminant food web behavior.

**Statistical Analyses of Contaminant–Food Web Relationships.** Prior to statistical analyses, contaminant concentrations were log-transformed, improving normality and reducing heteroscedasticity between species, and checked for outliers (z-score tests). Statistical analysis (Statistica, version 10.0; Statsoft, Tulsa, OK, USA) and reporting only proceeded for major contaminants (those with concentrations comprising at least 2% of the total concentration of all analyzed contaminants), and ≤2.5% nondetect values to avoid biased comparisons between contaminants due to different sample sets. These major contaminants consisted of Cl₃, Cl₄, Cl₅-PCB, Cl₆-PCB, Cl₍-PCB, Cl₅-PCB, Cl₆-PCB, t-nonachlor, heptachlor epoxide, p,p'-DDE, dieldrin, hexachlorobenzene (HCB), and α-HCH. The probability of making a Type I error was set to α ≤ 0.05. Tests were two-tailed unless otherwise indicated.

Effects of TP, carbon source, thermal group (poikilotherms versus homeotherms), and habitat range (resident versus transient species) on contaminant concentrations and biomagnification were tested using a general linear model (Type III sum-of-squares, n = 80)

\[
\log([\text{contaminant}]) = b_0 + b_1TP + b_2\text{group} + b_3\text{range} + b_4\text{carbon source} + b_5\text{group} \times TP + b_6\text{range} \times TP + b_7\text{carbon source} \times TP + e
\]

(2)

where \(b_0 \sim b_7\) are constants and \(e\) represents the error term, assumed to have a mean of zero and a constant variance. Predicted versus residual values for all individual contaminant models are provided in Figure S1, and normal probability plots are provided in Figure S2, for all reported contaminants.

Tolerance for each effect in the GLM models was consistently ≥0.01, indicating that the model did not violate the assumption of non-multicollinearity. An initial model also included season (April versus August) and season × TP terms, but excluded cetacean samples not having specific April or August collections. However, season terms were not significant for any contaminants. Therefore, the subsequent model excluded season terms and included the cetacean samples. Only first-order interaction terms involving TP were included in eq 2, in order to study how the contaminant–TP relationship, i.e. the biomagnification, is influenced by thermal group, habitat range, and carbon source. According to eq 2, average contaminant concentrations would differ between homeotherms and poikilotherms if group were significant and between transients and residents if range were significant. If carbon source were significant, average concentrations would differ with relative contribution of benthic and pelagic carbon sources. TMFs would differ between homeotherms and poikilotherms if group × TP were significant and between transient and residents if range × TP were significant. If carbon source × TP were significant, then TMFs would differ with relative benthic versus pelagic source contribution. The Type III sum-of-squares evaluates the effect of each factor after accounting for all other factors. Thus, the impact of each factor on the log-[contaminant]–TP relationship was illustrated by the GLM intercept and \(b\)-coefficients for TP, the given factor of interest, and their interaction term, to generate the log-[contaminant] values for different scenarios (i.e., homeotherm or poikilotherm, resident or transient, and benthic or pelagic (carbon source set to 0 or 1, respectively).

Food web accumulation was additionally investigated using TMFs as

\[
\text{TMF} = 10^{b_1}
\]

(3)

where \(b_1\) is the slope of the contaminant–TP regression

\[
\log([\text{contaminant}]) = b_0 + b_1TP + e
\]

(4)

considering a resident-only versus a resident-and-transient food web. TMFs significantly greater than 1 indicated a biomagnifying contaminant.

Biomagnification was compared between individual transient and resident species that occupied similar TPs: harp seal versus ringed seal, narwhal versus beluga whale, and Greenland shark versus arctic skate. Species-specific BMFs were calculated from teleost fish prey to marine mammal or elasmobranch fish predator as

\[
\text{BMF} = \frac{[\text{contaminant}]_{\text{predator, lipid weight}}}{[\text{contaminant}]_{\text{plankton, lipid weight}}} = \frac{TP_{\text{predator}}}{TP_{\text{plankton, lipid weight}}}
\]

(5)

where the prey were considered to be all lower trophic fish (herring, capelin, char, sculpin). Turbot were not considered prey fish since their TPs were higher than those of all other species investigated, likely due to a largely piscivorous diet. A TMF or BMF statistically >1 indicated biomagnification through the food web or from prey-to-predator, respectively.

**RESULTS AND DISCUSSION**

**Trophic Position, Carbon Source, and Contaminant Concentrations.** Estimated TPs from δ15N ranged from ≈2 in zooplankton (assigned to TP2) and sculpin to ≈4 in Greenland shark and turbot (Figure 1). Carbon sources ranged from ≈0 to ≈4.

![Figure 1: Trophic position (TP) versus carbon source for zooplankton (○), benthic invertebrates (▲; scallop, snail), teleost fish (●; herring, capelin, char, sculpin, turbot), elasmobranch fishes (◇; arctic skate, Greenland shark), pinnipeds (□; harp seal, ringed seal), and cetaceans (●; beluga whale, narwhal) from the Cumberland Sound food web sampled in 2007–2008.](Image 337x131 to 552x326)
(more benthic-type feeding) in scallop and snail to ≈1 (more pelagic-type feeding) in zooplankton, Atlantic herring, capelin, arctic char, turbot, arctic skate, beluga whale, and narwhal. Species with intermediate values, suggesting input from benthic and pelagic sources, included in order of increasingly pelagic contribution: sculpin < harp seal < ringed seal < Greenland shark.

In the overall model, lipid weight contaminant concentrations except for α-HCH, were positively correlated with TP, indicating biomagnification through the food web (Table 1, Figure 2, Tables S2, S3). A statistically significant proportion (8% for the trichlorinated-PCBs (Cl3-PCB) to 18% for t-nonachlor) of the explained variance in biomagnifying substance concentrations was explained by TP alone.

It bears noting that here, as in previous studies, we generated stable isotope ratios from fish and marine mammal lipid-free muscle samples to characterize food web structure. Yet, lipophilic substances are preferentially deposited in lipid-rich tissues. There are two potential issues with such an approach. In marine food webs, dietary consumption consists mainly of proteins and lipids, the proportions of which vary between prey items. Thus, due to isotopic routing, estimated TPs and carbon sources based on lipid-free muscle may reflect the protein portion of the diet, but not necessarily the overall diet. For example, we could have underestimated Greenland shark TPs because consumption of lipid-rich seals versus more proteinaceous fish would not be fully reflected in muscle stable isotope ratios. The result would be higher contaminant concentrations than expected based on δ15N-derived TPs. The second issue is that nitrogen and carbon turnover rates in muscle may be faster than the turnover rate of contaminants. Overall then, muscle stable isotope ratios may represent more recent feeding and than the turnover rate of contaminants. Overall then, muscle samples to characterize food web structure. Yet, due to isotopic routing, estimated TPs and carbon sources were derived from δ13C-values. Thermal groups were poikilotherms versus homeotherms. Habitat ranges were residents versus transients. Carbon sources were derived from δ13C-values.

<table>
<thead>
<tr>
<th>contaminant</th>
<th>whole model</th>
<th>parameter estimates: effect sizes (partial ( \eta^2 )), b-coefficients (*( p &lt; 0.05 ); **( p &lt; 0.01 ); ***( p &lt; 0.001 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cl3-PCB</td>
<td>0.19</td>
<td>0.08, 0.75a 0.07, 2.1a 0.07, −0.64a 0.00, 0.08 0.00, −0.07 0.02, −0.70 0.00, 0.10</td>
</tr>
<tr>
<td>Cl3-PCB</td>
<td>0.45 &lt;0.001</td>
<td>0.18, 1.0*** 0.04, 1.4 0.05, −0.48 0.05, 0.95 0.07, −0.34a 0.05, −0.94a 0.01, 0.10</td>
</tr>
<tr>
<td>Cl3-PCB</td>
<td>0.43 &lt;0.001</td>
<td>0.12, 0.75** 0.01, 0.72 0.02, −0.25 0.06, 0.93a 0.08, −0.35a 0.08, −1.0a 0.05, 0.22</td>
</tr>
<tr>
<td>Cl3-PCB</td>
<td>0.58 &lt;0.001</td>
<td>0.07, 0.75a 0.01, −0.77 0.00, 0.13 1.9, 2.5*** 0.24, 0.67*** 0.07, −1.3a 0.05, 0.36</td>
</tr>
<tr>
<td>Cl3-PCB</td>
<td>0.65 &lt;0.001</td>
<td>0.12, 0.82** 0.05, −0.35 0.00, 0.02 23, 2.2*** 0.29, 0.79*** 0.02, −0.59 0.01, 0.13</td>
</tr>
<tr>
<td>t-nonachlor</td>
<td>0.76 &lt;0.001</td>
<td>0.18, 1.0*** 0.02, −0.97 0.01, 0.16 1.9, 1.9*** 0.26, 0.72*** 0.02, 0.04 0.01, −0.10</td>
</tr>
<tr>
<td>heptachlor epoxide</td>
<td>0.87 &lt;0.001</td>
<td>0.10, 0.52a 0.06, −0.24 0.00, −0.02 2.2, 1.5*** 0.27, 0.54*** 0.07, −0.77a 0.04, 0.18</td>
</tr>
<tr>
<td>p,p′-DDE</td>
<td>0.76 &lt;0.001</td>
<td>0.10, 0.36** 0.07, −2.5a 0.04, 0.58 0.27, 3.3*** 0.34, 1.2*** 0.00, −0.02 0.00, 0.12</td>
</tr>
<tr>
<td>dieldrin</td>
<td>0.42 &lt;0.001</td>
<td>0.09, 0.60a 0.00, 0.09 0.00, −0.09 0.05, 0.86a 0.08, −0.32a 0.00, 0.14 0.00, −0.04</td>
</tr>
<tr>
<td>HCB</td>
<td>0.53 &lt;0.001</td>
<td>0.10, 0.81** 0.00, −0.19 0.00, 0.00 0.02, 0.62 0.04, −0.26 0.04, 0.87 0.01, 0.17</td>
</tr>
<tr>
<td>α-HCH</td>
<td>0.38 &lt;0.001</td>
<td>0.04, 0.31 0.02, 0.63 0.03, −0.25 0.00, 0.17 0.00, 0.00 0.04, 0.54 0.09, −0.27a</td>
</tr>
</tbody>
</table>

*General linear model (see Methods section), where the contaminant values are log-transformed ng/g lipid weight concentrations. The Cl3-PCB, Cl2-PCB, Cl1-PCB, Cl0-PCB, and Cl−PCB are the sum of tri-, tetra-, penta-, hexa-, and hepta-chlorinated PCB homologue group concentrations, respectively, representing on average 95% of the sum-PCB concentrations. Consumer trophic positions (TPs) were derived from δ13N-values. Thermal groups were poikilotherms versus homeotherms. Habitat ranges were residents versus transients. Carbon sources were derived from δ13C-values.
Atlantic, a more contaminated area relative to arctic regions. These seals may pick up a more contaminated North Atlantic-type signature during winter-feeding and transport it to the Arctic during summer migration northwards. This biovector hypothesis was supported by harp seal BMF patterns versus ringed seal (Figure 5). The specific contaminants that showed higher BMFs in harp seal (higher-chlorinated PCBs and t-nonachlor) are consistent with a more temperate region-type signature. Thus, increasing abundance of harp seal in Cumberland Sound has implications for contaminant exposures in top predators that opportunistically feed on marine mammals, such as polar bears, if they are shifting their diets to greater consumption of harp seals. 

Regardless of transient species acting as biovectors through higher accumulation (energetic requirements of greater movement) or higher exposures (in areas outside Cumberland Sound), the overall ecosystem contamination and the amount that is passed from one trophic position to the next (i.e., trophic transfer) is higher, on average, than if these species were not present.

Differences in BMF patterns between elasmobranchs and cetaceans versus pinnipeds suggested that other additional factors could also influence the BMFs. Unlike seals, both parent contaminant (t-nonachlor) and metabolite (heptachlor epoxide, p,p'-DDE) BMFs were higher in Greenland sharks and narwhals versus arctic skates and belugas, respectively (Figure 5). Although Greenland sharks and narwhals undergo extensive movements, these movements do not clearly bring them to more contaminated feeding grounds, supporting the bioenergetics versus biovector hypothesis for these transients. Yet,
Greenland sharks could also have higher BMFs from feeding on higher TP marine mammals or greater accumulation over longer life spans. Another explanation may involve using a constant trophic enrichment factor to assess TPs, whereas species-specific factors may be more appropriate. In the cetacean comparison, differences could partly be because the narwhals were all males, whereas only one of four belugas was a male. Limited indications from other published studies have suggested similar narwhal and beluga organic contaminant levels. Yet, differences between their contaminant levels here appeared larger than what would likely be explained by sex differences alone, and sex differences in diet have not been identified for either species. Moreover, Cumberland Sound belugas are known to be residents, whereas all other high-latitude (>60° N) populations are known to migrate considerable distances. Another possible issue is that for some species, certain age classes may exhibit long-distance movements, whereas others do not. For example, some juvenile ringed seals are known to migrate long distances, whereas evidence suggests that adult ringed seals show a much more restricted range. We were not able to obtain ages for all of the samples in this study, but it would be worthwhile in future work to investigate the consequences of these age-related habitat use differences on POP burdens. Season. Season did not significantly influence contaminant concentrations (Table S5). Low statistical power to detect differences or interyear variation may partly explain why no significant seasonal effect was observed. Although samples were collected in 2007 and 2008, the influence of interyear variation in contaminant concentrations could not be assessed in the current data set. Annual changes in arctic marine animal legacy contaminant concentrations are generally less than 2%/year.
Although higher for DDTs, HCHs, and ClBzs. Nonetheless, food web concentrations were generally higher in the ice-free versus ice-covered season. In the Northwater Polynya, ΣClBz, ΣHCH, and ΣCHL, but not ΣPCB and ΣDDT concentrations, increased from April to July in herbivorous zooplankton. Possible explanations for these patterns include seasonal changes in hydrophobic organic contaminant aquatic concentrations related to particulate organic matter concentrations or source water variation into the region, in addition to lipid dynamics and food availability differences between seasons. Significant seasonal concentration differences were also reported for a Norwegian Arctic marine food web. However, the species composition of the food web differed between seasons. For instance, higher concentrations and TMFs were generally found in July, but greater numbers of subarctic-type zooplankton and fish including capelin were sampled in July. Thus, the observed concentration change may partly be related to seasonal changes in species abundances, including transient species.

These findings may suggest that climate-associated shifts to more prolonged ice-free periods would not have a substantial direct impact on food web contaminant concentrations or TMFs. However, greater ice-free periods may lead to other indirect changes, such as increased volatilization from surface reservoirs and increased primary production. In the latter case, this would generally lead to decreased bioaccumulation due to decreased bioavailability from higher particulate organic carbon levels. Our data suggest that a main concern with respect to greater ice-free periods is increased abundance of more contaminated ice-avoiding transient or subarctic species.

Carbon Source. Carbon source was not a major factor explaining most contaminant concentrations or TMFs (Table 1). The exceptions were a small, but significant, main effect of carbon source for Cl4–PCB to Cl6–PCB and heptachlor epoxide concentrations. That is, TMFs were not significantly different for benthic versus pelagic carbon source, but across TPs, benthic source was associated with higher concentrations of these substances. For α-HCH concentrations, there was no main effect of carbon source, but significant interaction with TP showed that the TMF was higher for a benthic versus pelagic carbon source (Figure 3b).

Higher benthic concentrations for lower-chlorinated PCBs and heptachlor epoxide, and TMF for α-HCH, may be related to differing water-solubilities compared to other contaminants. Species having benthic-type carbon source values were collected in near-shore, shallow waters. Surface waters contain the highest concentrations of lower-chlorinated PCB and HCH, which decline with depth. Conversely, more hydrophobic contaminants undergo greater particle scavenging, allowing rapid deep water transport and more heterogeneous water column distributions. So, differences in benthic versus pelagic α-HCH, heptachlor epoxide, and lower-chlorinated PCB concentrations may be related to higher concentrations of these more surface-associated contaminants in shallow water, near-shore species.

Based on these findings, possible climate-associated shifts to increasingly pelagic-type food webs would not necessarily increase food web contaminant concentrations and TMFs. This speculation does not take into account other potential differences in pelagic replacement species, such as migration patterns or other changes in trophic relationships. Nonetheless, we would expect slightly lower food web concentrations of more water-soluble contaminants under increasingly pelagic-type food web regimes. Higher concentrations of these contaminants have also been found in surface water-associated sympagic amphipods than pelagic zooplankton. Therefore, shifts from benthic/nearshore and ice-associated food webs to pelagic-type food webs could potentially additively contribute to reducing food web concentrations of these contaminants, but would not necessarily have substantial impacts on the more recalcitrant and environmentally predominant higher-chlorinated PCBs and DDTs.

Thermoregulation. The inclusion of homeotherms and poikilotherms generally had no significant effect on contaminant concentrations or TMFs (Table 1, Figure 3c). The exceptions were small, but significant, relationships of p,p′-DDE to thermal group and of Cl3–PCB to thermal group and group × TP. Higher p,p′-DDE concentrations, and a similar though not statistically significant pattern for other recalcitrant pollutants, in homeotherms than in poikilotherms was likely related to higher energetic requirements, longevity, and lack of water-exchange as an elimination route in homeotherms. Concentrations of Cl1–PCB increased with TP, but the TMF differed between homeotherms and poikilotherms, related to different metabolic capacity toward lower-chlorinated PCBs.

It will be important, but logistically and financially challenging, to develop long-term temporal, full food web data sets to further consider the influence of ecosystem changes, and to consider these changes in combination with direct changes in concentrations in abiotic compartments (air, seawater) due to climate warming and changing emissions. Nonetheless, we have shown here that the presence of transient species may be a strongly influential factor given its importance relative to other established factors influencing bioaccumulation of persistent pollutants.

ASSOCIATED CONTENT

Supporting Information

Contaminant analysis, quality control, biological and collection data, TP and carbon source, raw contaminant concentrations, results of contaminant–TP correlations (TMFs), general linear model results including season factor, residual versus predicted values and normal probability plots for the GLM, additional panels showing relationship to TP of contaminant concentrations. This material is available free of charge via the Internet at http://pubs.acs.org.

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The authors declare no competing financial interest.

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