SETAC PRESS

Environmental Toxicology and Chemistry, Vol. 25, No. 9, pp. 2502–2511, 2006

Printed in the USA
0730-7268/06 $12.00 + .00

BIOMAGNIFICATION OF POLYBROMINATED DIPHENYL ETHER AND HEXABROMOCYCLODODECANE FLAME RETARDANTS IN THE POLAR BEAR FOOD CHAIN IN SVALBARD, NORWAY

EUGEN GRAVNINGEN SØRMO,† MARIA PETTERSVIK SALMER,† BJØRN MUNRO JENSSEN,*† HAAKON HOP,‡ KINE BÆK,§‖ KIT MAUREEN KOVACS,‡ CHRISTIAN LYDERSEN,‡ STIG FALK-PETERSEN,‡ GEIR WING GABRIELSEN,‡ ELISABETH LIE,§ and JANNECHE UTNE SKAARE§‖

†Norwegian University of Science and Technology, Department of Biology, NO 7491 Trondheim, Norway
‡Norwegian Polar Institute, NO 9296 Tromsø, Norway
§Norwegian School of Veterinary Science, POB 8146 Department, NO 0033 Oslo, Norway
‖National Veterinary Institute, POB 8156 Department, NO 0033 Oslo, Norway

(*Received* 14 *October* 2005; *Accepted* 2 *March* 2006)

Abstract—Concentrations of brominated flame retardants (BFRs), including polybrominated diphenylethers (PBDEs) and hexabromocyclododecane (HBCD), were investigated in an arctic marine food chain consisting of four invertebrate species: polar cod (*Boreogadus saida*), ringed seals (*Pusa hispida*), and polar bears (*Ursus maritimus*). The most abundant BFR, brominated diphenyl ether (BDE)-47, was found in detectable concentrations even in zooplankton, the lowest trophic level examined in this study. Most of the investigated BFRs biomagnified as function of tropic level in the food chain. A noticeable exception occurred at the highest trophic level, the polar bear, in which only BDE-153 was found to increase from its main prey, the ringed seal, indicating that polar bears appear to be able to metabolize and biodegrade most BFRs. In contrast, lower-brominated PBDEs, particularly BDE-47, showed clear signs of bioaccumulation in zooplankton, polar cod, and ringed seals. We suggest that this discrepancy in the fate of BFRs among the different species may be related to greater induction of oxidative detoxification activities in the polar bear. Absorption and debromination rates may be more important for bioaccumulation rates of BFRs in zooplankton, polar cod, and ringed seals. Lipid weight–based concentrations (LWCs) and whole body–based concentrations (WBCs) of BFRs were used to assess biomagnification factors (BMFs). Whole-body concentrations gave the most realistic BMFs, as BMFs derived from LWCs seem to be confounded by the large variability in lipid content of tissues from the investigated species. This study demonstrates that PBDEs and HBCD have reached measurable concentrations even in the lower trophic levels (invertebrates and fish) in the Arctic and biomagnifies in the polar bear food chain.

Keywords—Arctic Marine food chains Biomagnification Flame retardants Pollution

INTRODUCTION

Because of their environmental stability, persistence, and high production volume, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD) are among the most abundant brominated flame retardants (BFRs) detected in the environment and in wildlife and human tissues [1,2]. Environmental concerns about BFRs (PBDEs in particular) are due to their structural, chemical, physical, and toxicological similarities to other, better-known persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs), which suggest that they might have similar ecotoxicological potential [3]. Three PBDE technical products have been used commercially: penta-, octa-, and deca-BDE technical mixtures [4]. Currently, no laws prohibit the use of PBDEs in Asia or in the United States, but the European Union banned the use of penta- and octa-BDE technical products, commencing in 2004 [4]. In the United States, California has decided to ban penta- and octa-BDEs by 2008, and recently the only American manufacturer of penta- and octa-BDEs, Chemtura Corporation (Middlebury, CT, USA), reached a voluntary agreement with the U.S. Environmental Protection Agency (http://www.epa.gov/opptintr/pbde/qanda.htm) to remove these two products from the U.S. market by 2005. Deca-BDE, consisting mostly of BDE-209, is thought to be less threatening to the environment because the large molecular size of this congener is assumed to limit its global atmospheric transport potential and its bioavailability over biological membranes [4,5]. Currently, no restrictions exist on the use of technical deca-BDE products [6,7] or HBCD [6].

Bioconcentration and bioaccumulation of POPs in marine organisms have been conclusively linked to the lipophilicity of the various compounds. Highly lipophilic compounds (log K_{ow} values >5) tend to adhere to particles in the water column or to sediments and are bioavailable to aquatic organisms mainly through dietary intake of particulate organic matter, contaminated prey organisms, or detritus [7,8]. The high hydrophobicity of PBDEs, with increasing log K_{ow} values for more highly brominated congeners [9,10], suggests high biomagnification potentials for these compounds in marine food webs [11]. However, because of low bioavailability, the biomagnification potential of higher brominated PBDEs such as BDE-209 is likely to be low despite their high hydrophobicity [4,5].

Few local sources of POPs, such as BFRs, PCBs, and organochlorine pesticides, exist in the Arctic, and the dominant route for these chemicals in the region is believed to be long-range transport by the atmosphere, ocean currents, river inputs, and sea-ice drift [12]. Because many BFRs are resistant to biodegradation and are thus biomagnified, concern exists that

* To whom correspondence may be addressed (bjorn.munro.jenssen@bio.ntnu.no).

Table 1. Biological measurements of polar cod (*Boreogadus saida*), ringed seals (*Pusa hispida*), and polar bears (*Ursus maritimus*) sampled in Svalbard, Norway, 2002–03

Species	Date	Sampling area	Sex	Age (years)	Mass (kg)	Length (cm)	Girth (cm)	Blubber depth (mm)
Polar cod	September 26, 2003		—		16.3×10^{-3}	12	—	—
Polar cod	September 26, 2003		—		16.7×10^{-3}	12	—	—
Polar cod	September 26, 2003		—		13.3×10^{-3}	11	—	—
Polar cod	September 26, 2003		—		13.9×10^{-3}	11	—	—
Polar cod	September 26, 2003		—		10.1×10^{-3}	10	—	—
Polar cod	September 26, 2003		—		14.9×10^{-3}	11	—	—
Polar cod	September 26, 2003		—		16.2×10^{-3}	12	—	—
Ringed seal	May 5, 2003	Forlandssundet	M	8	74.5	131	107	36
Ringed seal	May 5, 2003	Forlandssundet	M	9	78.5	139	107	36
Ringed seal	May 5, 2003	Forlandssundet	M	25	73.5	134	104	37
Ringed seal	May 7, 2003	Forlandssundet	M	22	88.5	144	111	43
Ringed seal	May 7, 2003	Forlandssundet	M	17	73.5	134	105	31
Ringed seal	May 8, 2003	Forlandssundet	M	19	69.5	133	106	35
Polar bear	February 3, 2003	Austfjorden	M	17	467.0	236	172	40
Polar bear	July 10, 2002	Eholmen	M	9	353.0	227	159	50
Polar bear	May 13, 2003	Longyearbyen	M	3	229.0	196	113	40
Polar bear	May 17, 2003	Mushamna	M	5	275.0	220	135	30

concentrations of these chemicals could reach levels high enough to cause harmful effects in individuals and populations. In polar bears (*Ursus maritimus*) from Svalbard, Norway, high levels of PCBs and organochlorine pesticides have been associated with disruption of sex and thyroid hormone balance [13,14] and immune function [15]. Reports of PBDEs in ringed seals (*Pusa hispida*) and polar bears in Svalbard [16,17] and of rapid increases in PBDE concentrations in ringed seals from the Canadian Arctic have resulted in further concerns about the effects of BFRs on arctic wildlife [18].

Numerous reports have been made on organochlorine uptake and biomagnification in arctic biota, but at present few studies of transport and biomagnification of BFRs in arctic ecosystems have been conducted [12,16,18]. Although organochlorines and BFRs have several physical, chemical, and toxicological properties in common, they are also quite different, particularly in the substitution of higher-molecular-weight bromine atoms in place of chlorine atoms. Thus, the uptake and biomagnification of BFRs in arctic food webs may differ significantly from that of organochlorines.

The main aim of the present study was to investigate food web transfer of PBDE and HBCD flame retardants in the marine ecosystem around Svalbard, Norway, with samples ranging across the trophic web from pelagic zooplankton to polar bears. The hydrophobic nature of many BFRs means that they associate with lipids in the bodies of organisms, and their concentrations are generally presented as lipid weight concentrations (LWCs) in most food web studies. However, LWCs are highly susceptible to variability in the lipid content of the organisms [19,20], which may bias their use in estimation of food web transfer and biomagnification of BFRs. In our study we use wet-weight whole-body concentrations (WBCs) as an alternative to LWCs for presenting BFR data.

MATERIALS AND METHODS

Sample collection

Lower trophic sampling of the marine food web included four invertebrate groups: pelagic, herbivorous calanoid copepods (mainly *Calanus glacialis*); the herbivorous, pelagic krill *Thysanoessa inermis*; the pelagic omnivorous amphipod *Themisto libellula* (grazing mainly on phytoplankton but also preying on small pelagic zooplankton, such as calanoid copepods); and the ice-associated, omnivorous amphipod *Gammarus wilkitzkii* [21,22]. The ice amphipod *G. wilkitzkii* is part of the sympagic (ice associated) food web, and its diet consists mainly of ice algae, herbivorous zooplankton, and detritus, which implies that this species has a somewhat larger range in trophic position (both lower and higher) than the other invertebrates in this study [22,23]. The polar cod (*Boreogadus saida*) feeds mainly on calanoid copepods, krill, and pelagic amphipods, and stable isotope analysis (nitrogen) confirms its trophic position to be one level above these species [23]. The ringed seal feeds mainly on polar cod but also consumes pelagic and sympagic zooplankton [24]. The polar bear is the apex predator of the arctic marine food chain, preying mainly on ringed seals, bearded seals (*Erignathus barbatus*), and harp seals (*Phoca groenlandica*) [25].

All animals, except for one polar bear, were collected between February and September 2003 (Table 1). The zooplankton species were collected from the R/V *Lance*, operated by the Norwegian Polar Institute, Tromsø, Norway, during August 2003. All sampling sites for zooplankton, polar cod, and ringed seals were located north of 78°N (Fig. 1). Sampling of pelagic zooplankton species (calanoid copepods, *T. inermis* and *T. libellula*) was carried out using a Tucker trawl (1,000-μm mesh). The depth of trawling ranged from 0 to 350 m, depending on the location. The ice amphipod *G. wilkitzkii* was collected by scuba divers using an electrical suction pump beneath drifting sea ice [26]. Polar cod ($n = 7$) were collected during September 2003 in the marginal ice zone northeast of Nordaustlandet, Svalbard, using a bottom trawl from the R/V *Jan Mayen*, operated by the University of Tromsø, Tromsø, Norway. Ringed seal ($n = 6$) were shot during May 2003 on sea ice in Forlandssundet, Svalbard. The polar bears ($n = 4$) were shot at various locations around Svalbard during 2002–2003. The bears were shot by representatives of the Governor of Svalbard in protection of settlements or by others in self-defense when bears attacked. Samples of zooplankton were wrapped in aluminum foil and stored in 50-ml polyethylene containers. Individual specimen samples of whole polar cod and blubber/adipose tissue samples from the seals and bears were wrapped in aluminum foil and stored in plastic bags. All

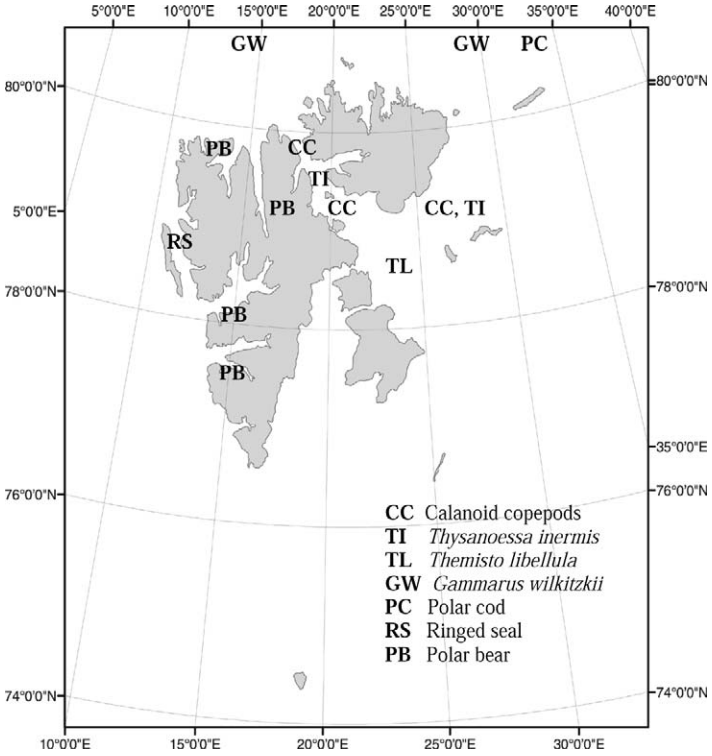

Fig. 1. Map of the Svalbard archipelago, Norway, indicating sampling sites.

samples were kept frozen at −20°C. Only large individuals of *T. libellula* and *G. wilkitzkii*, presumed to be adults, were collected in order to standardize the material. For ringed seals and polar bears, only males, mainly adults, were sampled.

Analytical method for BFRs

The chemical analyses of BFRs were done in the Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science in Oslo using gas chromatography/mass spectrometry (GC-MS) analysis. One pooled sample (~150 g) of each invertebrate species was crudely homogenized in a food blender. Two aliquots (~20 g) of the homogenates of the calanoid copepods, *T inermis* and *T. libellula* and five aliquots (~20 g) of *G. wilkitzkii* were made. Whole polar cod (~10 g), blubber from ringed seals (~2 g), and adipose tissue from polar bears (~2 g) were homogenized separately with scalpels in Petri dishes. The homogenates were transferred to 80-ml centrifuge tubes, and an internal standard mix (100 ng/ml) of brominated diphenyl ether (BDE)-77, BDE-119, BDE-181, and ^{13}C-BDE-209 (Cambridge Isotope Laboratories, Andover, MA, USA) was added to each sample. Cyclohexane (20 ml), acetone (15 ml), distilled water (10 ml), and 2% NaCl (2 ml) were also added to each sample from the mammals; in the invertebrates and cod samples, no water was added because of their naturally high water content. The samples were subjected to further homogenization using an Ika Ultra Turrax™ for 1 min (Janke & Kunkel, Staufen, Germany) and then an ultrasonic homogenizer (4710 Series; Cole Parmer Instrument, Chicago, IL, USA) for 2 min. The samples were centrifuged for 10 min at 3,000 rpm, and supernatants were collected. The lipid extraction was repeated by adding cyclohexane (10 ml) and acetone (5 ml) followed by ultrasonic homogenization treatment and centrifugation. The supernatants of both extractions were merged and concentrated to approximately 1 ml using a Zymark® evaporation system (TurboVap II; Zymark Corporation, Hopkinton, MA, USA) at 40°C with a gentle flow of nitrogen (pressure 0.6 bar). The concentrated lipid extracts were transferred to volumetric flasks, and the final volume was adjusted to 10 ml with cyclohexane. An aliquot of 1 ml from all samples was evaporated to dry condition on a sand bath (ST7; H. Gestigkeit, Düsseldorf, Germany) at 40°C for gravimetrical determination of the extractable lipid content.

For sample cleanup (removal of lipids), 4-ml aliquots of the lipid extracts from the polar bears and seals and 9 ml from the polar cod and zooplankton were treated with 6 ml ultraclean (purity 98.8%) concentrated H_2SO_4 (Scanpure; Chemscan AS, Elverum, Norway) and gently blended twice on a Whirl mixer for approximately 1 s. The samples were left in darkness for 60 min and then centrifuged for 20 min at 3,000 rpm. The supernatants and some of the acid layer were treated with 4 ml H_2SO_4 for a second time, left in darkness for 60 min, and then centrifuged. The polar cod and zooplankton samples were subject to a third cleanup with 2 ml H_2SO_4. Following the final centrifugation, the cleaned supernatants of polar bear and ringed seal were concentrated to 0.5 ml, polar cod to 0.3 ml, and invertebrate samples to 0.1 ml on a sand bath at 40°C under a gentle nitrogen flow. The sample concentrates were transferred to GC vials and put in dark containers.

For separation and detection of PBDEs (except from BDE-209) and HBCD, extracts (1 μl) were injected from an autosampler (Agilent 7683 Series; Agilent Technologies, Avondale, PA, USA) on a GC (Agilent 6890 Series; Agilent Technologies) configured with an MS detector (Agilent 5973 Network; Agilent Technologies). The injection was pulsed splitless. Both splitless and pulse were 1 min (pulse pressure 50 psi, temperature 250°C). The column was an SPB-5 of 60-m and 0.25-mm i.d., 0.25-μm film (Supelco, Bellefonte, PA, USA). Helium of purity 5.0 (Hydro Gas, Rjukan, Norway) was used as a carrier gas with a constant flow of 1 ml/min. The temperature program was as follows: 90°C (held for 2 min), 90 to 190°C (2.5°C/min), 190°C (held for 1 min), 190 to 250°C (5°C/min), 250°C (held for 1 min), 250 to 320°C (2.5°C/min), 320°C (held for 10 min). The total run time was 58 min.

For detection of BDE-209, extracts (10 μl) was injected on a GC-MS (Agilent 6890 Series/5873 Network) configured with a programmable temperature vaporization (PTV) injector (Agilent Technologies). The temperature program for this injector was as follows: 50°C; 50 to 90°C (27°C/min); 90 to 320°C (100°C/min). The ventilation time was 1.5 min, with a flow at 100 ml/min and a pressure of 0 psi. The column was a DB-5ms of 10 m, 0.25-mm i.d., 0.10-μm film layer (Agilent Technologies). The temperature program was as follows: 80°C (held for 2 min), 80 to 315°C (25°C/min), 315°C (held for 10 min). The total run time was 21.4 min.

In all GC-MS analyses, the temperature quadropole was set to 106°C, ion source to 250°C, and interface to 300°C. The GC-MS was operated in the electron capture mode (NCI) with methane of purity 4.7 as (Hydro Gas) reagent gas. To monitor the different BFRs, selected ion monitoring was used. The PBDEs (except from BDE-209) were monitored at *m/z* 79/81. Hexabromocyclododecane was monitored at *m/z* 79/81 and 159.8. Brominated diphenyl ether-209 was monitored at *m/z* 484 and 486 and ^{13}C-BDE-209 at *m/z* 495 and 497. Electron energy of 86.6 eV was used.

Chromatographic data were calculated using the software MSD ChemStation G1701 version C.00.00 (Agilent Technologies). Concentrations of the individual BFRs were determined

by corresponding components in the standards and analyzed for BDE-28 (2,4,4′-tribromodiphenyl ether), -47 (2,2′,4,4′-tetrabromodiphenyl ether), -99 (2,2′,4,4′,5-pentabromodiphenyl ether), -100 (2,2′,4,4′,6-pentabromodiphenyl ether), -153 (2,2′,4,4′,5,5′-hexabromodiphenyl ether), -154 (2,2′,4,4′,5′,6-hexabromodiphenyl ether), -183 (2,2′,3,4,4′,5′,6-heptabromodiphenyl ether), -209 (2,2′,3,3′,4,4′,5,5′,6,6′-decabromodiphenyl ether), and HBCD (hexabromocyclododecane). Quality assurance for the analyses included a six- to eight-point linear calibration curve of the analyzed standard solutions. Detection limits were set to about three times noise level and varied among species and chemicals: 0.012 to 1.299 ng/g lipid weight in invertebrates, 0.030 to 0.30 ng/g lipid weight in polar cod, and 0.014 to 0.75 ng/g lipid weight in the ringed seal and the polar bear. Hexabromocyclododecane consists of three diastereomers: α-, β-, and γ-HBCD. At temperatures above 160°C in the injection port, as used in this GC analysis, thermal rearrangement of the diastereomers leads to isomeric interconversion of β-, and γ-HBCD to α-HBCD [27]; thus, our results predict total HBCD.

The internal standards were used to detect and correct changes in compound concentrations during the chemical preparation and injection of the extracts into the GC-MS run. Recovery of samples of corn oil spiked with BFR standard solutions were also analyzed following each sample series. Mean percent recovery and coefficient of variance (CV) of the individual BFRs in the corn oil samples ranged from 70 to 115% and 1 to 28%, respectively. Standard solutions were run every 10 samples during the GC-MS analysis to detect any drift in the responses of the analysis. Reproducibility over time was tested continuously by analyzing the laboratory's own controls (seal blubber) at a minimum of one sample per series. Concentrations of the components in the seal blubber control we compared to the mean of previous years; they were within two times standard deviation of the mean. The laboratory is accredited by Norwegian Accreditation (Kjeller, Norway) for testing BFRs in biological material of animal origin according to the requirements of the NS-EN ISO/IEC 17025 (test 051, Norwegian Standard-English Standard International Organization for Standardization/International Electrochemical Commission). The laboratory's analytical quality for BFRs was approved in several intercalibration tests [28,29]. Because of high levels of BDE-153 and -183 in the blanks of the polar cod batch, coinciding with high levels of these compounds in polar cod extracts, we could not validate and report these compounds in this species. Moreover, because of the expectation of very low levels of BFRs in the lowest trophic levels of pelagic zooplankton species (*C. glacialis*, *T. inermis*, and *T. libellula*), relatively large samples (20 g) were used. The cleanup procedure used on these extracts was insufficient and caused contamination and subsequently destruction of the GC columns when running these samples. This made it impossible to report and validate BDE-209 and BFRs of long retention times on the SPB-5 column (BDE-153 and -183 and HBCD) in these species. Because of the financial risk of destroying even more columns, new attempts to analyze this material were not made.

Data presentation and statistical analysis

Lipid weight–based concentrations of BFRs in homogenates of zooplankton and polar cod and in blubber of ringed seals and adipose tissue of polar bears represented LWCs of BFRs. Wet weight–based concentrations (ng/g) of BFRs in homogenates represented corresponding WBCs of BFRs in zooplankton and polar cod, whereas the following (Eqn. 1) was used to calculate wet weight–based WBCs of BFRs in ringed seals and polar bears:

$$\text{BFR WBC ng/g} = \frac{\text{BFR LCW ng/g} \cdot \text{TLC (\%)}}{100} \quad (1)$$

where TLC is total lipid content (%). Total blubber content (TBC %) of ringed seals was calculated using the following formula (Eqn. 2) of Ryg et al. [30]:

$$\text{TBC (\%)} = 4.44 + 5{,}693 \cdot \sqrt{(\text{L/M})} \cdot \text{d} \quad (2)$$

with body mass in kg (M) and standard length (L) and blubber thickness (d) in meters. Total lipid content was then calculated from Equation 3:

$$\text{TLC (\%)} = \frac{\text{TBC (\%)} \cdot \text{blubber lipid content (\%)}}{100} \quad (3)$$

Estimated TLC in ringed seals ranged from 28 to 36%. The estimation of TLC in polar bears is a more complicated process. Farley et al. [31] reported TLC to range from 5 to 10% in polar bears after extensive fasting to as high as 50% during periods of hyperphagia. The bears of the present study were judged to be of normal or intermediate nutritional condition, and as a rough approximation, TLCs were set to 25% in all bears in the present study. Biomagnification factors (BMFs) of BFRs were calculated (Eqn. 4) based on Muir et al. [32]:

$$\text{BMF} = [\text{BFR}_{\text{predator}}]/[\text{BFR}_{\text{prey}}] \quad (4)$$

The BFR concentrations were expressed as either LWC or WBC (ng/g).

Statistical analyses were conducted using SPSS® Version 11.5 (SPSS, Chicago, IL, USA). The invertebrate data are represented by one observation for each species, which is the mean value of two replicates for *C. glacialis*, *T. inermis*, and *T. libellula* and the mean value of five replicates for *G. wilkitzkii*. The CV ranged from 1 to 20% for BDE-47, -99, and -100 on pelagic zooplankton. In *G. wilkitzkii*, CV ranged from 2 to 8% for BDE-47, -99, -100, -154, and -209. One observation from each species did not allow for statistical comparisons of concentrations between the zooplankton species. Pelagic zooplankton (calanoid copepods, *T. inermis* and *T. libellula*) were therefore merged into one group and compared to polar cod. Because of the relatively small sample sizes, with undetermined distributions, nonparametric Kruskal–Wallis (when comparing more than two groups/species) and Mann–Whitney tests (when comparing two groups/species) were used to compare BFR concentrations among the different trophic levels. Significance was set at $p < 0.05$.

RESULTS

In the lowest trophic level of the investigated food chain, BDE-47 and -99 were detected in all four species of invertebrates (Table 2). Brominated diphenyl ether-47 was about 10 times higher in WBC for *G. wilkitzkii* compared to pelagic zooplankton (calanoid copepods, *T. inermis*, and *T. libellula*; Table 3 and Fig. 2). Whole-body concentrations of BDE-99 were also relatively similar among the pelagic zooplankton species but about 10 times higher in the ice amphipod *G. wilkitzkii*. Brominated diphenyl ether-100 was detected in both species of amphipods but was 10 times higher in terms of WBC in *G. wilkitzkii* compared to *T. libellula*. Among the invertebrates, BDE-209 was analyzed only in *G. wilkitzkii*,

Table 2. Statistical data (ng/g lipid wt) for the analyzed polybrominated diphenyl ethers (PBDE) and hexabromocyclododecane (HBCD) flame retardants in different species of the polar bear food chain at Svalbard. ΣPBDEs = sum concentration of congeners BDE-28, -47, -99, -100, -153, -154, -183, and -209). NA = not analyzed; ND = not detected; SD = standard deviation. Different letters (A, B, or C) indicate difference in contaminant concentrations between polar cod, ringed seal, and polar bear[a]

	Lipid (%)	*n*		BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-209	HBCD	ΣPBDEs
Calanoid copepods	9.96	1	Mean[a]	ND	0.08	0.08	ND	NA	ND	NA	NA	0.16
Thysanoessa inermis	12.42	1	Mean[a]	ND	0.17	0.09	ND	NA	ND	NA	NA	0.26
Themisto libellula	9.01	1	Mean[a]	ND	0.33	0.05	0.11	NA	ND	NA	NA	0.53
Gammarus wilkitzkii	3.85	1	Mean[a]	ND	2.36	3.95	0.88	ND	0.26	7.22	ND	14.67
Polar cod (*Boreogadus saida*)	12.01	7	Mean	0.10A	0.81A	0.17A	0.18A	NA	0.09A	0.20	1.89A	1.99A
			Median	0.09	0.66	0.11	0.13		0.09	0.20	1.73	1.98
			SD	0.02	0.53	0.18	0.11		0.07	0.13	0.56	1.19
			Min–max	0.07–0.12	0.06–1.78	0.03–0.57	0.10–0.41		0.02–0.19	0.05–0.42	1.38–2.87	0.49–3.59
Ringed seal (*Pusa hispida*)	89.23	6	Mean	1.36B	49.30B	2.33B	4.69B	0.69A	0.71B		19.56B	59.08B
blubber			Median	1.02	46.96	1.96	3.65	0.68	0.55		16.95	55.36
			SD	0.81	14.16	1.46	2.68	0.35	0.50	ND–0.02[b]	7.57	19.73
			Min–max	0.67–2.87	36.45–73.83	1.01–4.79	2.84–9.78	0.34–1.27	0.38–1.69		14.6–34.5	42.04–94.23
Polar bear (*Ursus maritimus*)	83.85	4	Mean	0.19C	22.39C	0.79C	1.25C	5.24C	0.11A	0.09	11.53B	30.04C
adipose tissue			Median	0.19	21.86	0.72	1.00	4.25	0.11	0.08	12.16	27.44
			SD	0.08	7.360	0.22	0.56	2.71	0.08	0.05	5.299	10.32
			Min–max	0.09–0.27	14.10–31.73	0.62–1.11	0.90–2.09	3.28–9.18	0.02–0.20	0.03–0.16	5.31–16.51	20.74–44.55

[a] Mean of two replicates in Calanoid copepods (predominantly *Calanus glacialis*), *Thysanoessa inermis, Themisto libellula,* and five replicates in *Gammarus wilkitzkii.*
[b] BDE-209 was detected in only one ringed seal.

where it was found to be the most abundant BFR, constituting about 50% of the total PBDE load in this species (Table 2).

In the polar cod, BDE-28, -47, -99, -100, -154, and -209 and HBCD were detected. Because only BDE-47 and -99 were quantifiable in all pelagic zooplankton species (calanoid copepods, *T. inermis*, and *T. libellula*), statistical comparisons between pelagic zooplankton and polar cod were performed only for these compounds. Whole-body concentration of BDE-47 in the polar cod was 3 to 10 times higher than in its pelagic zooplankton prey ($p = 0.017$) but similar to the concentration in the ice amphipod *G. wilkitzkii* (Table 3 and Fig. 2). No significant difference was observed in WBCs between pelagic zooplankton and polar cod for BDE-99. Whole-body concentrations of BDE-99 and -209 were, however, 10 times higher in *G. wilkitzkii* than in polar cod (Table 3 and Fig. 2). The higher level of BFRs in *G. wilkitzkii* relative to pelagic zooplankton and polar cod was even more pronounced when considering LWCs (Tables 2 and 3).

In the ringed seal, BDE-28, -47, -99, -100, -153, and -154 and HBCD were detected in all specimens, whereas BDE-209 was detected in only one seal. The biomagnification in WBC of BDE-47 from polar cod to ringed seal was 200 times (Table 3 and Fig. 2). Whole-body concentrations of BDE-28, -99, -100, and -154 and HBCD showed more modest 20 to 85 times biomagnification from polar cod to ringed seal (Table 3 and Fig. 2). The corresponding BMFs from polar cod to seal, based on LWCs, were two to four times lower compared to those based on WBCs (Tables 2 and 3).

In the polar bear, all compounds except for BDE-183 were quantifiable, and WBCs and LWCs of most compounds were approximately two to four times lower than those found in ringed seals (Table 3 and Fig. 2) except for BDE-153, which increased five times from ringed seals to polar bears.

DISCUSSION

Analyses of the bioaccumulation and transfer of POPs within an assemblage of biota (a food web) requires an understanding of how key organisms interact (e.g., predator/prey interactions) [6]. The arctic marine food web tends to be relatively simple and fairly well understood, and this study covers the core linkages between trophic levels of the polar bear food chain. However, although pelagic zooplankton such as calanoid copepods, krill, and *T. libellula* [23] are the dominant prey for polar cod, in Svalbard they likely also consume abundant species of sympagic fauna, such as *G. wilkitzkii*, at least seasonally, and hence this species is included in the present study. Likewise, although ringed seals have a strong preference for polar cod, they do also consume some benthic fish and invertebrate fauna as well as sympagic zooplankton, likely including *G. wilkitzkii*. Finally, although ringed seals make up much of the polar bear diet, they are also known to eat a wide variety of other organisms; locally in Svalbard, bearded seals and harp seals are known to be important components of the diet, at least seasonally [24]. Hence, the species analyzed in this study represent the basic linkages in this food chain (web) but do not represent the complete picture of exposure to and bioaccumulation of BFRs in Svalbard polar bears.

Estimates of BMFs of lipophilic compounds in food chain studies are usually expressed as body burdens based on lipid concentrations. We, however, suggest that WBCs might be a more appropriate dose metric to use when exploring patterns across species in a food chain, this particularly when upper-trophic-level organisms tend to consume lower-trophic-level

Table 3. Biomagnification factors (BMFs) of polybrominated diphenyl ether congeners (PBDE), sum of all BDEs (ΣPBDEs), and hexabromocyclododecane (HBCD) in the polar bear food chain, calculated from mean whole-body concentration (ng/wet wt), and mean lipid-weight concentrations (ng/g) in brackets. Several BMFs could not be calculated (—), especially for zooplankton and polar cod, because of no detection in the lower trophic levels. Copepods = calanoid copepods (predominantly *Calanus glacialis*), *T. inermis* = *Thysanoessa inermis, T. libellula* = *Themisto libellula, G. wilkitzkii* = *Gammarus wilkitzkii,* polar cod = *Boreogadus saida,* ringed seal = *Pusa hispida,* polar bear = *Ursus maritimus*

BMF (predator/prey)	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-209	HBCD	ΣPBDEs
T. libellula/copepods	—	3.8	1.3	—	—	—	—	—	3.0
		(4.1)	(0.65)						(3.2)
G. wilkitzkii/copepods	—	11.4	19.0	—	—	—	—	—	34.7
		(29.1)	(47.6)						(87.8)
Polar cod/copepods	—	9.1	1.6	—	—	—	—	—	10.5
		(10.1)	(2.1)						(12.9)
Polar cod/*T. inermis*	—	3.3	1.2	—	—	—	—	—	5.2
		(4.7)	(1.9)						(7.9)
Polar cod/*T. libellula*	—	2.4	1.3	3.0	—	—	—	—	3.5
		(2.5)	(3.4)	(1.6)					(3.9)
Polar cod/*G. wilkitzkii*	—	0.8	0.1	0.4	—	—	0.1	—	0.3
		(0.4)	(0.04)	(0.2)			(0.03)	—qc	(0.1)
Ringed seal/*T. inermis*	—	632	54.5	—	—	—	—		496
		(285)	(26.8)						(227)
Ringed seal/*T. libellula*	—	463	60.0	214	—	—	—	—	364
		(148)	(43.1)	(42.3)					(111)
Ringed seal/*G. wilkitzkii*	—	153	3.9	31.5	—	—	—	—	29.0
		(20.9)	(0.6)	(5.4)					(4.1)
Ringed seal/polar cod	34.4	209	56.6	85.2	—	18.5	—	36.4	155.2
	(13.6)	(56.0)	(13.7)	(26.1)		(7.9)		(10.9)	(36.9)
Polar bear/ringed seal	0.16	0.40	0.29	0.23	5.2	0.25	—	0.6	0.41
	(0.1)	(0.5)	(0.3)	(0.3)	(7.5)	(0.32)		(0.7)	(0.5)

organisms whole. In the present study, BMFs calculated from WBCs and LWCs both revealed biomagnification of BFRs in the investigated food chain. However, some differences were observed between the two methods of expressing BMFs. The most noticeable difference was the much higher BMFs of BFRs based on WBCs from zooplankton and polar cod to the ringed seal as compared to BMFs calculated from LWCs (Table 3). These differences between the methods in terms of BMFs are almost certainly related to the 3 to 10 times higher lipid content of whole seals compared to their prey. This therefore suggests that use of LWCs may underestimate the biomagnification potential of BFRs in some species such as the ringed seal. Concerns also exist other than the great variability in lipid content between species that may favor the use of WBCs in studies of food chain transfer and dispersion of POPs in marine ecosystems. This includes the difficulty in measuring lipids by gravimetrical methods when lipid contents are low and that result in high variability for LWCs. The use of LWCs also assumes that the lipid measured represents the lipids in which POPs are stored. However, this assumption may be incorrect because POPs are associated mainly with triglycerides, and this fraction of the total lipid content of an organism varies between species [33,34]. Furthermore, several studies have argued that the nonlipid fraction of tissues could be important in explaining the fate of POPs in biota and that the nonlipid fractions are more important in small organisms (e.g., zooplankton) compared to larger organisms [7,35]. Additionally, many POPs are not uniformly distributed in the different lipid compartments of organisms, causing their LWCs to vary among different tissues types [19,20,36]. Thus, the use of LWCs may give inaccurate impressions of total body burden and even for tissue-specific concentrations of contaminants.

Although the use of WBCs can circumvent many of the limitations of LWCs listed here, challenges also exist with this method. One of the challenges of generating WBCs of POPs for large animals such as seals and polar bears is that they cannot be easily homogenized. In these species, estimated total contents of POPs in the blubber divided by total body mass can be used as an approximation for estimating WBCs of POPs. However, this means that POPs stored in lipid compartments other than the blubber will not be included in the calculation. In all probability, this is not a large setback when working with seals or polar bears since these lipids likely represent a negligible part of the WBCs because most of the lipids (triglycerides) and lipophilic POPs are stored in their blubber [37]. However, if compounds bind specifically to proteins or more polar lipids in tissues other than blubber, estimates using this sort of extrapolation will be inaccurate.

One of the more interesting findings in the present study is that despite one to two orders of magnitude higher WBC loads of most BFRs in the diet of the polar bear (ringed seals) compared to the diet of the ringed seal (zooplankton and polar cod), most BFRs were found in lower concentrations in the polar bear. This suggests a marked difference in biotransformation and biodegradation rates for BFRs in these two mammalian species. This is in accordance with previous findings of higher oxidative biotransformation and biodegradation capacities for other POPs, such as PCBs, in polar bears compared to seals [14,38,39]. This difference may relate to species-specific traits, or it may be the result of enhanced induction of detoxifying enzymes in the polar bear due to their much higher exposure to POPs. The apparently high biotransformation rate of BFRs in polar bears raises concerns about syntheses of potentially toxic BFR metabolites in this species [17]. The biomagnification of BDE-153 from the ringed seal to the polar bear indicates that this congener, relative to other BFRs, might be more resistant to biotransformation and thus biomagnifies even in polar bears. Marine mammals seem less able to biotransform and biodegrade 2,5- and 2,3,6-substituted PCBs when they have a 2,4,5-substitution on the second ring [38]. The substitution of bromine on BDE-153 fits with this prediction, which might explain the biomagnification of this con-

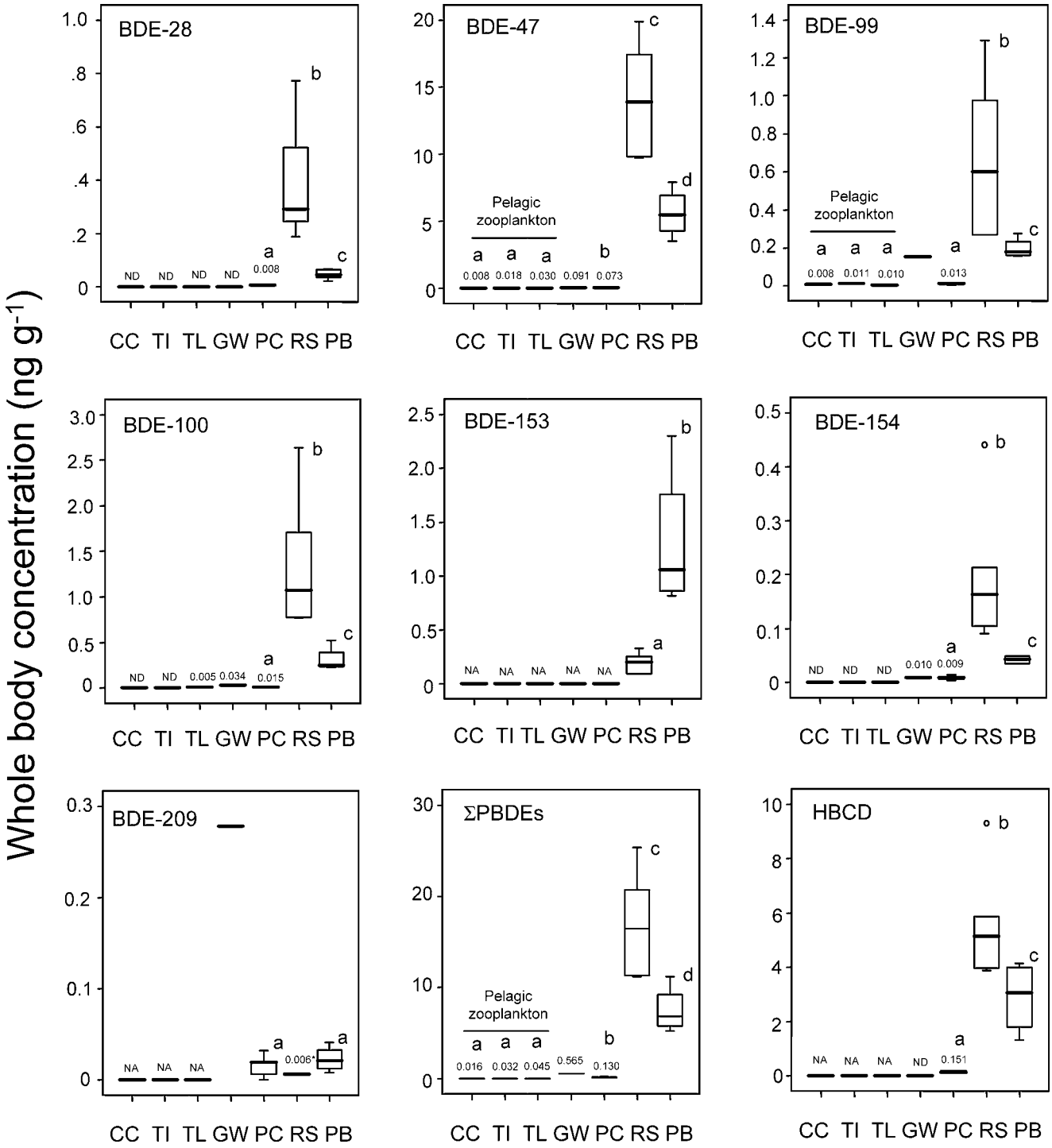

Fig. 2. Whole-body concentrations (ng/g) of individual polybrominated diphenyl ether (PBDE) congeners, ΣPBDEs (sum of BDE congeners), and hexabromocyclododecane (HBCD) in the polar bear (*Ursus maritimus*) food chain in Svalbard. ND = not detected; NA = not analyzed. See Figure 1 for definition of additional acronyms. Data are presented in a box-and-whisker plots. Unequal letters indicate significant differences ($p < 0.05$) in contaminant concentrations between pelagic zooplankton (as a group of three species), polar cod, ringed seals, and polar bears. * = BDE-209 was detected in only one ringed seal.

gener from ringed seals to polar bears. Thus, the most bioaccumulative PBDEs seem to be those that have halogen substitution pattern similar to the most bioaccumulative PCBs (e.g., PCB 153), at least when oxidative detoxifying processes are highly involved for the fate and degradation of these compounds in the organisms.

A slight increase of BDE-47 from pelagic zooplankton to the polar cod suggests that this compound can bioaccumulate and biomagnify in the lower trophic levels of the arctic marine food web. In contrast, BDE-99 showed no biomagnification from pelagic zooplankton to polar cod. Similar findings for the bioaccumulation of BDE-47 and the lack of biomagnification of BDE-99, measured as WBCs, have also been reported from zooplankton to arctic char (*Salvelinus alpinus*) on Bjørnøya, Svalbard [40]. These in situ findings are consistent with observations of a lower assimilation of BDE-99 compared

to BDE-47 in common carp (*Cyprinus carpio*) experimentally exposed to these compounds, probably as a consequence of intestinal or tissue debromination of BDE-99 to BDE-47 and other compounds [41]. Also among pelagic zooplankton, BDE-47 but not BDE-99 increased from calanoid copepods to *T. libellula*, indicating biomagnification of BDE-47. The detection of BDE-100 at a level just above detection limit in *T. libellula* but not in its copepod prey suggests biomagnification potential of this compound in the lowest trophic levels. Brominated diphenyl ether-47 also showed the highest biomagnification rate in the seals. In contrast to these findings in ringed seals, we found harbor seals (*Phoca vitulina*) from the Outer Oslofjord, Norway, to bioaccumulate BDE-153 more readily than BDE-47 (Gaustad et al., Norwegian University of Science and Technology, Trondheim, Norway, unpublished data). This discrepancy might be related to the fact that North Sea harbor seals are exposed to much higher POP loads (e.g., PCBs) than the arctic ringed seals [8,42]. This possibly enhances their expression of oxidative detoxifying enzymes and metabolism of metabolizable POPs in a concentration- or exposure-dependent manner as shown in Atlantic and Baltic grey seals (*Halichoerus grypus*) [43]. This suggests that factors such as absorption (bioavailability) and debromination rates of BFRs and, to a lesser extent, oxidative enzymatic reactions account for the variability in the bioaccumulative potentials of the different BFRs in arctic ringed seals. In contrast, oxidative enzymatic processes may be more important in predicting bioaccumulation potentials of BFRs in North Sea harbor seals and Svalbard polar bears.

In contrast to the modest biomagnification of BFRs in the lower parts of the arctic marine food web was a more than two orders of magnitude biomagnification of many BFRs from zooplankton and polar cod to the ringed seal. This is consistent with the much greater energetic demands and food intake of homeotherms (mammals) compared to poikilotherms (zooplankton and fish) [23]. Generally, this implies that homeothermic animals should have a higher concentration of POPs than poikilothermic animals and that the steps in the marine food chain with the highest biomagnification should be where homeotherms eat poikilotherms or other homeotherms. The differences in energetic demands between poikilotherms and homeotherms might be more pronounced in the Arctic compared to temperate or tropical waters because the energetic demands of poikilotherms (e.g., fish) are highly temperature dependent and thus particularly low in the cold waters of the Arctic [44].

Although BDE-209 is often by far the predominant PBDE in the abiotic media (e.g., sediments), it contributes in only a minor way to the total PBDE load in tissues of marine organisms [45]. This is presumably due to the large molecular size and subsequently limited digestive absorption of this compound in organisms [46]. Kinetics may also control uptake of BDE-209, as its strong binding to surfaces/particles may cause little of this compound to be free and available for intestinal accumulation in organisms. Thus, it is noteworthy that BDE-209 was found to account for more than 50% of the total PBDE load in the detritus-feeding ice amphipod *G. wilkitzkii*. However, it must be noted that since the samples of the ice amphipod are homogenates of whole specimens, BDE-209 could be located within their digestive system or even stuck onto their body surface and thus not subject to real uptake. Moreover, BDE-209 was also detected in polar cod (accounting for ~10% of the total PBDE load). Unfortunately, BDE-209 could not be analyzed in key prey species (pelagic zooplankton) of the polar cod in this study to establish BMFs for this compound in the polar cod. The presence of deca-BDE in the polar cod may represent novel uptake at the level of this species, or, alternately, they may get it from prey such as *G. wilkitzkii*. Moreover, since polar cod homogenates were used in this study, the possibility that the BDE-209 found in the polar cod originates from their gut content cannot be ruled out. The detection of BDE-209 in the blubber of one ringed seal and in the adipose tissue of all the polar bears, however, suggests that deca-BDE 209 may be subject to some uptake and food web transfer in arctic wildlife. Accumulation of BDE-209 has also been reported in peregrine falcon (*Falco pereginus*) eggs from Sweden [47] and in the blood of industrial workers exposed to BDE-209 [48]. The transport of high-brominated PBDEs is believed to be particle controlled and short range. However, the presence of BDE-209 in arctic biota indicates that these compounds probably are capable of long-range transport and dispersal. Detection of BDE-209 has also recently been reported in plasma of polar bears (<0.10 ng/g wet wt) and glaucous gulls (*Larus hyperboreus*) (0.03–0.43 ng/g wet wt) at Svalbard [17].

Gammarus wilkitzkii differed from pelagic zooplankton in having higher levels of BDE-99 compared to BDE-47 (Table 2), which might be related to its diet and gut contents (detritus). This as BDE-99 is often found in comparable or higher levels than BDE-47 in marine organic matter and sediments [5,45]. Furthermore, overall BFRs levels were also significantly higher in the ice amphipods compared to the pelagic zooplankton species of the present study. When organic matter is degraded by bacteria, the concentration of POPs in the remaining detritus (or decomposed organic matter) typically increases [49], causing detritus-eating marine organisms to be exposed to higher concentrations than pelagic feeding organisms.

Reports of HBCD in marine ecosystems and food chains are scarce [16,50,51]. The finding of no biomagnification from ringed seals to polar bears indicates that HBCD, like most of the investigated PBDEs, is biodegradable in the polar bears. However, the substantial biomagnification of HBCD from polar cod to ringed seals indicates its high bioaccumulation potential in other species, emphasizing that cause for concern and the need of risk assessment studies on HBCD at the ecosystem level might exist. For instance, complementary studies are needed to assess the bioaccumulative potential of the different HBCD diastereomers at the various trophic levels of the herein investigated polar bear food chain.

Recent observations in beluga whales (*Delphinapterus leucas*) from the St. Lawrence estuary, Canada, have shown that the time necessary for the concentrations of the most prevalent PBDEs (e.g., BDE-47) to double in the blubber was no longer than three years [52]. Concentrations of PBDEs have also increased in Canadian ringed seals over the past two decades [18], and LWCs of BDE-47 in the ringed seals in the present study sampled in 2003 were two to three times higher (17 vs 49 ng/g lipid) than levels reported in ringed seals sampled in Svalbard during 1999 [16]. This emphasizes that cause for concern about increasing levels of BFRs in the Norwegian Arctic might exist. However, the evidence for this is equivocal; LWCs of BDE-47 in polar cod in the present study were three to four times lower than those reported in 1999 [16]. The concentrations of BDE-47 in ringed seals at Svalbard are somewhat higher than those reported for ringed seals from the Canadian Arctic [18]. Total PBDE levels in polar bears of this

study were two to four times higher than in female polar bears at various locations in Alaska and the Canadian Arctic [53]. Boon et al. [11] reported LWCs of BDE-47 in Atlantic cod (*Gadus morhua*) and harbor seals from the North Sea that were more than an order of magnitude higher (133 and 1,236 ng/g lipid wt, respectively) than corresponding concentrations in polar cod and ringed seals in the present study. In the blubber of two harbor seals from the western Wadden Sea, concentrations of HBCD were found to range from 63 to 2,055 ng/g [50], which are higher than the corresponding LWC levels in the ringed seals (20 ng/g lipid) in the present study. This illustrates the generally lower contaminant load of BFRs in the European Arctic compared to continental European waters. It should, however, be noted that use of LWCs in studies aiming to monitor spatial and temporal trends in BFR contamination are susceptible to or biased by variability in lipid content in the organisms being compared. This may be particularly important for marine biota from the Arctic, where seasonal variability in day lengths and algae production severely affects the seasonal food availability and hence the lipid content of most organisms [7]. We therefore recommend the use of WBCs in studies concerning monitoring BFRs in marine organisms. This will probably result in more realistic descriptions of mechanisms involved in biomagnification processes and allow for development of better models to estimate dispersion of POPs in marine ecosystems.

Acknowledgement—E.G. Sørmo and M.P. Salmer contributed equally to this paper. This work was partly funded by the FIRE (Flame Retardant Integrated Risk Assessment for Endocrine Disruption; http://www.rivm.nl/fire) project (contract QLT4-CT-2002-00596) under the European Commission 5th Framework Program for research, technological development and demonstration activities (Quality of Life and Management of Living Resources, Key Action 4: Environment and Health). However, the contents herein do not represent the opinion of the European Community. Appreciation is expressed to the staff at the Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science and especially to Siri Føreid for assistance with the GC-MC procedures. Thanks are also given to Bjørn Krafft at the Norwegian Polar Institute for assistance with the sampling of ringed seals. The authors also thank the Governor of Svalbard (Sysselman) for providing the polar bear samples.

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