Research

Why Do Organochlorine Differences between Arctic Regions Vary among Trophic Levels?

KATRINE BORGÅ,*,†
GEIR WING GABRIELSEN,†
JANNECHE UTNE SKAARE,‡,§
LARS KLEIVANE,‡
ROSS J. NORSTROM,‖ AND
AARON T. FISK¶

*Norwegian Polar Institute, N-9296 Tromsø, Norway,
Norwegian Veterinary Institute, P.O. Box 8156 Dep.,
N-0033 Oslo, Norway, Norwegian School of Veterinary
Science, P.O. Box 8146 Dep., N-0033 Oslo, Norway,
Chemistry Department, Carleton University,
Ottawa, ON, Canada K1S 5B6, and Warnell School of Forest
Resources, University of Georgia, Athens, Georgia, 30602-2152*

Statistical analysis of organochlorine contaminants (OCs) in marine mammals has shown that, for most OCs, the European Arctic is more contaminated than the Canadian and U.S. Arctic. Recently, comparison of OC concentration ranges in seabirds, arctic cod (*Boregadus saida*), and zooplankton, found no difference between these regions. To address these inconsistencies, marine food web OC data from the European (central Barents Sea (CBS)) and Canadian Arctic (Northwater Polynya (NOW)) were simultaneously statistically analyzed. In general, concentrations of OCs were greater in seabirds and ringed seals (*Phoca hispida*) from the CBS as compared to the NOW; consistent with circumpolar trends observed in marine mammals. In contrast, levels of OCs were generally similar in zooplankton and arctic cod between the CBS and NOW. The main exception is HCH which had greater levels in the NOW across all trophic levels because of the greater proximity to sources in eastern Asia. The lack of differences in OC concentrations in zooplankton and Arctic cod from the European and Canadian Arctic suggest that regional differences in OC contamination in the Arctic have evened out. Reduced regional differences were not observed in marine mammals or seabirds because they are long-lived and also acquire contaminants from maternal transfer and hence reflect levels from the past when the European Arctic was more contaminated than the Canadian Arctic. In addition, seabirds may reflect exposure from other areas. This study highlights the potential problem of comparing spatial trends by using means and confidence intervals as compared to simultaneous statistical analysis of raw data. Differences in the spatial trends of OCs between trophic levels in the Arctic are important for consideration

* Corresponding author phone: +47 777 505 00; fax: +47 777 505 01; e-mail: katrine.borga@npolar.no.
† Norwegian Polar Institute.
‡ Norwegian Veterinary Institute.
§ Norwegian School of Veterinary Science.
‖ Carleton University.
¶ University of Georgia.

when assessing regional differences in spatial and temporal trends of discontinued and current-use contaminants.

Introduction

Recent reviews of organochlorine contaminants (OCs) in the Arctic marine environment have highlighted spatial trends in biota (*1*, *2*). OC data for marine mammals, particularly for polar bears (*Ursus maritimus*) and ringed seals (*Phoca hispida*), are the strongest for spatial comparisons and cover most of the circumpolar region (*3*−*7*). In general, levels of polychlorinated biphenyls (PCBs) and dichloro-diphenyl-trichloroethane (DDT) in polar bears and ringed seals are greater in the European Arctic (northeast Greenland to northern Norway and western Russia) than in the Alaskan and Canadian Arctic (*3*−*7*). These PCB and DDT trends are true also when adjusting for important biological factors such as age, sex, and blubber thickness (*3*, *4*). Circumpolar trends of chlordane in ringed seals were weaker, but similar to PCBs and DDT (*4*). In polar bears, chlordanes were highest in the European Arctic with lower levels in the Chukchi Sea (*7*). Spatial trends of hexachlorocyclohexane (HCH) concentrations in marine mammals are opposite to those of PCBs and DDT, with greatest concentrations in the Alaskan and western Canada Arctic and lower concentrations in the European Arctic (*3*, *4*, *7*), probably because of sources of HCH in eastern Asia (*4*, *8*). Spatial trends of hexachlorobenzene (HCB) in ringed seals and polar bears are similar to HCHs, with lower concentrations in the European Arctic than in the other Arctic regions, although the pattern for HCB is not as consistent as that for HCH (*4*, *7*).

Assessment of spatial trends of OCs in other arctic marine animals, namely, zooplankton, fish, and seabirds, has been limited by fewer data (*1*, *2*). Spatial trends of OCs in these organisms have only been based on comparing means and ranges between separate studies (*1*, *2*, *9*−*11*). These comparisons noted that spatial trends of PCBs and other OCs in arctic marine zooplankton, fish, and some seabird species differ from those observed in marine mammals. In general, levels of PCBs, DDTs, and chlordanes were similar (overlapping ranges) in zooplankton and Arctic cod (*Boregadus saida*) in the Canadian (northern Baffin Bay), Alaskan (Beaufort Sea), and European arctic (Barents Sea) (*1*). Depending on species, PCBs and DDTs concentrations in European Arctic seabirds were similar to, or in the higher range, of those reported from the Canadian Arctic (*1*, *10*, *11*). Levels of HCHs in zooplankton, Arctic cod, and seabirds follow similar trends to those observed in marine mammals.

The impact of global bans or use-reductions of OCs and other persistent halogenated compounds may best be assessed by examining temporal trends of OCs in Arctic biota. From several perspectives, the Arctic is a favorable ecosystem to study the mechanisms of OC dynamics in the environment and for assessing worldwide spatial and temporal trends (*1*, *2*, *12*). OCs accumulate to significant levels in this remote ecosystem in large part because of a net transport of OCs that are still in use, or are not at global equilibrium, from temperate regions to the Arctic (*13*). Several arctic ecosystem characteristics motivate the study of OCs, including limited breakdown of OCs because of low temperature, high levels in organisms because of high lipid content and longevity, and the lack of pollution point sources. In fact, the presence of contaminants in the arctic region, and the implication
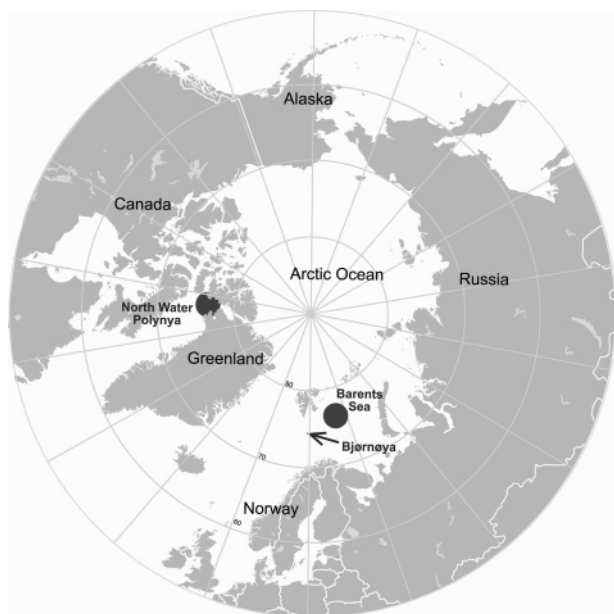
FIGURE 1. Location of food web studies in the northern Baffin Bay (Northwater Polynya) and the Barents Sea.

that contaminants are not limited by borders, was a driving force behind global bans of the most persistent and toxic organic contaminants (*1*). When assessing temporal change in environmental OC levels because of change in contaminant discharge, use, or production, one must make sure that the temporal change is not caused by other factors such as changed food web properties (and thus biomagnification) as a response to altered environmental conditions such as climatic changes or other factors.

As the contaminants follow the flux of energy through the food web, one would expect that the geographical OC trends in marine mammals and some seabird species are also seen in lower trophic levels of the food web and that geographic OC differences in predators reflect geographical OC differences in the diet. We hypothesize that differences between spatial trends of OCs in zooplankton, fish, and seabirds, and marine mammals, noted in recent reviews, are due to lack of rigorous statistical data analyses. To more fully examine spatial OC variation in the North American and European Arctic and the differences between OC trends in marine mammals and some seabird species, and lower trophic level animals, we combined selected data sets from food web studies in northern Baffin Bay (*11, 14, 15*) and Barents Sea (*9, 10, 16, 17*) and performed simultaneous statistical analyses.

Methods

Sampling Locations. Both the Northwater Polynya (NOW) in northern Baffin Bay and the central Barents Sea (CBS) are high-latitude marine seas with high primary productivity (Figure 1). The high productivity supports large, rich fisheries in the CBS as well as a rich feeding area for seabirds and marine mammals in both the NOW and the CBS (*18−21*).

Polynyas are areas of open water, often surrounded by sea ice, which persist throughout the winter in polar seas. The NOW is thought to be formed by a combination of latent (winds) and sensible (warm water cells) heat and receives water from the south along the Greenland coast and from the north through the Kane Basin (*22*). The CBS is an arctic shallow shelf sea (average depth 230 m), with water masses dominated by northeast flowing warm saline Atlantic water from the Norwegian Sea, which meets the colder and less saline Arctic water in the polar front. Details on the water masses from the NOW and the CBS during the respective

sampling campaigns can be found in Melling et al. (*22*) and Søreide et al. (*23*).

Sample Collection. Details on sample collection for the NOW (May and June of 1998) and the CBS (June 1995, May 1999) studies have been summarized previously (*9, 10, 16, 24, 25*). The present study includes only species collected from both sites (Table 1). In short, selected marine food web components from the NOW were collected in May and June of 1998 from CCGV Pierre Raddisson and from the CBS in May of 1999 from R/V Lance, respectively. In addition, some seabirds and seals were collected in June 1995 from the Barents Sea (Table 1). Zooplankton (copepods *Calanus hyperboreus* and *C. glacialis*, and amphipods *Themisto libellula*) were collected with zooplankton nets (520−1000 μm mesh) and sorted to species shortly after collection. Some pelagic amphipod samples from the crop of thick-billed murre (*Uria lomvia*) were included from the CBS, as analyses have shown that they did not differ in OC contents from those sampled from the water (*16*). *C. glacialis* and *T. libellula* are indicator species for Arctic water in the Barents Sea, and *C. glacialis* and *C. hyperboreus* are important numerically and in wet biomass (*23*). From the NOW, Arctic cod were sampled when observed swimming near the surface in broken ice at one location, whereas from the CBS most samples were from bottom trawls and some were collected by divers from the sea ice underside. Seabirds were collected opportunistically by shotgun from a zodiac (dovekie *Alle alle*, black guillemot *Cepphus grylle*, thick-billed murre, black-legged kittiwake *Rissa tridactyla*, glaucous gull *Larus hyperboreus*). Dovekie was collected from the CBS marginal ice zone in 1999, black guillemot and thick-billed murre from the CBS marginal ice zone both in 1995 and 1999, kittiwake from Bjørnøya in 1995 and the CBS marginal ice zone in 1999, whereas glaucous gull was only collected from Bjørnøya in 1995. Blubber from ringed seals was obtained from Inuit hunters from Grise Fjord, Canada and Qaanaaq, Greenland during the spring of 1998 (NOW) (*15*), whereas they were hunted from the CBS marginal ice zone in June 1995 (*25*). Only ringed seal males younger than 6 years and females all ages were included in the present study, as previous studies have demonstrated an age-related OC accumulation in older males (*15, 26*).

All samples were placed in Whirl Pak bags, polypropylene containers, cryovials, or aluminum foil and frozen until analyzed for OCs. Whole body was analyzed for OCs in zooplankton, whole body samples were analyzed for the CBS arctic cod, an approximately 3-cm cross section of the body posterior to head was analyzed for the NOW Arctic cod, and liver and blubber were analyzed for seabirds and seals, respectively.

OC Analysis. Methods with modifications for extraction, cleanup, separation, identification, and quantification followed standard procedures and are described in previous publications (*9, 10, 14, 15, 24, 25, 27*). The participating laboratories' analytical quality has been confirmed by good results from international intercalibration tests (*9, 24*). All NOW and CBS samples were analyzed on high-resolution gas chromatograph equipped with Ni-electron capture detector, except NOW ringed seals for which the gas chromatograph was equipped with a mass spectrometer detector.

The samples were analyzed for a suite of OCs, including HCB and various HCHs, chlordanes, DDTs, and PCBs, and only compounds analyzed in the same species from both areas were included in the regional comparison (see Table 1 footnote for detail on which compounds were included for the different animal groups). All compounds included in the present study were quantified above the tissue and compound dependent detection limit in more than 75% of samples from the respective species and geographic area,

TABLE 1. Arithmetic Means and 95% Confidence Interval (CI, $t_{0.05,df}$) for the Respective Sample Size of Organochlorine Groups (OC) (ng g^{-1} Wet Weight) and Lipid Content (%)[a] and Mean Stable Nitrogen Isotope Values (δ^{15}N) for the Respective Species and Area[g]

species	area	year	n	δ^{15}N[b]	lipid%[a] mean ± CI	CBS/NOW	HCB mean ± CI	CBS/NOW	ΣHCHs[c] mean ± CI	CBS/NOW	ΣChlordanes[d] mean ± CI	CBS/NOW	ΣDDT[e] mean ± CI	CBS/NOW	ΣPCBs[f] mean ± CI	CBS/NOW
Zooplankton																
Copepods	CBS	1999	15	7.0	2.6 ± 0.4	0.3	0.03 ± 0.01	0.1	0.3 ± 0.1	0.1	0.7 ± 0.1	0.9	0.5 ± 0.1	0.9	1.0 ± 0.2	0.5
Calanus spp.	NOW	1998	10	8.1	8.4 ± 2.0		0.4 ± 0.1	**CBS < NOW**	4.0 ± 3.2	CBS = NOW	0.8 ± 1.6	**CBS > NOW**	0.6 ± 0.3	CBS = NOW	1.9 ± 0.5	CBS = NOW
								$F = 32.1$, $p < 0.001$		$F = 0.3$, $p = 0.614$		$F = 14.9$, $p < 0.001$		$F = 1.2$, $p = 0.288$		$F = 3.1$, $p = 0.095$
Amphipods	CBS	1999	7	10.4	2.2 ± 0.5	0.9	0.8 ± 0.4	3.5	0.3 ± 0.1	0.2	2.0 ± 0.3	0.8	1.7 ± 0.4	0.9	1.9 ± 0.3	0.7
Themisto libellula	NOW	1998	5	10.0	2.4 ± 1.0		0.2 ± 0.1	**CBS > NOW**	1.5 ± 0.7	**CBS > NOW**	2.6 ± 1.7	CBS = NOW	1.9 ± 1.3	CBS = NOW	2.9 ± 1.3	**CBS < NOW**
								$F = 29.8$, $p < 0.001$		$F = 52.2$, $p < 0.001$		$F = 0.1$, $p = 0.739$		$F = 0.0$, $p = 0.883$		$F = 5.5$, $p = 0.044$
Fish																
Arctic cod	CBS	1999	11	12.5	3.8 ± 0.6	3.2	2.0 ± 0.2	2.5	0.8 ± 0.1	0.8	4.1 ± 0.6	1.5	3.4 ± 0.6	1.3	3.3 ± 0.4	2.2
Boreogadus saida	NOW	1998	8	14.0	1.2 ± 0.6		0.8 ± 0.2	**CBS > NOW**	0.9 ± 0.4	CBS = NOW	2.7 ± 0.7	CBS = NOW	2.6 ± 0.5	CBS = NOW	1.5 ± 0.7	CBS = NOW
								$F = 17.4$, $p < 0.001$		$F = 4.0$, $p = 0.063$		$F = 0.1$, $p = 0.817$		$F = 0.9$, $p = 0.365$		$F = 1.2$, $p = 0.293$
Seabirds																
Dovekie	CBS	1999	10	10.5	4.1 ± 0.4	1.0	18.5 ± 5.2	9.0	1.6 ± 0.6	0.6	24.1 ± 7.7	4.4	31.9 ± 7.6	3.5	129.5 ± 45.4	9.6
Alle alle	NOW	1998	10	11.6	4.0 ± 0.5		2.1 ± 0.6	**CBS > NOW**	2.8 ± 1.2	**CBS < NOW**	5.4 ± 1.8	**CBS > NOW**	9.1 ± 2.6	**CBS > NOW**	13.5 ± 4.5	**CBS > NOW**
								$F = 143.9$, $p < 0.001$		$F = 4.9$, $p = 0.040$		$F = 51.8$, $p < 0.001$		$F = 53.7$, $p < 0.001$		$F = 94.3$, $p < 0.001$
Black guillemot	CBS	1995	10	13.4	4.4 ± 1.9		14.8 ± 6.7	2.8	1.0 ± 0.4	0.2	12.8 ± 5.7	1.2	34.9 ± 23.7	0.6	100.5 ± 67.7	2.1
Cepphus grylle	CBS	1999	10	14.2	5.3 ± 2.3	1.4	26.9 ± 5.3	**CBS > NOW**	1.2 ± 0.3	**CBS < NOW**	14.3 ± 2.9	CBS = NOW	29.0 ± 6.8	**CBS < NOW**	99.5 ± 23.9	**CBS > NOW**
	NOW	1998	9	14.9	3.4 ± 0.8		7.5 ± 2.9	$F = 11.5$, $p = 0.002$	5.4 ± 1.6	$F = 92.1$, $p < 0.001$	11.6 ± 2.9	$F = 0.2$, $p = 0.688$	51.7 ± 25.1	$F = 11.6$, $p = 0.002$	46.7 ± 21.9	$F = 5.6$, $p = 0.025$
Thick-billed murre	CBS	1995	10	11.8	2.5 ± 1.2		11.4 ± 4.3	1.7	0.5 ± 0.1	0.3	4.4 ± 2.3	1.2	26.5 ± 17.4	0.8	43.8 ± 28.9	1.4
Uria lomvia	CBS	1999	9	13.1	3.4 ± 0.6	0.7	22.3 ± 4.7	**CBS > NOW**	0.7 ± 0.2	**CBS < NOW**	5.3 ± 1.1	**CBS > NOW**	24.6 ± 7.8	CBS = NOW	49.3 ± 16.6	**CBS > NOW**
	NOW	1998	10	13.8	4.1 ± 1.2		9.8 ± 2.7	$F = 19.8$, $p < 0.001$	2.2 ± 0.8	$F = 23.3$, $p < 0.001$	4.1 ± 1.1	$F = 10.5$, $p = 0.004$	31.8 ± 8.6	$F = 0.00$, $p = 0.834$	32.5 ± 10.5	$F = 11.1$, $p = 0.003$
Kittiwake	CBS	1995	11	12.7	3.9 ± 1.5		29.9 ± 9.1	2.9	1.4 ± 0.5	0.6	15.8 ± 5.3	1.8	46.2 ± 22.3	1.3	631.2 ± 319.2	4.6
Rissa tridactyla	CBS	1999	10	13.5	7.7 ± 2.9	1.4	37.7 ± 8.7	**CBS > NOW**	1.4 ± 0.4	**CBS < NOW**	37.4 ± 31.3	CBS = NOW	97.9 ± 32.8	CBS = NOW	491.9 ± 83.3	**CBS > NOW**
	NOW	1998	10	13.3	4.1 ± 1.2		11.6 ± 2.6	$F = 51.3$, $p < 0.001$	2.2 ± 0.6	$F = 10.5$, $p = 0.003$	14.7 ± 3.7	$F = 2.2$, $p = 0.148$	56.3 ± 14.2	$F = 2.7$, $p = 0.110$	122.8 ± 26.4	$F = 55.2$, $p < 0.001$
Glaucous gull	CBS	1995	15	15.2	4.1 ± 1.0	0.7	157.8 ± 68.3	6.0	9.0 ± 2.9	0.4	234.3 ± 133.4	2.8	1641.0 ± 775.6	4.5	5377.0 ± 3765.7	13.6
Larus hyperboreus	NOW	1998	10	16.4	5.6 ± 0.8		26.1 ± 4.3	**CBS > NOW**	23.7 ± 8.4	**CBS < NOW**	82.5 ± 21.5	**CBS > NOW**	365.9 ± 125.1	**CBS > NOW**	394.7 ± 162.2	**CBS > NOW**
								$F = 51.4$, $p < 0.001$		$F = 14.0$, $p = 0.002$		$F = 11.7$, $p = 0.002$		$F = 11.7$...		
														$F = 38.4$, $p < 0.001$		$F = 48.0$, $p < 0.001$
Marine Mammals																
Ringed seal	CBS	1995	7	14.5	94.8 ± 2.0	1.1	16.4 ± 5.6	1.0	59.5 ± 15.5	0.5	272.1 ± 98.1	1.4	678.2 ± 160.4	1.8	1070.4 ± 216.2	2.7
Phoca hispida	NOW	1998	31	17.3	88.2 ± 3.8		16.7 ± 3.3	CBS = NOW	109.1 ± 19.5	**CBS < NOW**	194.2 ± 40.8	CBS = NOW	371.1 ± 102.9	**CBS > NOW**	394.2 ± 93.4	CBS = NOW
								$F = 0.03$, $p = 0.860$		$F = 10.2$, $p = 0.003$		$F = 1.3$, $p = 0.269$		$F = 6.8$, $p = 0.013$		$F = 3.1$, $p = 0.095$

[a] NOW OC and lipid data from refs *11, 14, 15*. CBS OC and lipid data from refs *9, 10, 16, 17*. [b] NOW mean δ^{15}N values from refs *14, 15, 24*. CBS mean δ^{15}N values from refs *10, 12, 32*. [c] HCH is α and γ-HCH for zooplankton, β-HCH for seabirds, and α, β, γ-HCH for fish and seals. [d] Chlordanes is *cis*-chlordane, *trans*-chlordane, and *trans*-nonachlor for zooplankton; oxychlordane, *cis*-chlordane, *trans*-chlordane, and *trans*-nonachlor for fish; oxychlordane, *cis*-chlordane, and *trans*-nonachlor for seabirds; and oxychlordane and *trans*-nonachlor for seals. [e] DDTs is *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT for zooplankton and fish; *p,p'*-DDE for seabirds; and *p,p'*-DDE and *p,p'*-DDT for seals. [f] PCBs is CB-28, -31, -52, -99, -118, -138, -153, -180 for zooplankton and fish; CB-28, -47/48, -74, -95/66, -101/90, -99, -105, -118, -128, -137, -138, -153, -156, -170/190, -180, -187, -196/203, -194 for seabirds; and CB-28, -31, -47/48, -52, -74, -95/66, -101/90, -99, -87, -105, -110, -118, -128, -137, -138, -141, -149, -151, -153, -156, -157/200, -170/190, -180, -187, -196/203, -194 for seals. [g] Ratios of lipid content and OC concentrations between samples from the central Barents Sea (CBS) in the European Arctic and the Northwater Polynya (NOW) in the Canadian Arctic are based on mean values for the respective species. For seabirds from the CBS collected both in 1995 and 1999, the ratio is based on the mean of the two years. Summary statistics (F- and p-value) are given for the comparison of OC concentrations between the two areas, adjusted for lipid content (ANCOVA, Type III sum of square), with significant comparisons in bold.

and the recoveries were within acceptable ranges as described in previous publications (*9, 10, 14, 15, 24, 25, 27*).

Statistical Analysis. Organochlorine accumulation was compared between the two geographic areas using univariate and multivariate statistics: analysis of covariance (ANCOVA, Type III SS, SAS V8 for Windows) and principal component analysis (PCA, CANOCO 4.5 for Windows). As different OCs were quantifiable in the different animals, the analyses were done within each animal group (zooplankton, fish, seabirds, seal), in addition to one PCA including all animals, but only OCs quantified in all species. Missing values in zooplankton were replaced by randomly generated values for the respective OC and sampling site, using 0.5*DL as the mean and 40% variation for the CBS (3.5% of the samples), and one-third of the lowest concentration as the mean and 30% variation for the NOW (5% of the samples) (StatPlus in Excel for Windows). The ANCOVA included logarithmic transformed OC wet weight concentrations as response variable (sums of groups Table 1, selected individual compounds Table 2), geographic area as explanatory variable, and lipid content as covariable. For zooplankton and seabirds, where several species were included, species was included as a covariable for zooplankton, whereas the ANCOVA was performed for each seabird species. The PCA of OC concentrations included logarithmic transformed OC wet weight concentrations as response variable (individual compounds, see Table 1) and lipid content as covariable. Area was entered as an explanatory variable to evaluate the significance of separation of samples in the ordination space due to geographic area (redundancy analysis RDA) (*28, 29*). The significance was evaluated by partial Monte Carlo permutation test with 499 permutations. PCA of OC pattern included the proportion of each OC to ΣOC ($OC_i/\Sigma OC$) as response variables. For both concentration and pattern PCA, species was entered as a covariable when including more than one species to analyze the structure in the variance independent of species' variance; thus, the OCs were centered to an average value of 0.

Results and Discussion

The present study has been conservative by including only samples that were collected in the same season (May–June) from similar years (1998 and 1999; as well as Barents Sea (CBS) seabirds and ringed seal from 1995). CBS seabirds and ringed seals collected in 1995 and 1999 did not differ in OC concentrations in 78% of the comparisons ($F = 0.00-3.88$, $p = 0.0653-0.9740$), and thus the data were combined for each species. In addition, comparison within each organism group was either based on individual compounds (Table 2, Figure 2a–d) or on ΣOC group concentrations including only individual compounds quantified in both areas (Table 1).

The oceanographic and ecological characteristics of the two ecosystems examined in this study were similar, which is important when comparing OCs between systems. Characteristics such as habitat and trophic positions of species are important in the bioaccumulation of OCs in Arctic organisms (*12*). The NOW and CBS are both high latitude ecosystems with high seasonal productivity and similar assemblages of species. To investigate if diet in similar species differ between the two areas, the species' trophic position based on δ^{15}N values (*30, 31*) was compared between the NOW and CBS. In the present study, it is not of interest to asses the trophic position per se but rather to investigate if a difference in δ^{15}N values between the two areas ($\Delta\delta^{15}$N = $\delta^{15}N_{NOW} - \delta^{15}N_{CBS}$) is consistent across trophic levels from copepods to seabirds and seals. If the $\Delta\delta^{15}$N is consistent, a difference in signals at the base of the food web is reflected in the same magnitude throughout the food web, and the diet can be assumed to be comparable between the regions. The mean $\Delta\delta^{15}$N between the areas was 1.1 in copepods,

-0.4 in *T. libellula*, 1.5 in Arctic cod, 1.1 in dovekie, 1.1 in black guillemot, 1.4 in thick-billed murre, 0.2 in kittiwake, 1.2 in glaucous gull, and 2.8 in ringed seals (Table 1) (*10, 14, 24, 32, 33*). Thus, with the exception of amphipods, kittiwake, and ringed seals, the difference in δ^{15}N values, and hence trophic position, was relatively similar between the two areas within species. The $\Delta\delta^{15}$N indicates that both amphipods and kittiwake had a higher trophic position in the CBS than in the NOW, compared to the other species, whereas ringed seals had a lower trophic position in the CBS than in the NOW.

A recent comparison of trophic magnification factors (TMFs) among Arctic marine food webs (zooplankton–fish–marine mammals) demonstrated greater TMFs in the CBS than in the White Sea, the NOW, and the Beaufort Sea (*34*). This would result in higher OC concentrations in the upper trophic level species of the CBS compared to the NOW, probably because of a different food web composition between the two areas involving species not included in the above studies. However, OC TMFs in cold-blooded animals and OCs with similar TMFs between cold- and warm-blooded animals were similar between the NOW and the CBS marine food web (*24, 32*). We therefore assume that the trophic transfer of OCs is similar, and since trophic positions generally did not vary between the NOW and CBS, differences in OC concentrations between the two areas are due to other factors.

Because of the precautions used for data inclusion, the use of simultaneous statistical analyses (rather than comparing summary statistics), and the similarity of the two ecosystems, the present study represents the most thorough examination of differences of OCs in marine zooplankton, fish, and seabirds between arctic regions.

OC Concentration Comparisons. Significant differences in OC levels were found between the Canadian (northern Baffin Bay, NOW) and European (central Barents Sea, CBS) Arctic at most trophic levels, although the magnitude of difference, and the OCs contributing to a difference, was not the same for all animal groups. On the basis of ANCOVA and PCA results, spatial differences in OC concentrations in lower (zooplankton and fish) and upper (seabirds and marine mammals) trophic levels between the NOW and the CBS did not agree (Tables 1 and 2, Figure 2a–d). When all OCs were analyzed simultaneously with multivariate ordination, both OC concentrations and patterns differ between the CBS and NOW samples (Figure 3), although the compounds contributing to the difference varied with trophic level (Monte Carlo, F from 4.3 to 93.6, p from 0.002 to 0.008) (Figure 2). In general, with the exception of HCH, OC concentrations were greater in CBS seabirds and ringed seals compared with the NOW, consistent with circumpolar trends observed in ringed seals (*4*) and polar bears (*3, 6, 7*). Relative to seabirds and seals, however, OC concentrations in zooplankton and Arctic cod were often similar between the NOW and CBS, or greater in the NOW samples.

In general, concentrations of HCB and ΣHCHs differed consistently between the CBS and NOW for the various animal groups, with higher HCB and lower ΣHCH concentrations in the CBS than in the NOW (Table 1, Figure 2 a–d). The exceptions were HCH levels in copepods and arctic cod and HCB in copepod and ringed seals which did not differ significantly between the areas (Table 1, Figure 2 a, d). The NOW zooplankton and fish generally had larger variation among samples than the CBS samples for these OC concentrations, thus although the mean concentration of HCHs was higher in the NOW copepods compared to the CBS, the difference was not statistically significant (Table 1).

Spatial differences of the HCH in this study are the most consistent across animal groups for any of the OC groups examined here and agree with other circumpolar trends of HCHs observed in water (*35, 36*), and ringed seals (*4*). The

TABLE 2. Summary of Analysis of Covariance (ANCOVA) of Individual Organochlorine Compounds, Type III Sum of Square Statistics for Comparison of Samples from the Northwater Polynya (NOW) and the Central Barents Sea (CBS)[a]

	HCH			*trans*-nonachlor	*p,p′*-DDE	PCB							
	-α	-β	-γ			-28	-31	-52	-99	-118	-153	-138	-180
Copepods	CBS = NOW, F = 0.0, p = 0.969		CBS = NOW, F = 2.3, p = 0.148	**CBS > NOW, F = 19.2, p < 0.001**	CBS = NOW, F = 1.1, p = 0.300	**CBS < NOW, F = 19.4, p < 0.003**	**CBS < NOW, F = 35.2, p < 0.001**	**CBS < NOW, F = 7.8, p = 0.011**	**CBS > NOW, F = 35.8, p < 0.001**	CBS = NOW, F = 1.6, p = 0.224	CBS = NOW, F = 0.0, p = 0.987	**CBS > NOW, F = 18.6, p < 0.001**	CBS = NOW, F = 1.1, p = 0.301
Amphipods	**CBS < NOW, F = 65.2, p < 0.001**		**CBS < NOW, F = 15.6, p = 0.003**	CBS = NOW, F = 0.2, p = 0.694	CBS = NOW, F = 0.9, p = 0.369	**CBS < NOW, F = 22.6, p = 0.001**	**CBS < NOW, F = 29.0, p < 0.001**	**CBS < NOW, F = 11.0, p = 0.009**	CBS = NOW, F = 1.4, p = 0.270	CBS = NOW, F = 1.8, p = 0.211	CBS = NOW, F = 0.4, p = 0.571	**CBS > NOW, F = 5.4, p = 0.046**	CBS = NOW, F = 0.1, p = 0.732
Arctic cod	CBS = NOW, F = 0.1, p = 0.779	**CBS < NOW, F = 144.6, p < 0.001**	CBS = NOW, F = 1.8, p = 0.197	CBS = NOW, F = 2.4, p = 0.143	CBS = NOW, F = 0.8, p = 0.380	**CBS < NOW, F = 8.6, p = 0.010**	**CBS > NOW, F = 28.0, p < 0.001**	CBS = NOW, F = 0.2, p = 0.662	**CBS > NOW, F = 40.3, p < 0.001**	CBS = NOW, F = 0.3, p = 0.579	CBS = NOW, F = 2.0, p = 0.176	**CBS > NOW, F = 9.1, p = 0.008**	CBS = NOW, F = 0.9, p = 0.364
Dovekie		**CBS < NOW, F = 6.6, p = 0.020**		**CBS > NOW, F = 45.1, p < 0.001**	**CBS > NOW, F = 47.9, p < 0.001**	**CBS > NOW, F = 18.2, p < 0.001**			**CBS > NOW, F = 490.7, p < 0.001**	**CBS > NOW, F = 96.5, p < 0.001**	**CBS > NOW, F = 80.4, p < 0.001**	**CBS > NOW, F = 82.9, p < 0.001**	**CBS > NOW, F = 99.1, p < 0.001**
Black guillemot		**CBS < NOW, F = 110.9, p < 0.001**		CBS = NOW, F = 2.6, p = 0.123	**CBS < NOW, F = 10.0, p = 0.004**	CBS = NOW, F = 0.5, p = 0.511			**CBS > NOW, F = 36.2, p < 0.001**	**CBS > NOW, F = 14.8, p < 0.001**	**CBS > NOW, F = 12.5, p = 0.002**	**CBS > NOW, F = 14.7, p < 0.001**	**CBS > NOW, F = 4.5, p = 0.044**
Thick-billed murre		**CBS < NOW, F = 21.3, p < 0.001**		CBS = NOW, F = 3.7, p = 0.067	CBS = NOW, F = 0.1, p = 0.719	CBS = NOW, F = 2.9, p = 0.107			**CBS > NOW, F = 15.4, p = 0.008**	**CBS > NOW, F = 26.3, p < 0.001**	**CBS > NOW, F = 5.5, p = 0.030**	**CBS > NOW, F = 11.9, p = 0.003**	**CBS > NOW, F = 4.4, p = 0.049**
Kittiwake		**CBS < NOW, F = 10.5, p = 0.003**		**CBS < NOW, F = 5.5, p = 0.026**	CBS = NOW, F = 2.7, p = 0.110	**CBS > NOW, F = 5.1, p = 0.033**			**CBS > NOW, F = 94.4, p < 0.001**	**CBS > NOW, F = 52.1, p < 0.001**	**CBS > NOW, F = 76.5, p < 0.001**	**CBS > NOW, F = 61.4, p < 0.001**	**CBS > NOW, F = 65.9, p < 0.001**
Glaucous gull		**CBS < NOW, F = 13.0, p = 0.002**		CBS = NOW, F = 1.2, p = 0.291	**CBS > NOW, F = 38.4, p < 0.001**	**CBS > NOW, F = 19.6, p < 0.001**			**CBS > NOW, F = 107.8, p < 0.001**	**CBS > NOW, F = 73.6, p < 0.001**	**CBS > NOW, F = 38.2, p < 0.001**	**CBS > NOW, F = 56.2, p < 0.001**	**CBS > NOW, F = 40.1, p < 0.001**
Ringed seal	**CBS < NOW, F = 6.4, p = 0.017**	**CBS < NOW, F = 17.9, p < 0.001**		CBS = NOW, F = 1.5, p = 0.231	**CBS > NOW, F = 6.1, p = 0.019**	**CBS > NOW, F = 6.3, p = 0.017**		**CBS > NOW, F = 14.8, p < 0.001**	**CBS > NOW, F = 22.9, p < 0.001**	**CBS > NOW, F = 22.9, p < 0.001**	**CBS > NOW, F = 11.8, p = 0.002**	**CBS > NOW, F = 18.2, p < 0.001**	**CBS > NOW, F = 12.7, p = 0.001**

[a] Significant comparisons are in bold. Sample sizes for the respective comparisons are given in Table 1.

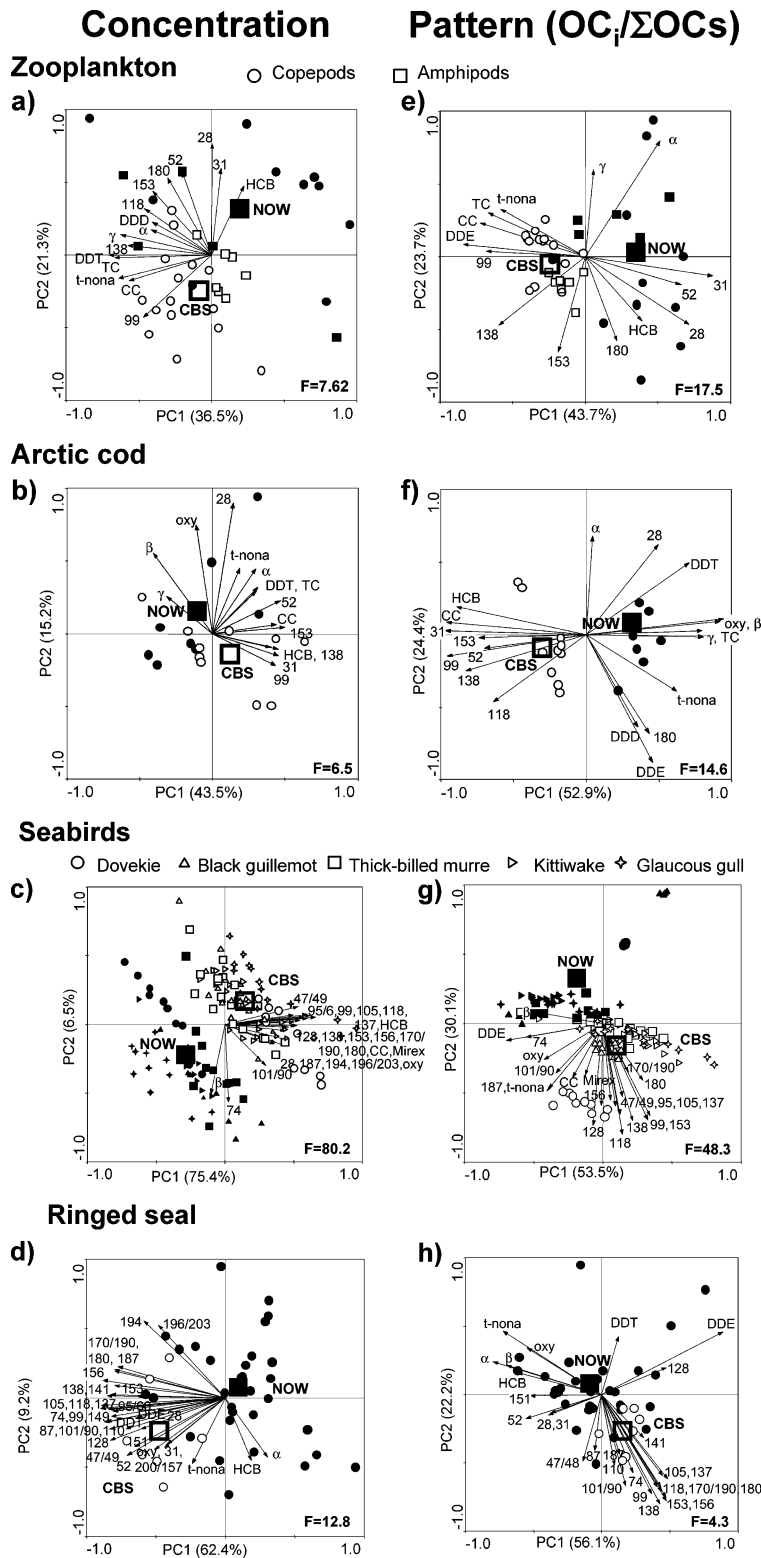# Concentration   Pattern (OC$_i$/ΣOCs)

FIGURE 2. Ordination diagram based on principal component analysis (PCA) of (a—d) log-transformed organochlorine (OC) wet weight concentrations in zooplankton, fish, seabirds, and seals with lipid content (%) as covariable and (e—h) PCA of relative OC pattern (OC$_i$/ΣOC) in zooplankton, fish, seabirds, and seals. In the PCA of zooplankton and seabirds, species was entered as a covariable to analyze the variance structure independent of species differences, and hence the OCs were centered to an average value of 0. Samples from the Northwater Polynya (NOW) are presented by filled symbols and from the Barents Sea (CBS) by open symbols, the mean of the samples indicated by a large square. *F* values are the result of Monte Carlo permutations; all *p* values were less than 0.008, showing that the OCs differed significantly between the NOW and the CBS. Only OCs (arrows) that correlated more than 15% in the ordination space are shown in the ordination diagram, and they are pointing in the direction of increased value. The relative percentage of the total variance explained by each of the principal components (PC) is given in brackets. Polychlorinated biphenyls are nominated by their congener number; α, β, and γ are hexachlorocyclohexane isomers; DDE, DDT, and DDD are all *p,p*′-isomers; TC, CC, oxy, and t-nona refer to *trans*-chlordane, *cis*-chlordane, oxychlordane, and *trans*-nonachlor, respectively.
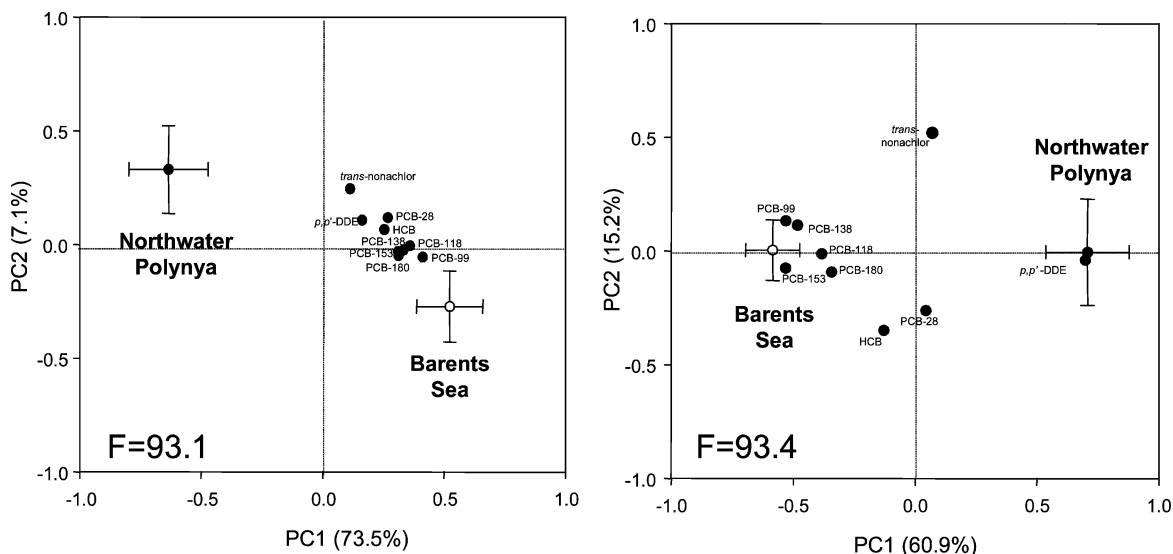
FIGURE 3. Ordination diagram of organochlorines (OCs) found in all animals combined from both the central Barents Sea and the Northwater Polynya. Biplot of loadings and samples' scores (mean ± 95%CI) is based on principal component analysis (PCA) of (left) log-transformed OC wet weight concentrations and (right) relative OC pattern ($OC_i/\Sigma OC$) of the compounds. Species, and lipid content for the concentration PCA, are accounted for as covariates (OCs were centered to an average value of 0), and thus the plots represent differences among samples because of sampling site. *F* values are the result of Monte Carlo permutations, both *p* values were 0.002.

fate of HCH is one of the best studied and understood OC in the Arctic, in part because it is easier to measure in air and water and its recent use has been confined to a narrower region of the globe (eastern Asia) (*37*). Higher levels of HCH in the NOW samples as compared to the CBS samples are probably due to the closer proximity of the Canadian arctic to sources of HCH in Asia, as from the ringed seal study (*4*).

HCB is a pesticide, and an industrial chemical byproduct that is still released to the environment throughout the northern hemisphere, and hence its circumpolar trends are likely to be more complex than HCHs. With the exception of copepods and ringed seals, HCB concentrations were greater in CBS biota. Higher HCB levels in the CBS than in the NOW may be due to the closer proximity of CBS to industry and agriculture as compared to the NOW. The greater concentrations of HCB in the NOW copepods may be a function of the 3.2 times higher lipid content in NOW copepods than in CBS copepods (Table 1). Higher lipid content in NOW copepods may be indicative of a postbloom situation after accumulating lipids compared to a prebloom or bloom situation in the CBS. During algal bloom, the amount of particulate matter in the water column increases as also the fraction of particle associated OCs (*38, 39*). Thus, during high primary production, there are lower concentrations of truly dissolved OC and lower concentrations of particle-associated OC as the algal biomass increases. Zooplankton are susceptible to slight changes in water concentrations of OCs; time of collection may have been when water concentrations of OCs in CBS were at their lowest or water concentrations of OC were at their highest in NOW. Thus, if the NOW and CBS copepods were experiencing different bloom situations, this could influence the OC bioaccumulation in copepods both from direct uptake from water and from the diet. On the basis of chlorophyll *a* biomass in the two areas during the sampling campaign, however, both regions were in a prebloom or bloom situation, depending on date and station (*23, 40, 41*). Prebloom or bloom in the NOW was supported by the copepods lipid content, which was still low in May and June compared to measurements from June to September (*14*). Thus, higher HCB in NOW copepods than in CBS copepods was not explained by different bloom situation.

The lack of difference in HCB concentrations between NOW and CBS ringed seals is similar to trends reported for

this species by Muir et al. (*4*), but there is no obvious explanation for this result. Ringed seal was one of the species where the difference in $\delta^{15}N$ values between the areas was greater than for the other species.

ΣChlordanes were higher in the Barents Sea than the NOW in copepods, dovekie, thick-billed murre, and glaucous gull, whereas no significant difference was found for the other animals (Table 1). ΣDDTs were significantly higher in the CBS than the NOW for dovekie, glaucous gulls, and ringed seals and were lower in the CBS than the NOW for black guillemot (Table 1). For the other animals, there was no significant difference between the two areas in ΣDDT levels. Individual chlordane compounds in zooplankton were higher in the CBS than in the NOW (Table 2, Figure 2a), whereas they did not differ significantly or contribute to separation of samples in the PCA in arctic cod, except oxychlordane that was higher in NOW than in CBS, and *cis*-chlorane that was higher in CBS than in NOW arctic cod (Figure 2b). Levels of individual DDT compounds in zooplankton and arctic cod were not different between the two areas, either analyzed separately or in the ordination (Table 2, Figure 2a–b). In seabirds and seals, the individual chlordanes and DDTs were higher in the CBS than in NOW (Figure 2c, d), although in some cases not significantly when analyzed separately in the ANCOVA (Table 2).

The lack of definitive geographic differences in Σchlordane and ΣDDT concentrations between the CBS and NOW are consistent with trends observed for Σchlordane and *trans*-nonachlor in polar bears (*3*) and ringed seals (*4*) and ΣDDT in ringed seals (*4*). The inconsistent spatial differences of chlordanes and DDT among trophic levels in the Arctic may reflect a combination of their history of use in North America and Europe. Chlordanes and DDT have been banned or have had restrictive use in North America and Europe since the 1970s (*42*), although DDT has continued to be used in Africa, Asia, and Russia. The reduction of use of these chemicals in North America and Europe has resulted in declines of these chemicals in arctic biota from these regions between the 1970s and 1990s (*1, 43, 44*) and may result in OC levels approaching equilibrium between the Canadian and European Arctic. The higher DDT and chlordane levels observed in the European Arctic in the past may have been a source of these contaminants to the Canadian Arctic. Temporal trends of several OCs in polar bear and other animals have

shown a slower decline in the Canadian as compared to the European Arctic (1), which supports this hypothesis. As discussed above for HCHs and HCB, the factors influencing lipid content may also influence dynamics of OCs such as Σchlordane and ΣDDT in zooplankton.

Another important factor that could influence observed Σchlordane and ΣDDT concentrations, and other OCs, in seabirds is winter migration (12, 44, 45). The CBS is closer to industrial, agricultural, and inhabited areas than the NOW, and migrating CBS seabirds are more likely to overwinter in more OC contaminated ecosystems. However, recaptures of CBS dovekie and thick-billed murre indicate a western migration to the southern tip of Greenland and Newfoundland (46), the same area where NOW dovekie and thick-billed murre overwinter (47). The present study's seabirds from the CBS marginal ice zone were likely nonbreeders in the year of collection, as they were captured far from the colonies. Some of these nonbreeders may be from colonies at Franz Josef Land, Novaya Zemlya at the northern and eastern border of the Barents Sea, respectively, as well as from colonies in the eastern Kara Sea such as Severnaya Zemlya (46; Strøm, personal communication). There are no recaptures of black guillemot tagged at Svalbard; however, they are considered to overwinter in ice-covered areas of the Arctic (46). Although some may migrate to the southern Barents Sea and coast of northern Norway, this is not well documented (Strøm, personal communication). Migration to the northern Norwegian coast for black guillemot, and sampling of nonbreeding dovekie and thick-billed murre individuals from eastern Kara Sea colonies, may explain elevated OC levels in these species as both the Norwegian coastline and the Kara Sea are more contaminated than the CBS and the NOW (1, 42). Barents Sea kittiwake and glaucous gulls probably migrate south and southwest dispersing over the whole north Atlantic region, where especially younger individuals are found overwintering close to Iceland, southern Greenland, and Newfoundland (48). Both kittiwake and glaucous gulls are also recaptured along the coast of north and mid-Europe (48), which would explain the elevated OC levels in these gulls.

ΣPCBs levels in copepods and arctic cod were not statistically different between the CBS and NOW (Table 1). In amphipods, the PCB levels were significantly higher in the NOW than in the CBS (Table 1). In seabirds and seals, ΣPCBs levels were significantly greater in the CBS as compared to the NOW (Table 1). The trends for the seabirds and ringed seals remained when individual PCB congeners were examined (Table 2, Figure 2c, d), and agree with past circumpolar studies of ringed seals (4) and polar bears (3). NOW zooplankton and Arctic cod had significantly greater concentrations of lower chlorinated PCBs but CBS zooplankton and arctic cod had greater concentrations of higher chlorinated PCBs (PCB-138) (Table 2, Figure 2a–b).

The PCB trends in the seabirds and ringed seals are consistent with past circumpolar trends in polar bears (3) and ringed seals (4) and reflect a general trend of higher PCB concentrations in the European versus North American arctic over the past 30 years (1, 42). The lack of such trends in the zooplankton and arctic cod may reflect an overall decrease in the differences of PCBs between these arctic regions in recent years. As discussed above for chlordanes and DDTs, PCBs have been banned in North America and Europe and the levels across the Arctic may be approaching steady state. Since the 1970s, PCB concentrations have declined faster in European Arctic than in Canadian Arctic animals (1). A change in PCB concentrations in the abiotic environment would be observed first in lower trophic level organisms since they are short-lived (<4 years for zooplankton and <10 years for Arctic cod) compared to seabirds and marine mammals that can live for decades. As well, maternal transfer of OCs to offspring

in zooplankton and fish is likely insignificant compared to mammals and birds. Since the half-life of recalcitrant OCs in seabirds and marine mammals is on the order of years, if not longer, OC concentrations in marine mammals and seabirds reflect a much longer time period (decades) than in zooplankton and arctic cod. Even young marine mammals and seabirds will reflect the past contamination, because of efficient maternal transfer of OCs to the offspring (26, 49). Thus, higher levels of PCBs in seabirds and marine mammals from the CBS are likely due in part to higher concentrations of PCBs in the past in this region.

Seawater was sampled during both field campaigns and analyzed for various OCs in the NOW (14) and for PCBs in the CBS (50). However, with the exception of HCHs, measurements of OCs in arctic seawater vary with orders of magnitude depending on study area and methods used for water collection and chemical analyses (17, 51). Therefore, to study spatial distribution of less water-soluble compounds, water is not recommended as the sample medium until the methods are improved and standardized.

The importance of different food sources for seabirds and seals between the two areas cannot be ruled out. However, the similarities in trophic positions except for kittiwake and ringed seals suggest that their diets are comparable. Previously, a diet of ice-associated fauna was suggested to explain elevated OC levels in polar bear living in ice-dense areas (3). However, later studies showed that ice fauna are not more highly contaminated with biomagnifying OCs than zooplankton (27). Similarly, benthic invertebrates with high OC levels (e.g. 52) are not likely prey for surface-feeding gulls.

Comparisons of Relative OC Pattern between Areas. The relative OC patterns in the NOW and CBS generally reflect the geographic variation seen from individual OC concentrations as described above. Although there was large variation among the samples in the ordination of OC pattern in all animals, even after adjusting for species difference, the two areas were significantly separated because of a higher relative contribution of p,p'-DDE to ΣOC in the NOW and a higher relative contribution of most PCBs (except PCB-28) to ΣOC in the CBS (Figure 3b). When the OC pattern within each animal group was analyzed (Figure 2e–h), zooplankton from the NOW had a higher relative contribution of HCHs, HCBs, and low-chlorinated PCBs than the CBS zooplankton, which had a higher relative contribution of chlordanes and highly chlorinated PCBs (Figure 2e). Arctic cod clearly differed in OC pattern between the areas with higher relative contributions of HCB, cis-chlordane, and most PCBs in the CBS arctic cod and higher relative contributions of β- and γ-HCH, oxychlordane, $trans$-chlordane, $trans$-nonachlor, and p,p'-DDT in the NOW arctic cod (Figure 2f). The two areas were not so clearly separated by the OC pattern in seabirds and ringed seals (Figure 2g, h). However, in general, NOW seabirds had higher relative contributions of β-HCH, p,p'-DDE, and PCB-74, whereas CBS seabirds had higher relative contributions of most PCBs (Figure 2g). The ringed seal OC pattern differs between the two areas mainly because of higher relative contributions of most PCBs in Barents Sea seals and higher relative contributions of HCHs, chlordanes, and HCB in NOW seals (Figure 2h).

The better area separation in OC pattern in lower trophic levels (zooplankton and fish) than in higher trophic levels (seabirds and seals) is probably due to the poor biotransformation ability in cold-blooded animals. Cold-blooded animals generally reflect the OC pattern seen in the surrounding water masses (e.g., 53). On the contrary, the poorer separation between samples from the two areas in OC pattern for seabirds and ringed seals reflects the species-specific ability of warm-blooded animals to biotransform and eliminate various OCs. Thus, the final OC pattern in warm-blooded animals is determined by their biotransformation

abilities (equal within a species across the Arctic) after exposure rather than by exposure alone (which might vary between regions).

Lessons Learned from the Present Study. The results of the present study illustrate potential problems with assessing trends in OC data on the basis of comparison of means or ranges, as compared to simultaneous statistical analysis of a data set. For example, by comparing concentrations it was concluded that ΣPCB levels in lower trophic level seabirds (e.g., thick-billed murre) did not differ between the CBS and NOW regions (*1, 10*). However, our statistical analysis of the same data as used in the above comparisons found that ΣPCB concentrations were greater in CBS thick-billed murre than in NOW thick-billed murre (Table 1).

The present study also demonstrates the potential problem associated with assessing differences or variation in contaminant data by using ΣOC groups as compared to individual compounds. The analysis of individual OCs in ANCOVA and by ordination analysis such as PCA demonstrates the importance of considering individual compounds rather than the sum of groups, as these summarize congeners of very different physicochemical properties, which may be subject to different transport pathways and other processes important for environmental distribution. Often, different studies use a different combination of congeners to calculate ΣOC concentrations, thus the comparison is flawed. Further, different congeners within a ΣOC group may show different environmental behavior that is lost when ΣOC groups are evaluated. This was observed in copepods where ΣPCB concentrations were not found to differ between the NOW and CBS (Table 1), whereas statistical comparisons of individual PCB congeners showed that NOW copepods had greater concentrations of lower chlorinated PCB congeners than the CBS copepods (Table 2). Similar conclusions regarding the problems associated with the use of ΣOC concentrations have been made when comparing HCH patterns among different animal groups (*53*).

As discussed above, there are several possible explanations for the variation in spatial OC differences observed between trophic levels in the Arctic, although none are likely to be exclusively correct for each OC or animal group (i.e., more than one factor is influencing observed concentrations). Regardless, differences in spatial trends of OCs among trophic levels or species are important for consideration when assessing regional differences in spatial and temporal trends of discontinued and current-use contaminants in the Arctic. This is particularly relevant since the Arctic has been widely used as a sentinel of global contaminant trends; for example, the banning of 16 persistent OCs by the Stockholm Convention of 2002 used the presence of contaminants in the Arctic as a persistence criterion (*37*). We suggest that any conclusions drawn from contaminant data, or any studies proposed for the Arctic that address spatial or temporal trends, consider the differences across many trophic levels. Questions or studies that address current inputs of contaminants to the Arctic are likely to be less biased if they consider lower trophic levels, as they more rapidly reflect environmental changes in OC exposure. However, studies of biological effects of contaminants in wildlife, or human exposure to contaminants in the Arctic, are likely more accurately assessed by studies of upper trophic level organisms.

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