

*Special Issue Honoring Don Mackay*

**BIOLOGICAL AND CHEMICAL FACTORS OF IMPORTANCE IN THE  
 BIOACCUMULATION AND TROPHIC TRANSFER OF PERSISTENT  
 ORGANOCHLORINE CONTAMINANTS IN ARCTIC MARINE FOOD WEBS**

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(Received 18 September 2003; Accepted 10 February 2004)

**Abstract**—Recent studies of arctic marine food webs have provided detailed insights regarding the biological and chemical factors that influence the bioaccumulation and trophic transfer of persistent organochlorine (OC) contaminants in aquatic systems. The present paper summarizes the recent literature with an emphasis on identifying important ecological factors for explaining variability of OC concentrations among organisms. The Arctic ecosystem has a number of unique attributes, including long food chains, reduced diversity of species, similar food webs across the entire region, and limited influence from pollution point sources. Lipid content, body size, age, gender, reproduction, habitat use, migration, biotransformation, seasonal changes in habitat conditions, feeding ecology, and trophic position have all been demonstrated to influence OC concentrations and bioaccumulation in arctic marine biota. The relative importance of each factor varies among OCs and organisms. Diet or trophic level is the dominant factor influencing OC concentrations and dynamics in seabirds and marine mammals, although biotransformation can significantly influence nonrecalcitrant OCs, such as hexachlorocyclohexane isomers. Dietary accumulation of OCs is also an important route of exposure for arctic fish and zooplankton, and biomagnification of OCs may also occur among these organisms. To date, only limited attempts have been made to model trophic transfer of OCs in the arctic marine food web. Although models developed to assess OC dynamics in aquatic food webs have included some biological variables (e.g., lipid content, feeding rate, diet composition, and growth rate), selection of processes included in these models as well as their mathematical solutions and parameterization all introduce simplification. This reduces biological validity of the models and may be particularly problematic in a highly seasonal environment, such as the Arctic Ocean.

**Keywords**—Biomagnification Migration Reproduction Age Lipid content

**INTRODUCTION**

Since man-made agricultural and industrial organochlorine (OC) contaminants were first detected in nontarget organisms in the environment (see, e.g., [1,2]), factors causing differences in concentrations between species have been a major focus of ecotoxicology and environmental chemistry research. Organochlorine compounds, such as polychlorinated biphenyls (PCBs) and DDT, were designed as persistent to be effective for their respective industrial and agricultural uses (for a detailed description of use, structure, and properties, see [3]). However, it was observed that many OCs increased in concentration with each trophic level in aquatic food webs, a process termed biomagnification. Uptake rates of most hydrophobic OCs are similar between species (see, e.g., [4]), and the elimination rate is what determines whether a chemical biomagnifies. Chemicals with fast elimination are less likely to biomagnify. Slow elimination, however, does not permit the chemical concentration in an organism to reach equilibrium with that in food or water, and biomagnification will thus occur.

The definition of biomagnification has varied slightly over the past three decades but was recently and, in our opinion, accurately defined by Gobas and Morrison [5] as “the process in which the chemical concentration in an organism achieves a level that exceeds that in the organism’s diet, due to dietary

accumulation.” A biomagnification factor (BMF) can be calculated using Equation 1:

$$BMF_{\text{lipid weight}} = \frac{[OC_{\text{organism}}]_{\text{lipid weight}}}{[OC_{\text{food}}]_{\text{lipid weight}}} \quad (1)$$

A second, less commonly used definition of biomagnification is “the process of accumulation of a xenobiotic from diet whether it increases in concentration or not” [6]. However, this definition is better described as trophic transfer or as dietary accumulation. Potential confusion may result from the second definition, because chemicals accumulated from food (i.e., trophic transfer) do not necessarily biomagnify, even when food is the dominant route of exposure.

Aquatic organisms may also accumulate OCs directly from water. Termed bioconcentration, this is a process by which the chemical concentration in an aquatic organism exceeds that in water as a result of exposure to waterborne chemicals [5]. The bioconcentration factor (BCF) can be calculated using Equation 2:

$$BCF_{\text{lipid weight}} = \frac{[OC_{\text{organism}}]_{\text{lipid weight}}}{[OC_{\text{water}}]_{\text{dissolved}}} \quad (2)$$

Exposure to most OCs through water only occurs solely for aquatic organisms under laboratory settings, because in the wild, these organisms are exposed to OCs through water, food, and sediment. The exception would be pelagic algae, which do not feed or have contact with sediment. However, uptake of hydrophobic OCs ( $\log K_{ow} > 5$ ) from water is considered to be negligible for most fish compared to dietary uptake. For

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aquatic birds and mammals, exposure to OCs through diet is the only relevant exposure pathway.

Bioaccumulation has been defined as the net process by which the chemical concentration in an aquatic organism achieves a level exceeding that in the water as a result of chemical uptake through all possible routes of exposure (i.e., water or food) and elimination from all possible routes [5]. The bioaccumulation factor (BAF) can be used to assess the ecological risk of OCs in aquatic ecosystems [5,7–9] and is calculated by Equation 3:

$$\text{BAF}_{\text{lipid weight}} = \frac{[\text{OC}]_{\text{field exposed organism}} \text{lipid weight}}{[\text{OC}]_{\text{water}} \text{dissolved}} \quad (3)$$

When field or laboratory organism data are not available, BAFs can be estimated by empirical or mechanistic models [7,9]. Bioaccumulation in aquatic organisms has successfully been described and quantified by empirical and mechanistic models, as reviewed and summarized by Mackay and Fraser [9] and by Gobas and Morrison [5]. Empirical models are relationships based on correlations of chemical measurement data, such as between a BAF in a particular species and the chemical's octanol–water partition coefficient ( $K_{ow}$ ). Mechanistic bioaccumulation models are based on hypotheses about the processes giving rise to the observed data. They assume that OC bioaccumulation in whole organisms can be predicted by quantifying uptake and loss process rates [5,9–11]. At steady state, the uptake and loss process rates are of equal magnitude, so the OC concentration ( $[\text{OC}]$ ) in an organism can be described by Equation 4:

$$[\text{OC}]_{\text{organism}} = \frac{[\text{OC}]_{\text{exposure}} k_{\text{in}}}{k_{\text{out}}} \quad (4)$$

where  $[\text{OC}]_{\text{exposure}}$  is the OC concentration to which the organism is exposed and  $k_{\text{in}}$  and  $k_{\text{out}}$  are the respective uptake and loss process rates (first-order rate constants with units of  $\text{time}^{-1}$ ) for the processes included in the model. Uptake processes usually include direct partitioning from the abiotic environment and ingestion of contaminated dietary sources. Loss processes include direct partitioning, biotransformation, growth dilution, parental transfer to offspring, and egestion. Equation 4 can be used to determine whether the OC concentration in an organism will exceed that in its food or if it will be governed by the direct partitioning with water. The equation may also be expanded to include the trophic relationships of the organisms involved, allowing modeling of the contaminant flow in food webs and the competing influences of uptake and loss. In addition to choosing the processes to include in the model, algebraic or numerical solutions must be chosen to describe each of the processes, and finally, parameters (i.e., values) must be chosen to scale the processes. In each of these steps (i.e., choosing and describing processes, and choosing values for the equations), assumptions are made regarding the specific system of study. Thus, although aquatic food web models developed during the last several decades successfully predict OC behavior in invertebrates and fish [12–14], as validated by a Lake Ontario dataset [15], a need exists for further validation of these and other conceptual models in different ecosystems, such as the marine. This has been difficult, however, because of a lack of sufficiently detailed, species-specific OC data.

The quantification of persistent OCs, such as PCBs, chlor-dane, and DDT-related products, in the arctic marine environment over the past four decades has dramatically increased the

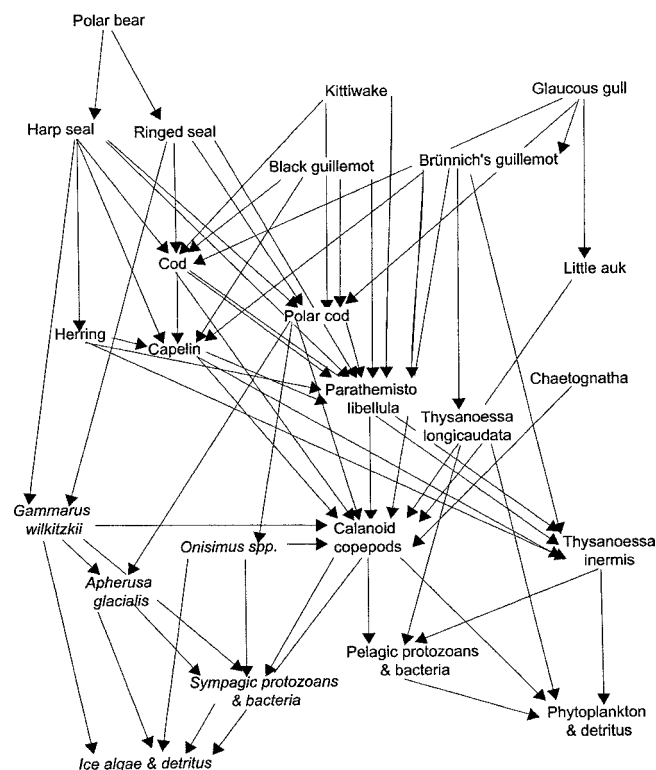


Fig. 1. Diagram of simplified arctic marine food web, including ice-associated (italic) and pelagic organisms. The diagram includes only some of the dominant species (by numbers and biomass), and the arrows illustrate trophic links by pointing to the organism that serve as food. For simplicity, some of the trophic interactions, such as seabirds feeding on ice fauna, are not shown, and the strength of trophic links is not represented.

breadth and depth of our knowledge regarding how this class of contaminant accumulates in biota and is subsequently transferred within food webs. The arctic marine ecosystem provides an excellent opportunity to study food web dynamics of OCs, because the contribution of point sources to regional contamination is relatively minor, a high dependence of lipids is found in arctic marine food webs, and the food webs tend to be long and simple (Fig. 1). Furthermore, marine mammals and seabirds are generally not endangered, and samples are readily available from Inuit hunters. Earlier field studies of contaminant bioaccumulation in arctic marine organisms included fish and species occupying higher trophic levels (e.g., seals and polar bears) (see, e.g., [16,17]), but marine invertebrates were either not included or were included without separation into taxonomic or functional groups [18]. Other studies lumped invertebrates into selected size classes [19,20]. Field studies of OC accumulation in arctic marine food webs have only recently begun to investigate the importance of biology and ecology of lower-trophic-level organisms in the bioaccumulation process. These studies either sort the marine invertebrates by species or genus [21–27], or they consider samples dominated by one species [28,29].

A number of comprehensive studies have examined the distribution of OCs in arctic marine food webs [21,26, 27,29,30]. This has resulted in detailed datasets and new knowledge concerning chemical and biological variables that influence the bioaccumulation and trophodynamics of OCs in marine food webs. We are using the Arctic Monitoring and Assessment Program boundaries for defining the Arctic region

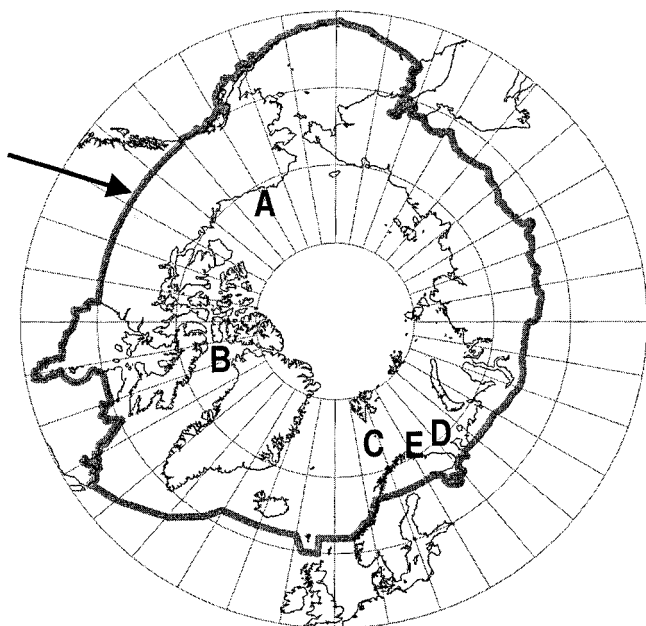


Fig. 2. Map of the Arctic region as defined by the Arctic Monitoring and Assessment Program (indicated by arrow; Murray [185]) and location of recent food web studies. A = Hoekstra et al. [26]; B = Fisk et al. [21] and Moisey et al. [127]; C = Borgå et al. [29]; D = Muir et al. [27]; E = Ruus et al. [30].

(Fig. 2). The present paper summarizes this new knowledge, with an emphasis on important ecological factors for explaining the variability of OC concentrations among arctic organisms. To emphasize factors that might need to be considered in greater detail as the discipline of food web modeling of persistent organic pollutants (POPs) behavior advances, we discuss and provide an overview of the chemical and biological (i.e., ecological and physiological) factors of importance in the bioaccumulation, trophic transfer, and concentrations of OCs in arctic biota; the methods and utility of examining bioaccumulation across whole food webs; and the use of models in understanding and studying the bioaccumulation and trophic transfer of OCs in aquatic food webs. This summary focuses on the arctic marine ecosystems, but the information is applicable and relevant to all marine and freshwater systems.

#### CHEMICAL FACTORS INFLUENCING BIOACCUMULATION AND TROPHIC TRANSFER OF OCs

The persistent OCs that are the focus of the present review have been identified as POPs [31]. They are a structurally diverse group of compounds; however, they have many physicochemical properties in common, such as high hydrophobicity and relatively low vapor pressure. They also share structural features, such as high proportions of chlorine substitution on cyclic aliphatic or aromatic rings. They generally have molecular weights of less than approximately 600 amu and molecular cross-sections of less than 9.5 Å and are resistant to chemical and microbial degradation [32]. Their properties have been well documented by Don Mackay and coworkers (compiled in [33]), and this information forms the foundation for use of physicochemical properties to predict bioaccumulation.

Perhaps the most important property—and the most widely studied—is hydrophobicity. Hydrophobicity is usually expressed in terms of  $\log K_{ow}$ , which accounts for a chemical's solubility in water but also in lipid (octanol is a surrogate for

lipids). As noted by Mackay et al. [34], differences in  $K_{ow}$  between OCs are generally the result of changes in water solubility, because OCs are all highly lipid soluble whereas their water solubilities differ substantially. The  $K_{ow}$  is a key parameter in most OC food web models [13,35], which assume that the OC uptake from water or food is directly determined by the organism's lipid content and the  $K_{ow}$  of the OC. The  $K_{ow}$  values are among the most accurately predicted physical properties [36]. For example, the program KOWWIN (Version 3.11; Syracuse Research, Syracuse, NY, USA) is based on 12,000 experimental  $K_{ow}$  values, many of which are chlorinated organics. Unfortunately, for many industrial chemicals in commerce, accurate and experimentally derived  $K_{ow}$  values are not always available; thus, the applicability of bioaccumulation models that require the  $K_{ow}$  as a basic input is often limited.

The  $\log K_{ow}$  values of the OCs discussed in the present paper range from approximately 3.0 to 8.0 and are positively correlated with both chlorine number and molecular size [33]. Although bioaccumulative compounds are defined as having a  $\log K_{ow}$  of 5.0 or greater [31], some recalcitrant OCs with a  $\log K_{ow}$  of less than 5 but greater than 3.5 (e.g.,  $\beta$ -hexachlorocyclohexane [ $\beta$ -HCH]) have been shown to biomagnify in aquatic food webs [21]. For OCs, elimination rates decrease, and bioaccumulation generally increases, with  $\log K_{ow}$  [5,12,37]. For example,  $\log$  BAFs increase with  $\log K_{ow}$  in arctic marine zooplankton [21,26]. Also, trophic magnification factors (TMFs; also called food web magnification factors) have been shown to increase with increasing  $\log K_{ow}$  in arctic marine food webs, with the greatest values for OCs in the  $\log K_{ow}$  range of 6.5 to 7.0 [21]. However, very hydrophobic OCs ( $\log K_{ow} > 7.5$ ) have lower BAFs and TMFs than OCs with  $\log K_{ow}$  values between 6.5 and 7.5 [5,12,21]. Although no consensus has been reached about the cause, possible explanations include lower bioavailability and faster elimination by feces [5,12].

The number and position of chlorines on the OC molecules play an important role in their food web dynamics. In general, increasing chlorination increases hydrophobicity [33,38,39] and reduces biotransformation rates [12,40], which result in greater bioaccumulation and longer half-lives of OCs in biota. The exceptions to this relationship are the highly chlorinated OCs that are very hydrophobic ( $\log K_{ow} > 7.5$ ). Chlorine position can play a role in the susceptibility of OCs to biotransformation. For example, PCB congeners without chlorine atoms at adjacent *para*- and *meta*- or *ortho*- and *meta*-positions are more readily biotransformed than congeners with such chlorine atoms [41,42]. Similarly, toxaphene congeners with 2-endo-, 3-exo-, 5-endo-, and 6-exo-substitution of chlorines on the six-membered ring are more recalcitrant than those with unsubstituted positions [43]. The  $\beta$ -HCH, which is the only fully equatorially substituted HCH isomer, is also the only recalcitrant HCH isomer in most mammal species [44].

#### BIOLOGICAL FACTORS INFLUENCING THE BIOACCUMULATION AND TROPHIC TRANSFER OF OCs

Beyond chemical factors, a number of biological factors influence the bioaccumulation and trophic transfer of OCs in arctic marine biota. None of the factors are completely independent in nature, and they are correlated, to some degree, due to species and chemical characteristics. They include lipids, seasonality, reproduction, body size, age, sex, life cycle, biotransformation, habitat use, migration, and feeding ecology. Typically, most studies of bioaccumulation and trophic transfer

of OCs have used lipid-based (or normalized) concentrations, which reduces the influence of variable lipid content on OC concentrations. However, some of the variables discussed, particularly seasonality and reproduction, also influence lipid dynamics in organisms. In these cases, the role of lipids will be discussed beyond lipid normalization.

### Lipids

Of particular importance in the Arctic ecosystem is the dynamics of lipids in organisms. The OCs discussed in the present paper are highly lipid soluble and, thus, are significantly influenced by lipid dynamics. Most comparative studies on OC levels in aquatic biota consider the influence of lipids by either lipid-normalizing the concentration (i.e., [OC]/lipid content) [35], by using lipid content as a covariate in statistical models [45], or by using the residuals of the regression of OC levels on lipids for further analysis [45,46]. All these approaches assume that the capacity to accumulate OCs is linearly correlated with the organism's lipid content. In addition, these lipid-normalizing methods assume that the neutral storage lipids are the main organic phase into which chemicals partition, although for phytoplankton, PCBs may also partition into other organic carbon phases [47,48]. Although a general linear relationship exists between lipid content and OC concentration both within and among temperate aquatic species, this relationship has rarely been studied for organisms with changing seasonal lipid contents, as is the case for arctic species. In addition, many arctic marine zooplankton do not feed during winter, and they store energy in more energy-dense wax esters as well as in triacylglycerols [49,50]. Triacylglycerol is the dominant neutral lipid class in vertebrates, and most studies that have shown an increasing OC concentration with lipid content have used triacylglycerols (see, e.g., [51]). To our knowledge, the relevance of wax esters has not been considered in OC bioaccumulation by arctic zooplankton. If OCs partition differently into wax esters and triacylglycerols, then OC bioaccumulation will differ among arctic zooplankton depending on their over-wintering strategies and lipid storage.

Lipid storage makes resources less available for maintenance, growth, and reproduction. Thus, arctic animals have a longer life span and a later maturation compared with temperate organisms having a more stable resource availability that favors earlier maturation and greater allocation to reproduction. An organism's adaptation regarding lipid storage therefore influences OC bioaccumulation as a result of increased life span (both in poikilotherm and homeotherms; see *Age, sex, and life cycle*), higher OC storage capacity as a result of the higher lipid content (given the lipid-soluble OCs), and efficient OC transfer to offspring as a result of lipid-rich eggs and milk (see *Reproduction*). It has been suggested that the high lipid content and diet of arctic organisms and their efficient energy transfer also increase the OC transfer from prey to predator [3]. Lipids are assimilated more efficiently from the gut than are proteins [52], and cotransport of OCs with lipids would support a more efficient OC uptake in a high-lipid diet [53]. However, a recent experimental study of OC accumulation in ringed dove showed that PCB assimilation efficiencies were similar between doves fed a high- and doves fed a low-lipid diet [54]. Although OC levels in arctic biota are generally low compared with those in industrialized and agricultural areas, the seasonal OC redistribution to target organs from the inactive lipids, as lipids are used for energy,

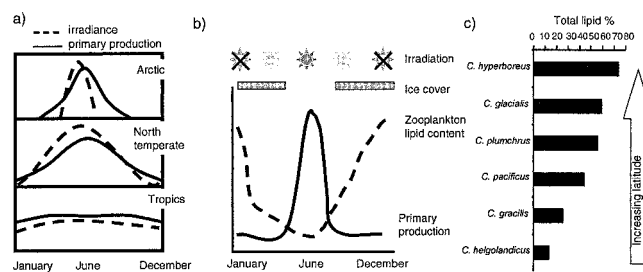


Fig. 3. (a) Schematic diagram of seasonal variation in irradiation and primary production with latitude. (b) Schematic diagram of seasonal variation in irradiation and ice cover, primary production, and arctic marine zooplankton lipid accumulation. (c) Increasing lipid content as a percentage of dry weight in *Calanus* copepods with increasing latitude (data from Lee et al. [186]).

renders arctic biota at greater risk of OC-related effects [55,56].

### Seasonality

Seasonality influences OC accumulation through altered organism properties and OC bioavailability as the marine ecosystem responds to changes in irradiation. In contrast to the distinct seasonal primary production in the Arctic, primary production and, thus, food availability are more evenly distributed throughout the year at lower latitudes (Fig. 3a). Arctic marine herbivores exposed to such marked variations in phytoplankton production have adapted by storing large amounts of energy (in the form of lipids) that can be mobilized in times of food shortage [49]. The utilization of lipids as an energy source is not unique to northern, high-latitude regions [57]; however, the magnitude of the processes is enhanced in the Arctic because of stronger seasonal variation in solar energy intensity (Fig. 3a) and primary productivity (Fig. 3b). Thus, the importance of lipids increases with increasing latitude from the tropics to the Arctic (Fig. 3c).

Increasing light intensity can also influence OC bioavailability because of changes in ice cover and phytoplankton production. Although the water concentrations of OCs seem to be determined mostly by vertical mixing and advection, air-water gas exchange may influence the surface-water concentrations during the open-water (i.e., ice-free) period [58]. For compounds that have been supersaturated in Arctic Ocean water, open-water periods lead to out-gassing from the water to the atmosphere and, thus, lower water concentrations [59]. In the water-surface layer (upper 50 m), the OC concentrations were highest during ice-covered periods of winter and lowest during the open-water period of summer [58]. This is reflected in algae, in which OC concentration decreased in May as concentrations in the water decreased [20]. Furthermore, melting sea ice might also lead to a release of contaminants into the water. These contaminants are transported along with the sea ice, accumulated from atmospheric deposition or from particle inclusion in the ice-production areas on the shelf seas [60]. However, OCs are removed from the water column and deposited to the benthic compartment of the marine environment because of the high particle affinity of OCs and the rapid release of particulate matter from sea-ice sediment in the spring [61] and, thus, are no longer available for uptake pelagic organisms (see *Habitat use*).

The increased light intensity and open water also influence the bioaccumulation of OCs in terms of increased phytoplankton production (see, e.g., [20]) (Fig. 3b). Because of sorption,

the portion of OCs available for direct uptake from water decreases with increasing phytoplankton biomass [62]. As a result, the fraction of OCs subjected to sedimentation increases with the increase in algal production. In addition, increased phytoplankton biomass dilutes the average OC concentration in algae; thus, dietary uptake by herbivorous zooplankton decreases [62]. Because OC concentrations in zooplankton respond quickly to water OC concentration (because of both direct partitioning and changed dietary exposure from algae), seasonal changes in water concentrations are reflected not only in primary producers but also in zooplankton [20,22]. In mixed zooplankton samples from Lancaster Sound in the Canadian Arctic, all OC concentrations on a lipid-weight basis (except  $\alpha$ -HCH) decreased during the open-water period, being lowest from August to September [20]. This resulted, in part, from the reduced OC water and algae concentrations, but it may also have been related to increasing zooplankton lipid content during this time.

Seasonality might also affect OCs in biota as a result of changing behavior in terms of migration and feeding preferences. For instance, many birds and mammals migrate to the productive Arctic region in summer to feed on the lipid-rich zooplankton and fish [63], which likely have much lower OC concentrations than prey from southern habitats [21]. In addition, feeding preferences of an organism may change during the year because of prey availability, resulting in different OC exposures. Thus, organisms may be exposed to different OC concentrations and profiles throughout the year. This may have a profound effect on food web structure interpretation and might produce erroneous results in terms of calculation of BMFs, which assume that an equilibrium has been reached between the OCs in the predator and in its prey.

The influence of seasonality on OC bioaccumulation has been studied in terms of primary production and bioavailability [20], seasonal lipid dynamics because of reproduction and build up of reserves in invertebrates [20,22] and seabird [64]. To our knowledge, however, it has never been studied over a complete-year cycle. Reasons for this include the high demands of fieldwork in the Arctic, where the polar night and extreme temperatures complicate the late-autumn and winter season, and inaccessibility of many marine organisms because of either partial or complete ice cover.

### Reproduction

Reproduction requires an energy investment by the mother, a large percentage of which is in the form of lipids. This provides a vehicle for the elimination of OCs, and for highly hydrophobic chemicals with long half-lives, this is often the dominant or even only relevant elimination route [65].

To our knowledge, no studies have directly assessed the influence of reproduction on OCs in arctic invertebrates, despite the fact that some arctic marine zooplankton live multiple years and reproduce more than once [50]. Many arctic marine zooplankton accumulate vast quantities of lipids (>50% of body dry wt) during the productive, open-water season [50,66]. These lipids are used for energy during the long, unproductive winter and for reproduction during the late winter or early spring, before onset of the phytoplankton bloom [49,57]. A recent study of OCs in the arctic marine copepod *Calanus hyperboreus* found that after reproduction, lipids increased over a four-month period, which subsequently influenced the concentrations of some OCs (e.g.,  $\Sigma$ HCH and  $\Sigma$ chlordanes) [22]. Marine zooplankton and ice fauna also transfer lipids to

their eggs. These are mostly phospholipids used as structural elements in cell membranes [57,67], however, and not neutral lipids, which are the storage lipids to which OCs have a higher affinity and a positive correlation [51]. Zooplankton maintain concentrations of most OCs that are near equilibrium with water concentrations, and this would suggest that the influence of reproduction on observed OC concentrations will be negligible.

Reproduction, at least regarding concentrations in males and females, has been shown to influence OC concentrations in fish. Studies have noted lower concentrations of OCs in females relative to males in individual populations of northern pike (*Esox lucius*) [68] and walleye (*Stizostedion vitreum*) [69]. In the walleye, both age and size at age were included in the statistical analysis. To our knowledge, however, no study has examined the influence of reproduction on observed OC levels in female arctic marine fish. The frequency of reproduction and the magnitude of the investment by the female likely will determine its significance regarding OC levels and bioaccumulation.

The influence of reproduction on seabird OC concentrations is dependent on the number of eggs produced by the female and is only significant for birds that produce a large number of high-lipid eggs [4]. Organochlorine concentrations were not found to differ between sexes in various arctic seabirds collected in northern Baffin Bay (North America) [70,71] and in the Barents Sea [72]. The sample size was low in both studies, however, and definitive conclusions cannot be made.

Reproduction significantly influences the OC concentrations observed in marine mammals, mainly resulting in decreased levels in reproductively active females compared with males [73–81]. The influence of reproduction on OC concentrations in marine mammals can be observed by examining the relationships between age/size and OC concentration (discussed below). Although production of the fetus provides a vehicle to eliminate OCs for the mother, the more important factor for marine mammals is the production of milk and its subsequent transfer to the neonate. The lipid content of seal milk is 40 to 60% [82], and that for whale milk is approximately 35%, much greater than the level in terrestrial mammals (e.g., human milk is only 4.5% lipid [83]) and providing an efficient means for lipophilic OCs to be eliminated in female marine mammals and transferred to their offspring.

Reproduction and lactation in the polar bear has been shown to result in increased concentrations of very hydrophobic OCs, such as PCBs, in the mother because of the loss of fat mass [65,84]. However, the overall body burden for these OCs decreases in the female bear during this time. This would suggest that although the OC concentrations mirror trends in lipid mass, the retention kinetics of fats and OCs in biota differ.

### Body size

Body size may be an important variable when considering different species or individuals within a population (e.g., different age classes). An organism's change in body size may influence OC bioaccumulation by altering process rates as a result of changed surface to volume ratio and, thereby, altering the elimination or metabolic rate; alternatively, the influence may result from shifting the level of OC exposure because of changes in the organism's behavior, such as diet preferences or habitat use (discussed below). For organisms having a direct exchange with water (i.e., invertebrates and fish), increased body size reduces the elimination of OCs via direct partitioning

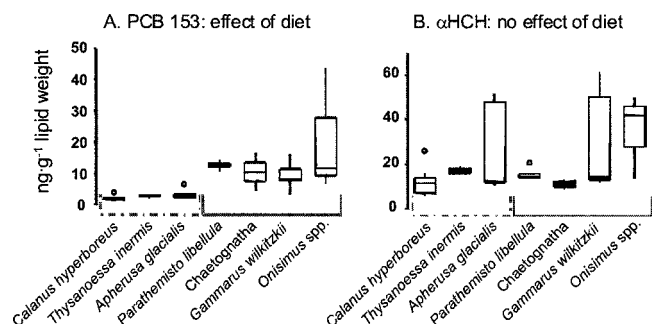


Fig. 4. Box plots of (A) polychlorinated biphenyl (PCB) 153 and (B)  $\alpha$ -hexachlorocyclohexane ( $\alpha$ -HCH) in predominantly herbivorous (---) and carnivorous (—) zooplankton and ice-associated amphipods from the marginal ice zone in the Greenland Sea and north of Svalbard in September/October 1999. (From Borgå et al. [109].)

to water because of a reduced surface to volume ratio and a lower rate of respiration [35,85]. Uptake rates of OCs with high lipophilicity and low water solubility, as inferred from  $\log K_{ow}$ , by direct diffusion also decrease with increasing body size and reduced surface to volume ratio [85]. However, the change in uptake rate with size is highly species dependent, as shown in a combined field and laboratory study of clams and oysters [86], and it likely is highly dependent on the physiology of the organism. As well, given the high lipid solubility of compounds and additional accumulation from dietary exposure, OC concentrations are expected to increase with size because of slower elimination.

The influence of size on OC bioaccumulation will be greater for more hydrophobic OCs ( $\log K_{ow} > 5$ ) because of slower partitioning out of lipids that results in longer half-lives and concentrations [37,87–89]. However, some high-molecular-weight, highly chlorinated OCs, such as octachlorodibenzo-*p*-dioxin and decachlorobiphenyl, have faster elimination rates than lighter and less chlorinated congeners in invertebrates and fishes, possibly because of limited partitioning into lipid pools and increased loss via fecal elimination [89,90]. For these highly chlorinated OCs, the influence of body size may be less important.

In arctic zooplankton, a comparison of OC concentrations in different size groups found no differences [19,20]. However, these zooplankton size groups consisted of a mixture of both herbivores and carnivores, and if trophic position influences bioaccumulation in aquatic invertebrates (see below), then the influence of the various factors is difficult to decipher. Marine zooplankton and ice fauna with similar diets but different body sizes had similar concentrations of hydrophobic OCs (Fig. 4A) [23], whereas for zooplankton species with different diets but similar sizes, carnivores had higher OC concentrations than herbivores [23–25] (Fig. 4A). In general, organisms sharing the same trophic position had similar OC concentrations regardless of their size. On the contrary, less hydrophobic compounds, such as HCH, did not show any difference among zooplankton and ice fauna that could be related to their size or diet (Fig. 4B). This corresponds to previous bioconcentration studies, suggesting that less hydrophobic OCs are not biomagnifying in marine invertebrates but are controlled by direct partitioning and equilibrium with water [20,22,91,92].

Limited information is available regarding OC levels in arctic marine fish with which to assess the influence of body size. Fish tend to be gap feeders (i.e., consume only what they can swallow), so larger fish tend to feed at a relatively higher

trophic level, which confounds efforts to assess the importance of size between species. In laboratory experiments, the elimination of OCs decreases with fish size [37,87]; therefore, if two fish species of similar size are feeding at the same trophic level, the higher trophic level species likely will have higher OC concentrations. Body size has been demonstrated to be positively related to OC concentrations in freshwater fish when examining different individuals within a population, but studies examining this phenomenon in arctic marine fish are limited. Johnston et al. [69] found that both gender and body size influence OC concentrations in freshwater walleye from temperate lakes. In the Greenland shark (*Somnios microcephalus*), collected in the Arctic, OC concentrations did not vary with size, although the unique feeding behavior of this shark and the limited size range studied may have confounded analysis of this relationship [93]. Despite the lack of data for arctic marine fish, OC concentrations likely increase with fish size for the same reasons as those discussed above.

Efforts to examine relationships between OC concentrations and body size in birds and mammals are confounded by the fact that once maturity is reached, birds and mammals do not continue to grow. The influence of body size on OC bioaccumulation in birds and mammals is best examined by comparing different species. Unfortunately, there are few examples of arctic seabird or marine mammal species that are of significantly different size but feed at the same trophic level, just as few species are available without some other characteristic (e.g., migration) that would confound comparisons. Ringed seals (*Phoca hispida*) and beluga whales (*Delphinapterus leucas*) offer a possible comparison, because they have been shown to feed at a similar trophic level based on stable isotopes of nitrogen [94]. On average, however, adult beluga are approximately 10-fold heavier than seals. Beluga also have approximately 10-fold higher levels of PCBs (in blubber) than do ringed seals from the same regions [95]. However, species differences in biotransformation capacity for PCBs between cetaceans and pinnipeds may account for some of this difference [96].

Trophic level and age more often overwhelm the influence of body size on observed OC concentrations in birds and marine mammals. This is exemplified by a comparison of OCs in the bowhead (*Balaena mysticetus*) and beluga whale. The bowhead, a baleen whale that feeds primarily on pelagic invertebrates, is many times larger and longer-lived than the beluga, a toothed whale that feeds on fish, but it has OC concentrations that are an order of magnitude lower [97].

Increasing size of an organism may result in dilution of the accumulated OC concentrations, called growth dilution (see, e.g., [12,13]). However, given the high influence of changed surface to volume ratio on elimination rates in water-respiring animals, growth dilution is more easily observed in growing, young, homeotherm species (i.e., birds and mammals). For instance, chicks and pups/cubs have initially high OC concentrations because of, respectively, parental transfer to the egg or via placenta and milk [74,98]. These initial OC concentrations and exposure via milk in offspring are most often much higher than what will be experienced by the young animal when it starts to feed. Therefore, the growing bird or weaned mammal is adding mass that is less contaminated compared with the milk or initial body burdens, and so the whole-body concentrations will decrease. Once the bird or mammal matures and stops growing, OC concentration may begin to

increase again with age until equilibrium is reached between OCs in the animal and in the diet.

#### Age, sex, and life cycle

Because OCs are slowly eliminated, continuous exposure to OCs during an organism's lifetime may lead to increased concentrations with age until an equilibrium is reached between the organism and the exposure routes. Often, this steady state is not reached for very hydrophobic, recalcitrant OCs, such as PCBs in arctic ringed seals [80]. As well, other factors related to age, such as increased size and trophic level, may obscure the impact of age.

Although many marine invertebrates live for several years, they have a shorter life span than fish, birds, and mammals. Because of the generally low OC levels and limited availability of samples, it is difficult to assess the influence of age on OCs in invertebrates. As well, other factors, such as size or behavior, appear to be more important in arctic marine invertebrates. For instance, OC concentrations in omnivorous, three- to five-year old ice amphipods were comparable to or lower than those in omnivorous, one- to three-year-old ice amphipods [24,25]. Similarly, herbivorous, three- to five-year-old copepods had OC concentrations similar to or lower than those of similar-sized, one- to two-year-old herbivorous ice amphipods [23].

Information regarding the influence of age on OC concentrations in marine fish is scarce; however, based on data from temperate freshwater species, age is likely to be a significant variable. For example, Larsson et al. [68] found that OC concentrations significantly increased with age in the freshwater eel (*Anguilla anguilla*). Age is correlated to body size in fish, however, so deciphering the impact of each variable individually is difficult. The relative importance of age and size on OC concentrations will undoubtedly vary with the characteristics of the species examined, such that the linear relationship between OC concentrations and age for slow-growing arctic fish will likely be much different and less steep (because of lower feeding) compared to faster-growing, temperate species.

A number of arctic seabirds are long-lived (>20 years), and an effect of age on OC concentrations is likely. Except for tagged birds, no methods to age adult seabirds are currently available; hence, it is difficult to evaluate the importance of age on OC bioaccumulation. A unique opportunity with banded thick-billed murres (*Uria lomvia*) allowed a comparison of OC concentrations between different age classes [99]. That study found that recalcitrant hydrophobic OCs (e.g., PCBs and DDT) were greater in older birds than in prebreeding birds, although the number of samples and age classes was limited. In correspondence with other studies on long-lived birds (e.g., [100]), a recent study of the long-lived adult glaucous gull (*Larus hyperboreus*) suggests that the seabird's OC concentrations reach steady state with the OC exposure from food relatively fast, at least before the age of first breeding [101]. Thus, the trophic level of the bird, rather than the age, is what determines the OC level.

Age has been shown to be a highly significant factor influencing OC concentrations in arctic marine mammals (Fig. 5). Results of recent studies are consistent with previously recorded trends showing that most OCs occur at lower levels in juveniles than in adults and are lower in adult females than in adult males [73–81]. Relative proportions of PCBs have also been shown to vary between juvenile and adult gray seals, but not between male and female adults [101]. Hydrophobic and recalcitrant OC concentrations generally increase with age

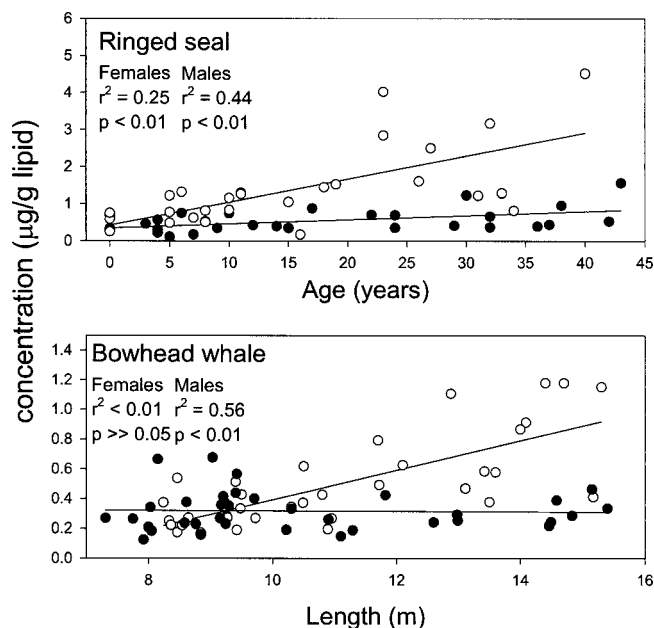


Fig. 5. First-order linear relationship between total polychlorinated biphenyl concentration in blubber and age in ringed seal from northern Baffin (derived from Fisk et al. [80]) and body length of bowhead whale from the Bering-Chukchi-Beaufort Sea stock (derived from Hoekstra et al. [81]). Male mammals (○) and females mammals (●) are represented.

in males, and they may either decrease, remain relatively constant, or increase with age in females, although in the latter case, generally at a slower rate than in males [80,81,102,103]. Stern et al. [74] found no increase in PCBs with age in male belugas and declining concentrations in reproductively active females. Lower OC concentrations in females normally are primarily attributed to the transfer of compounds to offspring during gestation and lactation and are discussed above. Whether females experience age-related changes in contaminant levels is variable and likely depends largely on their level of contaminant exposure, how often they successfully reproduce and wean offspring, and/or the level of lipid investment in offspring. Significant relationships between age and OC concentrations observed in female ringed seals from northern Baffin Bay likely result from the inclusion of very old female seals (>40 years) that have probably stopped reproducing [80]. Wolkers et al. [102] found that PCBs increased with age in both sexes of Svalbard ringed seals, and they suggested that in these seals, continued feeding by females during lactation may compensate for loss of OCs during lactation. Nonetheless, it is important to remember that age- and sex-related trends in contaminant concentrations may vary both within and between species depending on the compound being examined and the location of the study.

For OCs that are not recalcitrant or have lower hydrophobicity (e.g., total HCH isomers, or  $\Sigma$ HCH, and total tetra-, penta-, and hexachlorobenzenes, or  $\Sigma$ CBz), age has not been found to be a significant factor in the bioaccumulation of these OCs by marine mammals. For example, no sex-related differences were observed for  $\Sigma$ CBz and  $\Sigma$ HCHs in seals from northern Baffin Bay [80]. Similarly,  $\Sigma$ HCH and  $\Sigma$ CBz or hexachlorobenzene (HCB) levels were independent of age and sex in Alaskan (USA) bowheads and Barents Sea minke whales [76,81]. Male polar bears have 30% lower levels of chlordanes than female polar bears [104]. Polischuk et al. [65] showed

that male polar bears are capable of metabolizing chlordanes during a seasonal fast, whereas females were not, resulting in the lower levels of chlordanes observed in males.

#### *Habitat use and migration*

Habitat use is important for OC bioaccumulation in terms of changing the exposure, both by direct partitioning with water and by dietary uptake, and has been recognized by various food web models (see, e.g., [12,13,14,105]). Because of the high particle affinity of OCs and the benthopelagic coupling with sedimentation of organic matter and particles [61], higher OC concentrations are expected in benthic than in pelagic organisms of similar feeding ecology [106]. This is particularly true in the Arctic, where much of the primary productivity in the water column and marginal ice zone settles either ungrazed to the benthos or as fecal pellets from zooplankton [107,108].

Concentrations of OCs with high particle affinity (e.g., DDTs and highly chlorinated PCBs) have been found to be higher in benthic amphipods relative to zooplankton and ice fauna [19,106,109]. A similar pattern of greater concentrations in high-particle-affinity OCs in deep-sea fish compared to surface-water fish was explained by lower retention time of these OCs in the surface water and accumulation in the benthos [110,111]. Furthermore, differences exist among nonbenthic, water-respiring species depending on the vertical OC distribution in the water column [23]. Compounds with low particle affinity (e.g., HCHs) have a higher residence time in surface waters and diminishing concentrations with depth because of microbial degradation in the water column and possible input from the atmosphere [59,112–115]. Ice fauna have higher HCB,  $\gamma$ -, and  $\alpha$ -HCH concentrations than zooplankton, whereas other OCs (e.g., DDTs and PCBs) did not differ between ice fauna and zooplankton. Organochlorines with high particle affinity will sediment to the deeper waters and the seafloor, resulting in a relatively homogeneous vertical distribution of OC concentrations in the water column [112,113]. Thus, although the diet might not change in terms of feeding preference and prey composition, the direct exchange with water can influence the concentrations of OCs in water-respiring animals.

For higher-trophic-level organisms, such as seabirds and seals, a diet of ice fauna will increase the exposure to HCHs and HCB, whereas for compounds like DDTs and PCBs, the prey's trophic level will be more important than the habitat (sea ice vs pelagic) in determining the predator's exposure to contaminants [23]. Changes in dietary OC uptake with habitat use may be caused by the same diet being more (or less) contaminated, or by changing feed preferences, in the new habitat. For example, concentrations of OCs were greater in predatory gulls that resided in a colony near a seabird cliff and, hence, could prey on seabirds than in those that lived away from the seabird cliffs [101,116]. Although this is a result of different diets and is covered by the trophic level section of the present review, it does demonstrate that habitat selection influences OC levels and bioaccumulation pathways.

A number of arctic seabirds and marine mammals migrate, which can influence accumulated OC concentrations. For species that migrate from temperate regions, the arctic ecosystems likely are less contaminated, and OC concentrations in the migrating species likely are greater than expected in similar seabird species that are Arctic residents [21,29,70,71,117]. A good example of this is the black-legged kittiwake (*Rissa tridactyla*) in the eastern Canadian Arctic. This population mi-

grates from the eastern seaboard of North America, spending the summer in the Canadian Arctic. Concentrations of recalcitrant OCs, particularly PCBs, in this species are much higher than would be expected based on comparison with other seabird species feeding at a similar trophic level but remaining in arctic waters year-round [70,71,117].

Migrating species may also provide a means of delivering OCs to other arctic species. Harp seal migrations from the White Sea in northwestern Russia to eastern Svalbard and other areas of the northern Barents and Kara seas have been proposed as a pathway for the contamination of polar bears in the Barents Sea [118]. Using satellite-tagged polar bears, a study also showed that bears using sea ice in the Barents Sea had higher levels of OCs than those feeding mainly in near-shore areas, possibly because of higher caloric requirements and differing diets (harp seals vs ringed seals on landfast ice) [119].

Subtle changes of OC profiles have also been observed to be associated with habitat or migration changes within populations of marine species. Multivariate statistical analyses (i.e., direct and indirect ordination) have been used to give a better understanding of OC profiles in marine mammals resulting from potential regional and seasonal differences in accumulation [73,95,120,121]. For example, Weis and Muir [122] noted increasing proportions of more volatile OCs in ringed seal blubber with increasing latitude in the Canadian Arctic, whereas Muir et al. [103] found increasing proportions of HCH and decreasing PCBs with western longitude in ringed seals from Canada, Greenland, Svalbard, and Russia. Relative proportions of PCB congeners were not found to vary between adult gray seals (*Halichoerus grypus*) across wide geographic areas [101]. Hobbs et al. [120] suggested that geographic variation in OC patterns in minke whale (*Balaenoptera acutorostrata*) blubber in the North Atlantic are caused by regional differences in exposure. The ordination plots of OC concentrations in bowhead whale blubber samples suggest that proportions of OCs, such as  $\beta$ -HCH, fluctuate with the seasonal migration of this population between the Bering, Chukchi, and Beaufort Seas [81]. Ultimately, many of these observations likely are the result of different OC concentrations in seawater among regions within the Arctic Ocean [123,124], although accurate measurements for OCs other than HCH isomers in seawater are very limited.

#### *Biotransformation*

As mentioned, the elimination rate for hydrophobic OCs is more influential than uptake rate in determining concentrations in biota. Biotransformation usually contributes to the elimination of a chemical, and if an OC is biotransformed to a more polar compound, which is readily excreted, the metabolite will not biomagnify. For OCs, enzyme-mediated biotransformation is dependent on both the chemical structure and the metabolic capacity of the organism, typically mediated via cytochrome P450 monooxygenases (CYP). This diverse class of enzymes exhibits broad but overlapping substrate and product specificities that may be species- and gender-specific (see, e.g., [125]).

Assessing biotransformation of OCs is difficult, and few studies have reported biotransformation rates, which are required for mechanistic modeling. Most of our current understanding about biotransformation in wild marine organisms results from comparison of the relative abundance of individual OCs between species or groups of organisms (e.g., birds vs fish) (see, e.g., [42]) or using *in vivo* techniques, such as liver microsome studies [126]. Based on relative abundances of



individual OCs, biotransformation capacity was estimated to increase from invertebrates to fish to seabirds and marine mammals [41]. However, this varies significantly depending on the OC and the species. For example,  $\alpha$ -HCH biomagnifies in ringed seals, and evidence suggests that these organisms cannot biotransform this OC [127,128]. However,  $\alpha$ -HCH does not biomagnify in arctic seabirds, because they readily biotransform and eliminate this isomer [127]. As well, seabirds and marine mammals are efficient at CYP1A-type biotransformation (e.g., non- and mono-*ortho* PCBs with adjacent hydrogen at the *ortho*- and *meta*-positions). However, seabirds and seals have limited CYP2B-type biotransformation (e.g., PCBs with two or more chlorines at the *ortho* position and with adjacent hydrogen at the *meta*- and *para*-positions) [41], whereas polar bears are proficient at CYP2B transformations [17]. The limited data regarding CYP2B-like biotransformation of nonplanar contaminants in cetaceans, including *ortho-meta* and *meta-para* unsubstituted PCB congeners, is relatively low compared to data regarding other marine mammals, such as pinnipeds and polar bears (see, e.g., [129,130]). Recent studies concerning the formation of PCB metabolites, and changes in PCB congener patterns relative to prey, suggest that whereas CYP2B-like gene expression and subsequent isozyme activity appear to be limited in cetaceans, as-yet-uncharacterized CYP isozymes with sufficient catalytic activity may be present and capable of mediating the biotransformation of several PCB congeners [131,132].

Arctic marine zooplankton and benthic invertebrates do not appear to efficiently biotransform OCs, and concentrations and relative abundances of OCs in these organisms are mainly determined by exposure from water and diet and by direct elimination to water and fecal matter [23,25,92]. Based on laboratory studies, evidence suggest that both freshwater and marine fish can biotransform some OCs (see, e.g., [133,134]).

Biotransformation is most significant for seabird and marine mammals and can have a substantial influence on observed levels and patterns of OCs in these animals. This was demonstrated for OCs in the arctic marine food web using arctic cod, ringed seal, and polar bears [16]. The number of OCs decreased from cod to seal to bear, because less recalcitrant chemicals did not biomagnify. In fact, biotransformation has a profound impact on the patterns of OCs observed in polar bears [17]. Concentrations of OCs like *p,p'*-dichlorodiphenyldichloroethylene (DDE), which are recalcitrant in most arctic biota, are low in polar bears, because these animals can metabolize this OC [17].

The biotransformation of OCs by CYP enzymes is not always a beneficial process. In many cases, the resulting metabolites are more bioaccumulative than the parent compounds. For example, DDE and heptachlor *exo*-epoxide have TMFs that were much higher than expected based on their  $\log K_{ow}$  values, likely because an organism can both bioaccumulate these chemicals and form them from other contaminants [21]. Another example is methyl sulfone (MeSO<sub>2</sub>)-containing PCB and DDE metabolites in the ringed seal–polar bear food chain [135]. Some of the MeSO<sub>2</sub>-PCBs quantified in polar bears are present because of biotransformation of precursor PCB congeners, but a significant proportion of this class of PCB metabolites in this species likely is the result of bioaccumulation from ringed seals.

Chiral pollutants, which exist as two mirror-image forms called enantiomers, have also been used recently to assess relative biotransformation capacity between species

[21,127,128,136]. Enantiomers have identical physicochemical properties and abiotic degradation rates, but they can also have different rates of biotransformation, resulting in a change in the relative proportions of each enantiomer (enantiomeric fractions [EFs]) [137]. The quantification of EFs provides additional information regarding the biotransformation capacity of biota that would have been overlooked by conventional, achiral contaminant analysis. For example, the stereochemical disposition of chiral OCs appears to vary significant between seabird and marine mammal species, suggesting that the biotransformation process is stereoselective and, possibly, species-dependent. Proportions of chlordane components in seabirds were shown to be related to taxonomy based on the magnitude of EF values of chiral components and relative proportions of all components [70]. In particular, thick-billed murre (*Uria algae*) had a superior ability to biotransform and eliminate chlordane based on high proportions of oxychlordane and the highest EFs for oxychlordane and heptachlor epoxide among seven species of seabirds. The  $\alpha$ -HCH EF values varied between invertebrate species, although the chemical was found to biomagnify in these organisms [127]. However, differences in EFs may also be attributed to different exposure, depending on where in the water column the invertebrates and fish are living, and not to biotransformation, because the EF of  $\alpha$ -HCH may vary with depth [59]. The idea that exposure rather than biotransformation is important for invertebrates and fish is supported by the similarity in EFs between invertebrates, arctic cod, and seawater [127,128].

#### Feeding ecology and trophic position

The relative importance of water versus food regarding OC exposure in aquatic organisms continues to be a topic of debate in ecotoxicology (see, e.g., [138–140]). Although the accumulation of OCs from water is rapid and BCFs (Eqn. 2) are orders of magnitude greater than BMFs (Eqn. 1), a significant amount of the exposure to most hydrophobic OCs for fish and, of course, birds and mammals would be through diet. For example, it is estimated that more than 99% of PCBs in lake trout (*Salvelinus namaycush*) are derived from dietary exposure [141]. Because much of the OCs in oligotrophic fresh and marine waters will be freely dissolved [142], a very large increase in concentration occurs between water and the first trophic level (i.e., phytoplankton) as a result of bioconcentration. A second-trophic-level organism will, on consuming a first-trophic-level organism, be exposed to a highly concentrated source of the contaminant compared to water. Because energy obtained from food is also used for purposes other than building biomass, the biomass per trophic level decreases when moving up the food web (generally quoted as 10%, but can vary from 3–13% [143]). However, many OCs are not readily eliminated with each step in the food web, or at least not to the extent of biomass dilution. Thus, with each step in the food web, the discrepancy between the concentrations in food and water increases.

To illustrate the discrepancy between exposure to contaminants through water and through food, an estimate of 1,2,3,4-tetra-CBz and 2,2',4,4',5,5'-HCB (PCB 153) uptake by arctic cod (*Boreogadus saida*) from water and from herbivorous copepods (*C. hyperboreus*) from northern Baffin Bay is presented (Table 1). The trophic levels of *C. hyperboreus* and arctic cod have been established as 2 and 2.9, respectively, based on  $\delta^{15}\text{N}$  values and stomach contents [144]. *Calanus hyperboreus* were also found in the stomach of these arctic

Table 1. Estimate of exposure to 1,2,3,4-tetrachlorobenzene (CBz) and 2,2',4,4',5,5'-HCB (PCB 153) by arctic cod via water and a common food item (*Calanus hyperboreus*)<sup>a</sup>

	1, 2, 3, 4-tetra-CBz	PCB 153
Log $K_{ow}$ <sup>b</sup>	0.000 007	0.000 006
Water concentration (ng/ml)	0.167	1.368
Maximum exposure rate of arctic cod through water (ng/d) <sup>c</sup>	0.073	0.062
Maximum exposure rate of arctic cod through food (ng/d) <sup>d</sup>	0.054	0.438
Percentage of total exposure to chemical via food	42.5	87.6

<sup>a</sup> Data for water and *C. hyperboreus* adapted from Fisk et al. [22].

<sup>b</sup> Log  $K_{ow}$  of 1, 2, 3, 4-tetra-CIBz is from Mackey et al. [33] and for PCB 153 from Hawker and Connel [38].

<sup>c</sup> Maximum exposure rate of arctic cod through water = [water] × VR, where [water] is the water concentration and VR is the ventilation rate of the arctic cod. The ventilation rate is the water volume pumped per day ( $\text{ml d}^{-1}$ ) derived from the rate determined for a 100-g striped mullet (*Mugil cephalus*) [184] in ambient conditions ( $36 \text{ ml min}^{-1}$ ) scaled to the weight of the arctic cod (21.5 g) by multiplying mullet weight/arctic cod weight.

<sup>d</sup> Maximum exposure rate of arctic cod through food = [*C. hyperboreus*] × FC, where FC is the amount of food consumed per day based on a feeding rate of 1.5% body weight  $\text{d}^{-1}$  and a weight of 21.5 g.

cod, so it is a legitimate predator–prey comparison. Concentrations of 1,2,3,4-tetra-CBz, a relatively water-soluble OC, and PCB 153, which has very low water solubility, are much higher in *C. hyperboreus* than in water (Table 1). If some reasonable assumptions about ventilation volumes and feeding rates of arctic cod are made, the exposure via food is significant for 1,2,3,4-tetra-CBz and the dominant route for PCB 153 (Table 1). Dietary assimilation of CBz and PCBs has been estimated to range from approximately 50 to 75% for small fish [37] and may be as high as 90% [145]. Therefore, food exposure is the more significant factor—even if exposure is adjusted for assimilation efficiency. Arctic cod also consume more contaminated prey, such as predatory amphipods, and not all of the contaminant that passes over the gills is absorbed. Thus, food likely plays a larger role than is suggested by this example.

Evidence is mounting that dietary accumulation is an important process for marine zooplankton [22,23–25,92,146]. However, it has been stated that even if dietary uptake is important, the direct partitioning with water is sufficiently fast that biomagnification will not occur in zooplankton but, instead, that the organisms will always remain in equilibrium with water and OC concentrations will be explained solely by the organism's lipid (see, e.g., [138]). When adjusting for lipid differences between zooplankton, however, marked differences remain in OC concentration between species. Although some of this variation may be attributed to factors such as size or habitat, as described above, a close comparison of species-specific samples of zooplankton of different sizes, diets, and habitat suggest that a large proportion of the variation in hydrophobic OCs between zooplankton can be attributed to their diets [23] (Fig. 4). Concentration and pattern differences among zooplankton reflect their diets, as also shown for higher trophic levels [23]. However, OC bioaccumulation from diet by invertebrates and other poikilotherms (and, consequently, biomagnification) is not as great as in homeotherms. This re-

sults from a number of factors, mainly that homeotherms have much greater feeding rates than poikilotherms [21,46,147].

Various studies have demonstrated that diet choice and trophic level play a dominant role in recalcitrant OC concentrations in seabirds and marine mammals. For example, OC concentrations have been shown to be correlated to trophic level in seabirds [29,70,71]. Muir et al. [148] showed how walrus (*Odobenus rosmarus*) that fed on ringed seals had much higher OC concentrations than those that fed only on benthic crustaceans. Other examples of the importance of diet choice in OC concentrations have been given throughout the present overview.

## QUANTIFICATION OF OC TROPHIC TRANSFER

### Defining food webs

The determination of OC bioaccumulation and subsequent transfer within a complex assemblage of biota (i.e., food web) implies a rudimentary understanding of how these organisms interact. The concept of trophic dynamics was first discussed by Lindeman [149], and the literature regarding food chains and food web structure has become quite substantial. The usefulness and value of defining food web structure is an ongoing debate in ecology (see, e.g., [150]). Regardless, the organization of ecosystems into trophic levels provides a simplistic and practical way to conceptualize energy and contaminant flow within an ecosystem as well as predator–prey interactions. However, these classifications are subject to various pitfalls [151]. One such criticism is that trophic interactions are dependent on direct observations of feeding behavior or indirect empirical measurements, such as gut contents, that may not account for all linkages or may overestimate the importance of a trophic link. As a result, a continuous variable to represent assimilated diets is required.

Over the past decade, improvements in analytical techniques have permitted the use of naturally occurring, stable isotopes of elements in a broad array of ecological applications [152]. The application of stable isotopes to better understand ecological linkages has advantages over traditional methods, such as analysis of gut contents, because it averages dietary assimilation over a longer period of time and does not suffer from subjective interpretations of contents [153]. This approach is based on the principle that ratios of stable isotopes, mainly carbon ( $^{13}\text{C}/^{12}\text{C}$ ;  $\delta^{13}\text{C}$ ) and nitrogen ( $^{15}\text{N}/^{14}\text{N}$ ;  $\delta^{15}\text{N}$ ) in consumer tissues can be related in a predictive way to those in their diet. The ratio of heavier stable to lighter stable isotopes of nitrogen ( $^{15}\text{N}/^{14}\text{N}$ ), expressed relative to a standard as  $\delta^{15}\text{N}$ , generally increases with trophic position in aquatic food webs, providing a continuous variable with which to assess trophic status [154]. Stable carbon isotope ratios ( $^{13}\text{C}/^{12}\text{C}$ ) show less enrichment with trophic transfer but can be useful in evaluating sources of primary production in marine systems as well as general patterns of inshore/benthic versus offshore/pelagic feeding preferences [94,155]. In addition to providing novel insights regarding feeding dynamics and migration, stable isotopes can also quantify the influence of trophic status on the transfer of environmental pollutants between biota. The potential for stable isotopes, most notably  $\delta^{15}\text{N}$ , to be used in biomagnification studies was first demonstrated by Broman et al. [156]. Since then,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  signatures in biota have been used to interpret the trophic transfer of persistent anthropogenic contaminants in several freshwater [157–160] and marine systems [21,26,27,46,161].

Table 2. Trophic magnification factors (TMFs)<sup>a</sup> for selected persistent organic pollutants reported for marine food webs based on the magnification power defined by Broman et al. [156] for polychlorinated dibenyl-*p*-dioxins and polychlorinated dibenzo furons for the Baltic marine food web

	Arctic marine											BMF			
	Alaskan Arctic	Canadian Arctic				European Arctic			Russian Arctic	Other marine		Fresh-water	Harp seal		
	Beaufort-Chukchi Seas (A)	Northern Baffin Bay (B)		Barents Sea (C)		White Sea (D)	Norway Oslofjorden (E)		BMF <sup>h</sup>	BMF <sup>i</sup>	BMF <sup>j</sup> TL <sup>j</sup>				
	Whole <sup>b</sup>	Whole <sup>b</sup>	Whole <sup>c</sup>	Cold <sup>c</sup>	Cold <sup>d</sup>	Warm <sup>d</sup>	Whole <sup>b</sup>	Whole <sup>e</sup>				Whole <sup>f</sup>	Cold <sup>f</sup>	Cold <sup>g</sup>	
$\alpha$ -HCH	1.6	2.2			<i>1.6</i>	<i>1.6</i>	1.5	1.2				5.3	2.0	2.7	
$\beta$ -HCH	2.9	4.2	7.2		1.8	8.1	1.5	0.6				4.3	3.0	4.1	
$\gamma$ -HCH	0.7	1.4					1.7	0.6				1.3	5.0	6.8	
$\Sigma$ HCH	1.4		2.7												
HCB	1.4	1.8	4.1		4.7	4.7	1.8	2.3				3.6	6.0	7.3	
<i>cis</i> -Chlordane	0.7	1.7	1.6		2.6	59.6		0.7				0.2			
<i>trans</i> -Chlordane	1.0											0.2			
Oxychlordane	9.3		6.5		4.4	17.5		9.6				10.6	141.0	180.8	
<i>trans</i> -Nonachlor	5.2	5.1	5.5		5.0	5.0	10.4	4.8			4.4	8.2	111.0	141.7	
$\Sigma$ Chlordanes	4.8		6.6						6.8			4.5			
<i>p,p'</i> -DDE	5.4	7.4	13.7	5.5	3.7	31.8	14.5	3.6			4.1	21.3	250.0	319.3	
<i>p,p'</i> -DDT	1.5	2.8	5.7					1.3				5.4			
$\Sigma$ DDT	3.0		10.8						8.9	4.8		11.0			
PCB 28	1.3	1.3	2.1		5.4	5.4	3.3	1.3				0.5			
PCB 31					3.5	3.5									
PCB 47			2.5		2.8	14.5							8.0	10.4	
PCB 52	2.6							1.9			2.2	4.1			
PCB 99	5.9	5.5	7.5		3.1	28.4	11.9	2.6				19.7	115.0	147.0	
PCB 101	3.9		3.6									5.7			
PCB 105	5.8		6.1		3.4	21.4		1.4				6.2	14.0	18.1	
PCB 118	3.8		3.7		3.9	26.2		1.5				9.7	33.0	41.6	
PCB 149			2.3		3.4	14.3						2.5	1.0	327.7	
PCB 153	6.7	5.9	9.7		4.1	26.3	18.8	2.9			3.7	22.1	325.0	416.0	
PCB 138	4.7	5.6	8.8	6.7	3.7	27.8	17.0	3.4				22.1	256.0	327.7	
PCB 180	6.5		10.7					0.8				17.8			
PCB 194								2.2							
$\Sigma$ PCB 10								2.2							
$\Sigma$ PCBs	3.3	2.9	4.6					1.5	6.2	3.9					

<sup>a</sup> Significant TMFs ( $r^2 > 0.05$  or  $p < 0.05$ ) are in italics. Standard biomagnification factors (BMFs) for mixed prey (Eqn. 3) and adjusted to exactly one trophic level ( $<$  Eqn. 4) for harp seals in the Barents Sea are shown. Empty spaces are not reported in the respective studies. DDE = dichlorodiphenyldichloroethylene; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane; PCB = polychlorinated biphenyl.

<sup>b</sup> Hoekstra et al. [26]. Invertebrates, fish, and marine mammals in one relationship to estimate TMFs. Significance level is not given for TMFs for the Barents Sea and the Northern Baffin Bay food web relationship.

<sup>c</sup> Fisk et al. [21]. Whole: Invertebrates, fish, seabirds, and marine mammals in one relationship; Cold: relationship with only invertebrates and fish.

<sup>d</sup> Hop et al. [46]. Cold: Relationship with only cold-blooded species (invertebrates and fish); Warm: Relationship with only warm-blooded species (seabirds and seals).

<sup>e</sup> Muir et al. [27]; invertebrates, fish, and seals in one relationship to estimate TMFs.

<sup>f</sup> Ruus et al. [161]. Invertebrates, fish, seabirds and seals are in one relationship to estimate TMFs. Values are reported for individual compounds, only sums. Hexachlorocyclohexanes and HCB did not correlate with trophic level and were not reported.

<sup>g</sup> Kidd et al. [167]; algae, invertebrates, and fish in one relationship to estimate TMFs

<sup>h</sup> Fraser et al. [164]; predator and prey from the central Barents Sea in 1997.

<sup>i</sup> Borgå et al. [29], Kleivane et al. [118], and Hop et al. [46]; predator and prey from central Barents Sea in June 1995.

<sup>j</sup> Hop et al. [46]; predator and prey from central Barents Sea in June 1995.

### Quantifying biomagnification

Variation in OC accumulation in food webs is often illustrated by magnification factors that describe the increase in concentration between two adjacent trophic levels in a food web [5,162]. Commonly, BMFs are calculated by dividing the lipid-adjusted OC concentration in the predator by that in its prey (Eqn. 1). Alternatively, BMFs can be adjusted to represent exactly one trophic level in difference using the trophic level estimated from  $\delta^{15}\text{N}$  [21] (BMF–trophic level [BMF-TL], Eqn. 5). Commonly, BMFs are calculated relative to one prey species, not to the mixture of species or functional groups that are actually fed on by a predator (see, e.g., [29,30,163]). Although one could argue that considering a mixed diet is un-

important, OC concentrations in prey likely are relatively low compared to those in the predator, so the use of ratios (BMF) makes the variation evident and results in magnification factors that differ depending on the prey [28,93]. As a result of a fluctuating and variable environment with respect to food availability, many arctic marine predators are omnivorous opportunists rather than specialist feeders. Therefore, using a diet of mixed prey items is recommended if calculating BMFs. However, even when considering mixed prey, the BMFs for a predator may vary substantially depending on the study (Table 2). For instance, BMFs varied widely for harp seals (*Phoca groenlandica*) when compared between two studies from the Barents Sea in the mid-1990s [46,164] (Table 2).

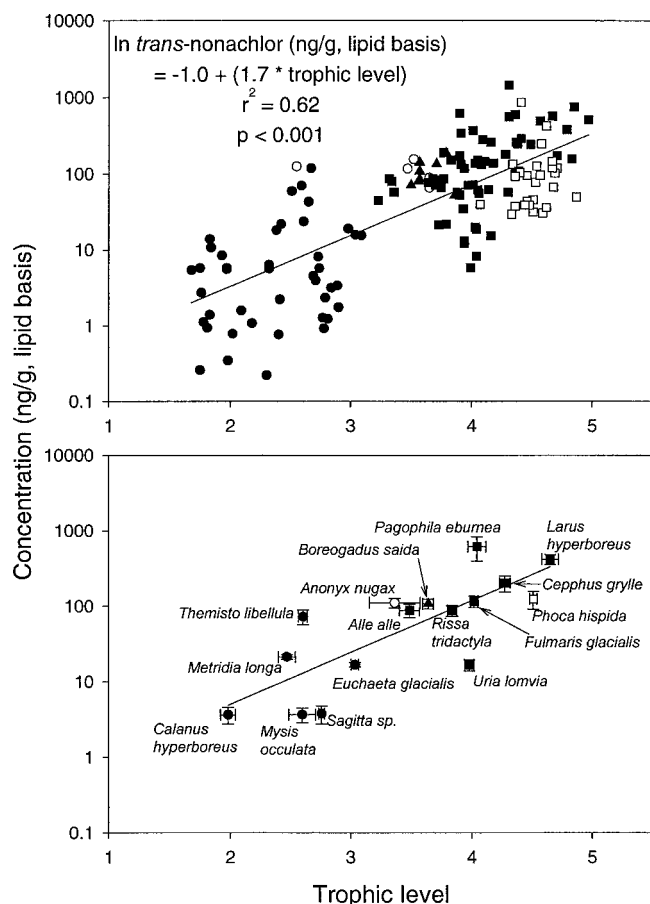


Fig. 6. Relationship between concentrations (lipid wt) of *trans*-nonachlor and trophic level (as defined by  $\delta^{15}\text{N}$ ) in the marine food webs of northern Baffin Bay (derived from Fisk et al. [21]). The top graph contains all data points, and the bottom graph contains the mean ( $\pm 1$  standard error) values for each species. Lines are linear regression. Tissues analyzed for *trans*-nonachlor were as follows: Invertebrates and arctic cod, whole body; seabirds, livers; and ringed seals, blubber. Symbols: ● = pelagic zooplankton; ○ = benthic amphipods; ▲ = arctic cod; □ = ringed seals; ■ = seabirds.

If the investigated prey items reflect the actual diet of the predator, then BMF-TLs and BMFs should be equal, because these ratios assume steady-state OC concentrations between predator and prey. Large differences between BMF and BMF-TL may indicate that predators have sources of contaminants other than the investigated prey and are a potential ecological tool [93]. The BMF-TL is calculated as

$$\text{BMF-TL} = \frac{(\text{BMF}_{\text{lipid weight}})}{(\text{Trophic level}_{\text{organism}} - \text{Trophic level}_{\text{diet}})} \quad (5)$$

Trophic magnification factors have been used to describe the increase of OCs from one trophic level to the other (Fig. 6). The TMFs are derived from the slope ( $b$ ) of the relationship between an organism's log lipid-normalized OC concentrations ( $[\text{POP}_{\text{organism}}/lw]$ ) and trophic position (TL) as determined by stable nitrogen isotopes [21,26,27,46,156,161,165] and as shown by Equation 6:

$$\text{Log} [\text{POP}_{\text{organism}}/lw] = a + b\text{TL} + e \quad (6)$$

where  $a$  is the intercept,  $l$  is the error estimate, and TMF is  $10^b$ .

This linear regression approach provides an average rate of increase per trophic level based on the assumptions that both

biomass transfer and biotransformation efficiencies are similar for all the organisms included in the regression analysis [156,165]. Thus, because poikilotherms and homeotherms have different energy requirements and biotransformation abilities (see, e.g., [41,166]), an organism's physiology should be identified (by a dummy variable) when the relationship between OC concentrations and trophic position is analyzed [46]. This will allow the observed, higher biomagnification in homeotherms than in poikilotherms as visualized by the regular BMFs, to be reflected. The use of a single biomagnification relationship for the whole food chain will overestimate and underestimate the degree of biomagnification in poikilotherms and in homeotherms, respectively [21]. The TMFs also assume diet as the main OC exposure route, although as noted previously, direct partitioning is important in the uptake of more water-soluble OCs, such as HCHs, in water-respiring animals.

#### Comparison of TMFs between studies

Although not without limitations, TMFs may be advantageous over BMFs and BMF-TLs when making comparisons between studies and ecosystems in the degree of biomagnification of an OC. The TMFs represent the average increase in OC concentration in food webs rather than the variability shown between species and compounds in BMF and BMF-TL ratios, which represent only specific predators. In addition, variability in food web magnification because of different OC exposure at the base of the food web (e.g., between different lakes) or between ice (pelagic or benthic) is accounted for in the TMFs (by the intercept in the regression), so the rate of increase per trophic level can be studied independently [156].

A recent study summarized TMFs for arctic marine food webs from the Alaskan, Canadian, European, and Russian arctic and temperate regions [26] (Table 2). Those investigators found that whereas the TMFs calculated for food webs (invertebrates, fish, and marine mammals) from the Alaskan Arctic, the Canadian Arctic, and the temperate regions were similar, results were different from TMFs for the Barents and White Seas (European and Russian Arctic). This indicates that global spatial variability of OC concentrations in higher-trophic-level arctic marine predators [123] may not only be caused by regional differences in contamination but by regional differences in uncharacterized trophic interactions [26]. However, compounds that have similar TMFs for poikilotherm and homeotherm species, such as *trans*-nonachlor (Table 2), also had TMFs that were similar between all food webs. In addition, when comparing only estimates for cold-blooded species, TMFs are comparable among the arctic marine food webs [21,46], a temperate marine food web [165] and a temperate freshwater food web [167]. Because TMFs for individual OCs that could be compared were generally similar between different food webs (both temperate and arctic marine as well as freshwater), this suggests that the process of biomagnification is similar between ecosystems even though characteristics of the food webs, such as lipid dynamics, physical environments, and average annual temperatures, differ widely. However, as this review shows, few datasets are available for marine temperate and freshwater food webs with which to do a proper comparison (Table 2).

#### MODELS AS USEFUL TOOLS IN SINGLE-SPECIES AND FOOD WEB STUDIES

##### Different models, common aim

As the sections above illustrate, many factors contribute to variation in OC concentrations among different organisms. The

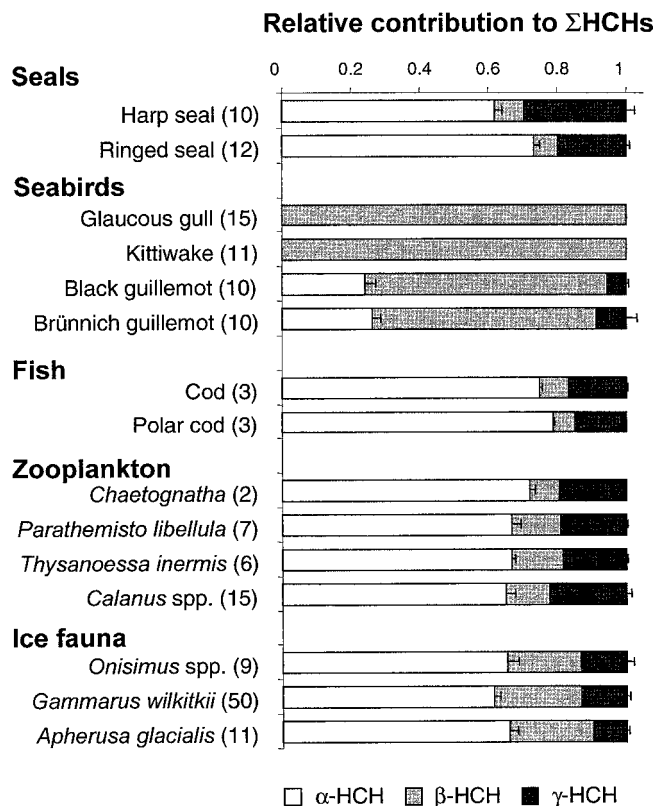


Fig. 7. Changes in hexachlorocyclohexane (HCH) pattern (relative contribution + standard error of each isomer to  $\Sigma$ HCH) in the arctic marine food web. Summary of published and unpublished invertebrate and fish data from the Barents Sea, north of Svalbard, and Greenland Sea in 1995, 1998, and 1999 and seabird and mammal data from the Barents Sea in 1995 (derived from Borgå [109]). The pooling of the invertebrate data is appropriate given the similar pattern across area, year, and season (derived from Borgå et al. [23–25,29]). Numbers in brackets are the sample size.

aim of the present section is not to give a comprehensive review of existing bioaccumulation models; this can be found in the detailed reviews by Gobas and Morrison [5] and by Mackay and Fraser [9]. Rather, the aim is to discuss the use of bioaccumulation models with arctic marine food webs. Over the last few decades, several mechanistic models have been developed to describe the OC bioaccumulation for either a single aquatic species or aquatic food webs. These include models principally based on chemical diffusion (see, e.g., [11–14,35,141]) and bioenergetics (see, e.g., [10,168–170]). The chemical exchange and bioenergetics-based models have been the conceptual basis of other models and/or have been used directly to investigate different aspects of food web contaminant flux (see, e.g., [105,111,157,164,171–174]). Rather than treating the organism as one box, as in the general food web approach, pharmacokinetics/physiological-based models are concerned with the distribution of contaminants within an organism, such as in seabirds using the bioenergetics approach (see, e.g., [175]) and in marine mammals using the chemical exchange approach (see, e.g., [172]).

Common for all mechanistic models, however, are the aims of simplifying the phenomenon of interest to understand the most important underlying processes and the aim of predicting behavior or outcome given the specified phenomenon. Both aims (simplification and prediction) will be discussed below in the context of the arctic marine ecosystem and the factors

shown to be important in the OC bioaccumulation in single species and/or on a food web basis.

#### *Simplification to understand importance of processes involved*

Although simplification of processes reduces the realism of a model (which is also the difference between laboratory experiments and field observation studies), it is important to isolate the processes to understand their relative importance. Simplification and isolation of processes is also the approach in statistical and empirical models, as exemplified in the present summary by results from the use of analysis of variance (see, e.g., *Body size*), principal component analysis (see, e.g., *Habitat use*), and linear regression (see, e.g., *Age, sex, and life cycle* and *Quantifying biomagnification*).

The simplification of the phenomenon may be in terms of choosing and reducing the processes included, the mathematical solution of the processes, and the parameterization. For example, in arctic marine systems, simplifications could include the following: Assuming negligible biotransformation, limiting temporal changes in lipid and feeding rates, and assuming steady-state contaminant concentrations. Assuming negligible biotransformation may be valid for recalcitrant PCBs (e.g., 2,2',4,4',5,5'-substituted PCB 153) but not for others (e.g., HCH). Figure 7 illustrates the varying proportions of HCH isomers in arctic marine food web organisms because of the biotransformation and elimination of the  $\alpha$ - and  $\gamma$ -isomers and retention of recalcitrant  $\beta$ -HCH. Temporal changes in food availability at the base of the food web may be substantial and important to consider (as illustrated for primary production and zooplankton lipid content in Fig. 3). Effects of sex and age as well as their influence on contaminant levels within the population of organisms are important, and assumptions of steady-state concentrations will introduce high uncertainty (as illustrated for seals and whales in Fig. 5).

All models aim at describing a phenomenon as simply as possible, but the study of food webs involves several different processes given the high complexity of ecosystems. Thus, the predictors included in statistical, empirical, and mechanistic models are many. Models that are too complex, however, reduce their usability, both in terms of user-friendliness and by increasing the uncertainty of model predictions, and they will not necessarily contribute to increased understanding of OC bioaccumulation [176,177]. By reducing the predictors, the whole process of modeling will be more practical, less costly, and less demanding; however, the generality of the model will decrease. An example of the importance of including more detailed properties of biota was illustrated by Jackson [178]. That study compared the use of constant carbon assimilation efficiency versus a bioenergetics approach to describe allocation to various metabolic processes and the effect on PCB concentrations in growing Lake Ontario fish. The comparison resulted in a 450% difference in predicted PCB concentration when lake trout growth rates were reduced by 60%. Thus, given that the bioenergetics approach accounts for the difference in carbon allocation with changed process rates, its use is beneficial in models considering trophic interactions, nutrient cycling, and contaminants [178]. Chemical diffusion models have also been developed, starting with relatively simple exchange models (see, e.g., [179]), that include bioenergetics solutions to the parameter estimates (see, e.g., [12,13]).

### *Predictability of models: Influence of simplification and assumptions made*

A model's output must be compared with empirical data to evaluate its realism and predictive ability. The simplification done by selection of processes, their equations, and scaling values assumes a certain behavior that might affect the model's predictive strength. Not only mechanistic but also empirical models and BAFs suffer from assumptions that might not be fulfilled in all situations, which in turn might reduce the predictive power of a model. For instance, BCFs, BMFs, their correlation with  $\log K_{ow}$ , and the existing food web models usually assume equilibrium between OC concentrations in the organisms and that of the environment, either in the diet or in the water. By using different food web models based on slightly different concepts [12,13] and a Lake Ontario dataset [15], Burkhard [7] obtained similar and satisfactory results from the different models in terms of predictability. Good correspondence was also seen between model outcome of the Gobas [13] and the Campfens and Mackay [14] models [180]. This illustrates the robustness and the sufficient predictability of some existing single-species or food web models for selected datasets. However, the assumption of equilibrium may not be valid in a highly fluctuating situation, such as the arctic ecosystem, and therefore may not be applicable in terms of mirroring the OC dynamics at the base of the food web. Equilibrium seems to occur between OCs in water and arctic zooplankton during low-productivity months (winter) [20,22]. On the other hand, during high-productivity months (summer), when the ecosystem undergoes rapid changes in terms of primary production, OC concentration in water, and organism lipid content, equilibrium may not be reached and thus should not be used to describe the current processes [20,22]. Skoglund et al. [48] noted this effect for PCB sorption to phytoplankton and were able to predict the effect more closely using a kinetics rather than equilibrium modeling approach.

Another important issue is the use of existing models in environments that have not been extensively studied, such as the Arctic Ocean, where few data were previously available and few attempts have been made to model the OC behavior in single species or the food web (but see, e.g., [164]). Most single-species and food web models are validated for temperate lakes. Validation for arctic marine food webs will be interesting, because these organisms have very specific adaptations to the highly fluctuating environment and primary production (Fig. 3). Thus, the influence of factors relevant in the Arctic, such as special lipid dynamics, open marine environment (and not a closed lake), and different temperature regime, are all potentially interesting to study with mechanistic models. Another special feature of the arctic marine ecosystems is that as a result of the lipid-rich, lower trophic levels, they sustain large populations of various species of seabirds and marine mammals. Given that most existing aquatic food web models include fish as the highest trophic level, it will be valuable to extend these models to birds and mammals by linking bioenergetics or fugacity models for seabirds and mammals.

### *Quantifying processes and identifying influential parameters*

Single-species and food web models are advantageous and useful tools for quantifying the magnitude of competing processes for OC bioaccumulation. Many of the factors influencing OC bioaccumulation, such as body size, affect both uptake and elimination rates. Thus, single-species or food web models

are helpful to investigate the relative importance of uptake and elimination processes. Increased understanding of the processes involved can be achieved, because the confounding effect of correlated and dependent factors on OC bioaccumulation can be broken and, thus, studied separately. An example is the use of models to quantify processes of uptake and elimination in invertebrates and fish. The effect of growth dilution to reduce OC concentrations caused by increasing size of an organism may be overshadowed by the correspondingly reduced elimination rate by direct diffusion, which will lead to higher concentrations [35,87,88]. Another example of the utility of mechanistic mass-balance models is the empirical BMF. The BMFs are not only the result of exposure (e.g., diet) but also of elimination (e.g., biotransformation). Thus, these processes (uptake and elimination) are confounded in the BMF, and many OCs, such as PCBs with different configurations, will have BMFs mainly reflecting the half-life of the OC in the specific organism. By assuming no elimination, food web models break the dependencies between BMFs and half-life. This approach was used for a fugacity-based bioaccumulation model for the Barents Sea harp seal (*Phoca groenlandica*), with a diet of pelagic amphipods and krill, to estimate the maximum biomagnification (called  $Q$ ) assuming no biotransformation or elimination [164]. This  $Q$  value was then assigned to all compounds, and half-lives could be calculated and compared to the structure of the PCB and its potential for being metabolized. Interestingly, differences in calculated half-lives between various PCBs corresponded to their categorization as being either persistent or metabolizable [41,42]. A third example is the study of chemical distribution in the organism during the course of the fish life cycle [181] or during chick embryo development [54].

Another advantage of the mechanistic food web model is that the relative influence of input parameters can be investigated, both separately and combined, by systematic or random adjustment of input variables (see, e.g., [7,182,183]). For instance, using the food web model of Campfens and Mackay [14], the benthic invertebrate *Diporeia* was sensitive toward sediment OC exposure, organism lipid content, and bioavailability (as a function of organic carbon in sediments) [183]. Because the sensitivities of the input variables are driven by the conceptual model, the same variables will be sensitive for similar species from different ecosystems, although the actual ecosystems may behave differently, as will be seen from empirical data. Which parameter is important for the model output is also species-dependent.

### *Stable isotopes and TMFs in food web modeling*

An important question is whether TMFs can be incorporated into food web models. The TMF would seem to be a useful food web and chemical-specific parameter that might be used to replace other parameters with unknown values in most food webs (e.g., assimilation efficiencies and feeding rates in individual species). In Canadian lakes, the empirical linear relationship between log contaminant concentration and stable nitrogen (i.e., TMF) was used to estimate the prey-Hg concentration for fish predators [157]. Prey-Hg concentrations were then used in a bioenergetics-based food web model to successfully predict predator-Hg concentrations [10]. This illustrates the potential future use of TMFs and stable nitrogen isotopes in mechanistic food web models as the empirical relationships become available for different systems, lakes and seas, and temperate as well as arctic ecosystems (Table 2). In

this respect, it would also be valuable if the TMFs could be derived theoretically rather than empirically. Whether this is possible remains an open question, however, until more studies are conducted and data are available regarding POP behavior in food webs.

### CONCLUSION

Recent studies of arctic marine food webs have provided new, detailed information regarding the influence of chemical and, especially, biological factors on bioaccumulation and trophic transfer of persistent OCs. Studies in other regions, such as the Great Lakes and the Baltic Sea, have provided the data for improved understanding of bioaccumulation processes and for validating food web bioaccumulation models. Even so, the Arctic is unique because of longer food chains, reduced diversity of species, multiple and widely separated areas with similar food webs, and limited influence from pollution point sources. So far, however, only limited attempts have been made to model trophic transfer of OCs in the arctic marine food web.

Because of limitations, many previous field studies have often lumped together lower-trophic-level organisms with very different ecological and physiological characteristics. As the present summary of recent arctic marine food web studies shows, even organisms such as zooplankton show highly species-specific bioaccumulation of contaminants. Thus, species specificity is important to keep in mind, and bioaccumulation models likely will be improved if they are specified in terms of life stage and other parameters (because both ecological and physiological properties of an organism change during its lifetime).

Biological factors account for a high degree of the variation in OC levels and patterns both within and among species of the arctic marine ecosystem. The factors resulting in OC (or other contaminant) bioaccumulation are confounded and difficult to study separately in field studies. Thus, the use of models to assess the importance of competing processes might be crucial as a next or continuing step in our understanding of bioaccumulation in this system. The present overview has summarized the factors that have been shown to be important in the present empirical food web studies and that need to be considered in more detail during future studies. The factors shown to be of importance for describing differences in OC concentration and pattern among species are as follows: The organism's body size (because of physiological and ecological changes, e.g., diet and age), habitat use (e.g., OC exposure and diet change), reproductive status (e.g., transfer of lipids to offspring), seasonality (e.g., altering lipid content, bioavailability, feeding preferences, migration), biotransformation (e.g., depending on a species' capacity and a chemical's molecular structure), and trophic position (e.g., not only among seabirds and seals but also among invertebrates and fish). For instance, whereas the OC pattern in invertebrates and fish is predominantly influenced by direct elimination and physiochemical properties, the elimination pattern in seabirds and seals is determined by physiological parameters in terms of biotransformation capacities and transfer to offspring. The TMFs, which represent the average increase per trophic level among species with comparable physiology, may be useful descriptors of the magnification process, and they have been used with stable isotopes of nitrogen to estimate prey contamination.

The improvement in our knowledge regarding biomagnification of OCs that has occurred during the 1990s is valuable

for our understanding of other persistent halogenated compounds that are being detected in marine environments, such as the polybrominated diphenyl ethers and perfluoroacids. It is not clear, however, that the knowledge gained by the study of persistent OCs can be transferred easily to the huge array of other chemicals in commerce, including many high-production-volume chemicals, which typically are not halogenated and are less hydrophobic. A future challenge will be to include a better understanding of metabolism within the bioaccumulation and modeling framework, because so many chemicals in commerce have some structural features that are amenable to biotransformation compared to the recalcitrant OCs.

*Acknowledgement*—We thank A. Di Guardo for valuable comments on the modeling discussion. Katrina Borgå was supported by a Marie Curie Fellowship of the European Community program Improving the Human Research Potential and the Socio-Economic Knowledge Base under contract number HPMF-CT-2002-02045. Funding for PhD postdoctoral fellowship of P. Hoekstra was provided by the Department of Wildlife Management, North Slope Borough (Barrow, AK, USA).

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