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Polybrominated diphenyl ethers (PBDEs) in marine mammals from Arctic and North Atlantic regions, 1986–2009

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ABSTRACT

A selection of PBDE congeners was analyzed in pooled blubber samples of pilot whale (*Globicephala melas*), ringed seal (*Phoca hispida*), minke whale (*Balaenoptera acutorostrata*), fin whale (*Balaenoptera physalus*), harbor porpoise (*Phocoena phocoena*), hooded seal (*Cystophora cristata*) and Atlantic white-sided dolphin (*Lagenorhynchus acutus*), covering a time period of more than 20 years (1986–2009). The analytes were extracted and cleaned-up using open column extraction and multi-layer silica gel column chromatography, and the analysis was performed on a GC-MS system operating in the NCI mode. The highest PBDE levels were found in the toothed whale species pilot whale and white-sided dolphin, and the lowest levels in fin whales and ringed seals. One-sided analyses of variance (ANOVA) followed by Tukey comparisons of means were applied to test for differences between years and sampling areas. Due to inter-year sampling variability, only general comparisons of PBDE concentrations between different sampling areas could be made. Differences in PBDE concentrations between three sampling periods, from 1986 to 2007, were evaluated in samples of pilot whales, ringed seals, white-sided dolphins and hooded seals. The highest PBDE levels were found in samples from the late 1990s or beginning of 2000, possibly reflecting the increase in the global production of technical PBDE mixtures in the 1990s. The levels of BDE #153 and #154 increased relative to the total PBDE concentration in some of the species in recent years, which may indicate an increased relative exposure to higher brominated congeners. In order to assess the effect of measures taken in legally binding international agreements, it is important to continuously monitor POPs such as PBDEs in sub-Arctic and Arctic environments.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are aromatic compounds substituted with up to ten bromine atoms that have been used extensively as flame retardants in for example textiles, plastics, and electronics. Since most brominated flame retardants do not react with the material in which they are incorporated, an extensive usage on a global scale has caused a release of PBDEs into the environment. In the early 1980s, PBDEs were found by Andersson and Blomkvist (1981) in eel, pike, bream and sea trout from the Viskan–Klosterfjorden water system, close to Gothenburg in Sweden. Bioaccumulation in wildlife has since then been reported in numerous studies, even in places with no local point sources or industrial production (Law et al., 2003; Strandberg et al., 2001; ter Schure et al., 2004). Various detrimental effects related to PBDE exposure have been studied in recent years. Changes in neurobehaviour, effect on thyroid hormone levels and fetal toxicity have been observed in exposure studies of rodents (Darnerud, 2003; Eriksson et al., 2001). PBDEs have mainly been available in three technical mixtures, penta-BDE, Octa-BDE and Deca-BDE. The penta mixture consists primarly of tetra- (BDE #47), penta- (BDE #99, #100) and hexa- (BDE #153, #154) congeners, the octa mixture consists primarily of a hepta congener (BDE #183) and a few octaBDEs. DecaBDE consists primarily of the fully brominated BDE #209 (La Guardia et al., 2006). As a result of the ratification of the WEEE and RoHS directives, the penta- and Octa-BDE mixtures were banned in the European Union in 2004 and in 2009 they were added to

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the list of persistent organic pollutants (POPs) under the Stockholm convention. The DecaBDE mixture was banned from the Swedish market in the beginning of 2007 and from the whole of the European market in the middle of 2008 (Kemmlein et al., 2009). In order to assess the effect of measures taken in legally binding agreements, for example the LRTAP Protocol and Stockholm Convention, it is important to continuously monitor POPs such as PBDEs in remote environmental compartments. However, since POPs are long lived and globally distributed it may take several years before effects of implemented reduction measures can be evaluated. Many marine mammal species are suitable for studies of long-term exposure of chemicals in the marine environment since they feed at the top of the aquatic food chain and have a relatively long life-span. PBDE concentrations as high as 3000–5000 ng/g lipid weight (lw) have been reported in blubber of pilot whales from the Faroe Islands and sperm whales and harbor seals stranded along the Dutch coast (de Boer et al., 1998; Lindström et al., 1999). PBDEs have been hypothesized to be transported to remote regions like the Arctic by similar atmospheric pathways as PCBs, and that tetra- and penta-brominated PBDEs have accumulation potential comparable to that of hexa- to heptachloro biphenyls (Wania and Dugani, 2003). Although a general trend in the last decades has been that the traditional POPs have declined in arctic biota whereas the PBDEs have increased, PBDE concentrations have shown different patterns of change in different species and geographical areas (Braune et al., 2007; de Wit et al., 2006; Helgason et al., 2008; Rigét et al., 2010; Vorkamp et al., 2008). In the Canadian Arctic, an increase in PBDE levels was found in burbot liver, ivory gull eggs and East Canada beluga sampled in the period 1980–2006 (Braune et al., 2007; Stern and Tomy, 2007; Tomy et al., 2007). A possible decline or tendency to leveling off was found in West Canada ringed seal and eggs of thick-billed murre and northern fulmars between 2000 and 2006 (Braune, 2006, 2007; Ikonomou et al., 2005; Riget et al., 2006). In the Barents Sea region, temporal trend studies in seabird eggs indicate a rapid increase of PBDEs from 1983 to 1993, after which the concentrations leveled off (Helgason et al., 2010).

The aim of the present study was to provide information on PBDE levels in marine mammals from Arctic and North Atlantic regions over the last 20 years. The current data is based on analysis of selected PBDEs in blubber samples of seven whale and seal species sampled in East Greenland, Faroe Islands, Iceland, West Ice and Norway between 1986 and 2009.

2. Materials and methods

2.1. Sampling

Sampling parameters of pilot whale (*Globicephala melas*), ringed seal (*Phoca hisida*), minke whale (*Balaenoptera acutorostrata*), fin whale (*Balaenoptera physalus*), harbor porpoise (*Phocoena phocoena*), hooded seal (*Cystophora cristata*) and Atlantic white-sided dolphin (*Lagenorhynchus acutus*) are summarized in Fig. 1 and Table 1. In general, either length and/or teeth parameters were used for age determination in order to produce comparable pooled samples. The pools combined individuals of similar age/size and of a single sex in species where sex is known to influence the contaminant concentration. In order to minimize the variability stemming from age and sex-related processes, it was decided to perform the chemical analyses primarily on samples of males, and preferentially within a fixed age/size group. As a means also to minimize the variance in the data without increasing the number of samples to be analyzed, it was decided to analyze pooled samples, comprising 3–5 individuals of a predefined age/sex. Pooled samples can be used to reduce the biological variation, which is advantageous in the sense that the reduction of within group variation will increase the sensitivity of the experiment to changes between groups. Samples of Atlantic white-sided dolphin and pilot whale were taken in connection with the traditional drive hunts by sampling crews of the Environment agency and the Museum of Natural History in the Faroe Islands. The samples are part of larger collections of biological materials taken in the yearly drive fishery. Samples of minke whales were obtained by licensed whalers from southwest and central West Greenland between May and October 1998. The minke whales caught along the Norwegian coast were part of a scientific catch for the Marine mammal's program funded by the Research Council of Norway and obtained from the bio-bank at the Norwegian School for Veterinary Science (NVH). The whales were caught using a transect method by which the ship follows a certain predefined route, aiming at estimating the minke whale stock. The minke whales from Icelandic waters were derived from a scientific program carried out under the auspices of the Ministry of Fisheries in Iceland during 2003–2007. Altogether 200 animals were caught during these five years from all around Iceland by random sampling from small defined areas. Blubber samples of female hooded seal were made available from the bio-bank at the Norwegian School of Veterinary Science and from the institute of

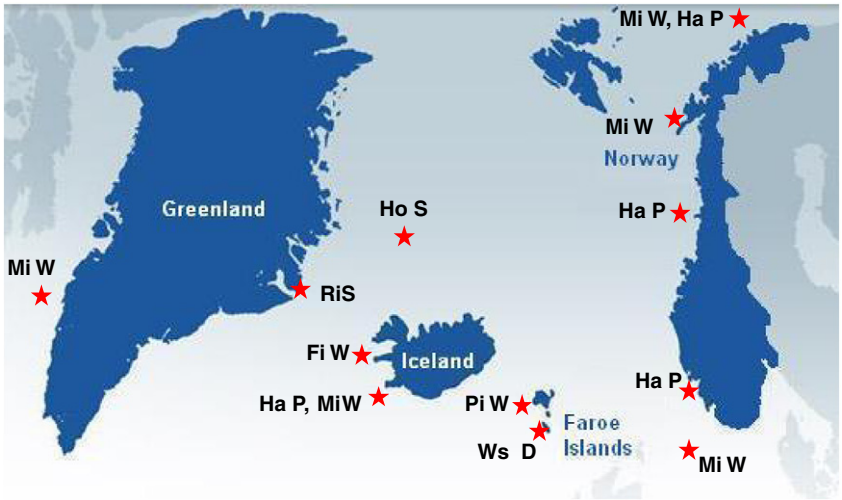

Fig. 1. Indicative map showing sampling locations of pilot whales (Pi W), minke whales (Mi W), hooded seals (Ho S), ringed seals (Ri S), harbor porpoise (Ha P), fin whales (Fi W) and white-sided dolphins (Wh d).

Table 1
Sample characteristics of seven marine mammal species sampled around Faroe Islands, East Greenland, Norway and Iceland.

Species	Geographic location	Sampling years	Sex	Tissue	No./pool	No. of pools/year	∑ Pools
Pilot whale	Faroe Islands	1986, 1997, 2006/2007	Male	Blubber	3–5	3	9
WS dolphin	Faroe Islands	1997, 2001, 2006	Male	Blubber	3–5	3	9
Ringed seal	E Greenland	1986, 2000, 2006	Male/Female	Blubber	4–5	3	9
Minke whale	W Greenland	1998	Female	Blubber	4	3	3
Hooded seal	West Ice	1990, 1997, 2007	Female	Blubber	4–5	3	9
Minke whale	Norway	1993, 1999	Male	Blubber	5–6	3	6
Harbor porpoise	Norway	2000	Male	Blubber	5	3	3
Minke whale	W Iceland	2003–2006	Male	Blubber	5	3	3
Fin whale	W Iceland	86–89, 2006	Male/Female	Blubber	3	2	4
		86–89, 2009	male	Blubber	5	3	6
Harbor porpoise	W Iceland	1992, 1997	Male	Blubber	5	3	6

marine research (IMR). The seals were shot on the ice, and immediately dissected for biological sampling. Samples of juvenile male and female ringed seals were obtained by local hunters from Ittoqqortoormiit, central East Greenland. The hunting mainly took place in May and June and the seals were either shot on the ice or caught in monofilament nets under the ice. Samples of blubber from harbor porpoises from Iceland were provided by the bio-bank at the Marine Research Institute of Iceland (MRI). The samples were from by-catches collected in south west Iceland for scientific purposes in the years 1992 and 1997 (March–December). Samples of blubber from harbor porpoises from Norway were made available by the bio-bank at IMR. The animals were by-caught in coastal fisheries in March, April, and June 2000 at three locations along the Norwegian coast. The fin whales were from catches within the Icelandic scientific program on fin whales and sei whales (*Balaenoptera borealis*) in Icelandic waters conducted by the Marine Research Institute of Iceland (MRII). All samples were stored at −20 °C until analysis.

2.2. Chemicals

All organic solvents (methanol, n-hexane, dichloromethane, toluene) used were of pesticide grade and purchased from Sigma Aldrich (Steinheim, Germany). Anhydrous sodium sulfate (Na_2SO_4) were obtained from Fluka (Steinheim, Germany), silica gel 60 (70–230 mesh) and sulfuric acid (H_2SO_4) of pro analysis grade from Merck (Darmstadt, Germany), potassium hydroxide (KOH) (reagent grade pellets, Ph Eur) from Scharlau (Barcelona, Spain). Prior to use the silica gel was washed with methanol and dichloromethane and activated and stored in 120 °C. Tetradecane was used as a keeper and obtained from Sigma Aldrich (Steinheim, Germany). The ^{13}C-labeled internal standard (BDE #77) was purchased from Cambridge Isotope Laboratories Inc. (Andover, MA, USA), and ^{13}C #139 was made available by Wellington Inc. (Guelph, Canada). Individual native PBDEs congeners #28, #47, #66, #85, #99, #100, #138, #153, #154, #183 mixed into a quantification standard were also made available from Wellington Laboratories. ^{13}C-labeled PCB congeners #81, #114 and #178 were used as recovery standards and were provided by Cambridge Isotope Laboratories Inc. (Andover, MA, USA).

2.3. Sample preparation and analytical determination

10–20 g of pooled blubber made up from 3 to 6 individuals (Table 1) was homogenized in a mortar with anhydrous sodium sulfate and approximately 5 g of the homogenate was transferred to glass columns (18 mm diameter). The internal standards were added and the lipids were eluted with hexane/dichloromethane (1:1, v/v). After solvent evaporation using low-pressure rotary evaporation, the lipid contents were determined gravimetrically. Sample clean up was performed using a multilayer silica column (18 mm diameter) containing KOH silica gel, neutral activated silica, 40% H_2SO_4 silica gel, 20% H_2SO_4 silica gel, neutral activated silica gel and activated Na_2SO_4. The analytes were eluted with hexane. Prior to instrumental analysis the ^{13}C-labeled PCB-mix used as recovery standards were added. Individual PBDEs congeners #28, #47, #66, #85, #99, #100, #138, #153, #154, #183 (∑PBDEs) were analyzed by an Agilent 6890 GC coupled to a low-resolution mass spectrometer (Agilent 5975) operating in the NCI mode monitoring m/z 79 and m/z 81. The recovery standard of PCBs #81, #114 and #178 were monitored at m/z 408. The negative chemical ionization-MS (NCI-MS) operating mode may in many instances be a more sensitive technique than EI-MS for the determination of PBDEs, although the Br-ions (m/z = 79 and 81) used as quantifier and qualifier are less selective compared with the ions formed in EI (Covaci et al., 2007). ^{13}C-labeled BDE #77 and BDE #139 were used as internal standards because they can both be used in the NCI mode for most biota samples (Athanasiadou et al., 2008; Karlsson et al., 2006), as well as in the EI mode if co-elution occurs. The samples of white-sided dolphins were analyzed by EI-MS due to a co-eluting peak with that of one of the internal standards (BDE #77). The same chromatographic set-up and parameters as in the NCI mode were used and the analysis was performed in the selective ion recording mode (SIR), monitoring the two most abundant ions of the molecular bromine cluster for QA/QC purposes. Splitless injection was used to inject 1 μl of the final extract and quantification was carried out using the internal standard method. The column used for separation of analytes was a 30 m SGE DB-5 (0.25 mm, 25 μm) and the temperature program was set to initial 180 °C for 2 min, ramped 15 °C/min to 205 °C, 2 °C/min to 251 °C and 6 °C/min to 325 °C.

2.4. Quality assurance

PBDE congeners #28, #47, #66, #100 and #99 were quantified against ^{13}C-labeled BDE #77 and PBDE congeners #85, #154, #153, #138, #183 against ^{13}C-labeled BDE #139. The identification of PBDEs was based on accurate isotope ratio and retention time. The recovery ranged between 55 and 100% in 67% of the samples and 101–130% in 31% of the samples. Ten of the calculated recoveries were either below 50% or above 150% and those samples were hence quantified with a higher uncertainty (Table 2). With every batch of 6–9 samples extracted, an extraction blank was also prepared and analyzed as well as monitoring instrumental blanks of toluene. Detection limits were set to three times the signal to noise (S/N) and calculated for each sample individually. Repeatability was assessed by spiking experiments of six replicates of a whale blubber sample on two separate days, resulting in a relative standard deviation (RSD) between 9% and 29%. Reproducibility was calculated from four individual analyses of a blubber sample on different days. The RSD was calculated to between 15% and 33%, except for BDE #85 and BDE #138 where levels were close to the detection limit.

Table 2
Temporal trends in PBDE concentrations in marine biota.

Species and location	Years	Trend	Reference
Ringed seal CW Greenland	1982–2005	Increasing levels	Vorkamp et al., 2008
Ringed seal W Canada	1981–2000, 2005	Increasing levels, leveling off in 2005	Ikonomou et al., 2002, 2005
Ringed seal E Greenland	1986–2004	No significant trend	Riget et al., 2006
Ringed seal Svalbard	1998 and 2004	Decrease, 70–80% lower in 2004	Wolkers et al., 2008
Beluga Whale Canada	1988–1999	Increasing levels	Lebeuf et al., 2004
Harbor porpoise UK	1992–2008	Peak in 1998, Decrease >50% in 2008	Law et al., 2010
Blue mussels, Seine estuary, France	1981–2003	Increasing 1981–1993, peak 99–2000. Decreasing 2002–2003	Johansson et al., 2006
California Sea lion	1994–2006	No trend	Meng et al., 2009
Harbor seals, NW Atlantic	1991–2005	No trend	Shaw et al., 2008
Northern Fur seals, Japan	1972–1998	Peak 1991–1994, Decrease 50% in 1998	Kajiwara et al., 2004
Ivory gull eggs, Canada	1976–2004	Increasing levels	Braune, 2007
Indo-pacific dolphin and Finless porpoise, Hong Kong	1997–2008	No trend	Lam et al., 2009

2.5. Data treatment

In order to make the results comparably across species and geographical area approximately the same number of individuals contributing approximately the same volume to each pool was chosen. The measured contaminant concentration of a pooled sample can be considered as an arithmetic average of levels in the individual samples making up the pool, assuming an equal sample size. If the contaminant concentrations in the population are normal distributed, pooled-sample concentrations will also be normal distributed with the same mean but reduced variance compared to individual measurements. However, contaminant concentrations are often better described by a lognormal distribution than a normal distribution which leads to biased estimates of the central tendency of the samples making of the pool (Caudill, 2010). Due to lack of knowledge of the variability within the pools it was not possible to correct this bias (Caudill et al., 2007). Preliminary data scrutinizing indicate that contaminant concentrations were more likely log-normal distributed than normal distributed even though samples were pooled and data were therefore log-transformed prior to statistical tests. One-sided analyses of variance (ANOVA) followed by Tukey comparisons of means were applied to test for differences between time periods and sampling areas. A significance level of 5% was used when evaluating test results.

3. Results and discussion

3.1. PBDE levels and species differences

PBDE concentrations were examined in pooled blubber samples of pilot whale, ringed seal, fin whale, hooded seal, Atlantic white-sided dolphin, minke whale and harbor porpoise covering a time period of 23 years (1986–2009). The highest PBDE levels were found in samples from the late 1990s or beginning of 2000 in the toothed whale species pilot whale and white-sided dolphin, and the lowest levels in fin whales and ringed seals (Fig. 3). Fin whales most commonly have lower contaminant levels than those of toothed whales since they are macroplanktophages and feed at mid-trophic level (Fossi et al., 2003). Two pools of fin whales, comprising two females and one male caught in 1986–1989 and 2006, were separated by age (12–20 years and 26–35 years). In both sampling periods the highest total PBDE concentration was found in the pool with the older individuals, 5 and 12 ng/g lw (1986–1989) and 20 and 31 ng/g lw (2006). No significant difference in PBDE concentration was found compared to the pools from 1986 to 1989 (8–9 ng/g lw) and 2009 (19–27 ng/g lw), comprising only male individuals. The PBDE levels measured in pilot whales from 1997, range 568–2428 ng/g lw, are among the highest reported in arctic marine mammals. The values are in accordance with a study on pilot whales sampled in 1996 (Lindström et al., 1999). In Atlantic white-sided dolphins, the median ΣPBDE concentrations were 230, 673 and 249 ng/g lw in 1997, 2001 and 2006, respectively. This was lower than in males stranded in Massachusetts between 1993 and 2000, where a mean value of 1820 ng/g lw was reported (Tuerk et al., 2005). The median ΣPBDE concentration in ringed seal blubber from East Greenland ranged from 20 ng/g lw (1984) to 34 ng/g lw (1998). This is comparable to levels measured in ringed seals sampled in the same area between 1986 and 2004 (Riget et al., 2006). Compared to PBDE levels found in ringed seals from Arctic Canada, the levels measured in the present study are one order of magnitude higher (Ikonomou et al., 2002) and slightly higher than those found in ringed seals from Svalbard collected in 1998 (Wolkers et al., 2004). Higher PBDE concentrations were found in the hooded seals taken on the Greenland Sea pack ice compared to the ringed seals, between 38 ng/g lw and 148 ng/g lw. The PBDE concentration range was relatively narrow in harbor porpoises caught in Icelandic waters in 1992 and 1997, between 67 and 98 ng/g lw. The harbor porpoises sampled in 2000 at three different locations along the coast of Norway displayed total PBDE concentrations between 71 and 540 ng/g lw, with the lowest

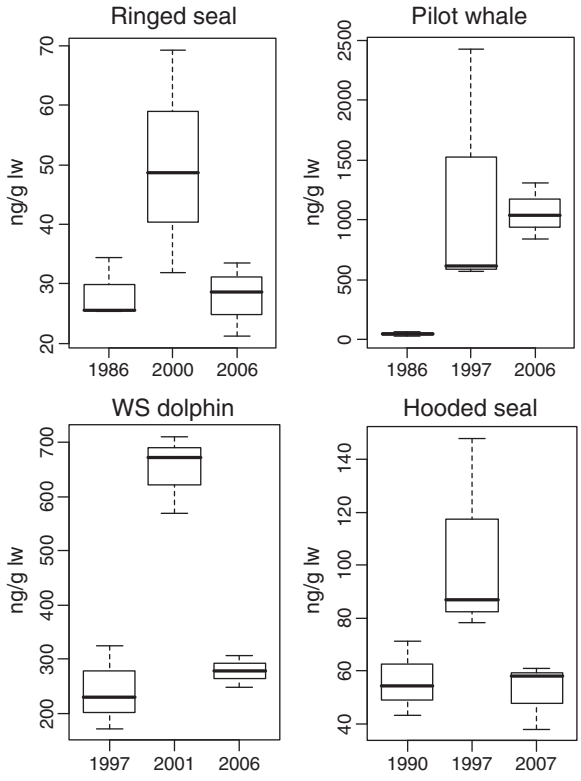

Fig. 2. ΣPBDE concentrations (ng/g l.w.) in pooled blubber samples of ringed seals, pilot whales, Atlantic white-sided dolphins and hooded seals (n = 3). Solid bars indicate median concentrations and max and min values are shown by whiskers.

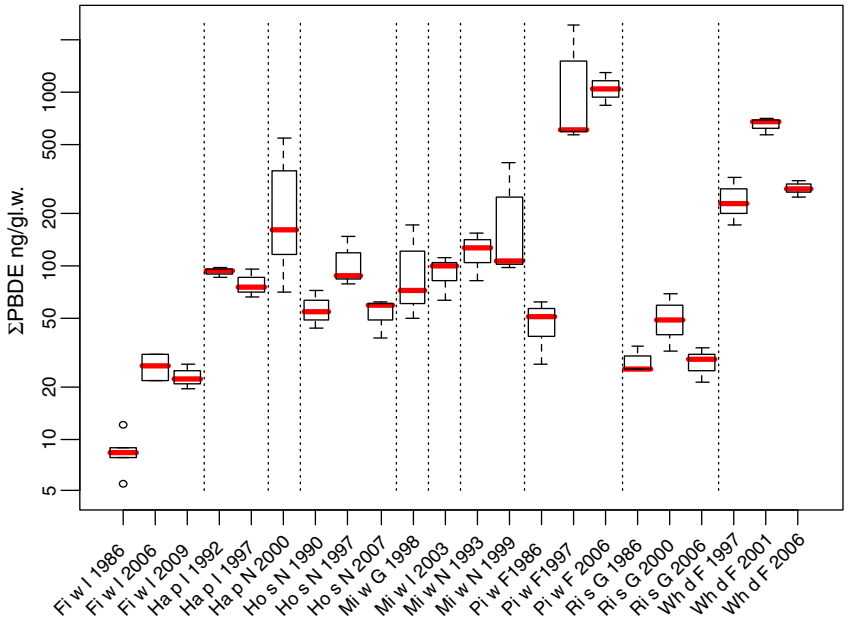

Fig. 3. Inter- and intra-species comparison of Σ PBDE concentrations in fin whales (Fiw I) from Iceland, harbor porpoises (Ha P I, Ha P N) from Iceland and Norway, hooded seals (Ho s N) from Norway, minke whales (Mi W I, Mi W N) from Iceland and Norway, pilot whales (Pi W F) from Faroe Islands, ringed seals (Ri s G) from East Greenland and white-sided dolphins (Wh d F) from Faroe Islands. Solid bars indicate median concentrations and max and min values are shown by whiskers.

levels found in the most northern part (Finnmark). Compared to harbor porpoises stranded in the U.K between 1992 and 2008, displaying median total PBDE levels between 320 and 2230 ng/g lw, the levels from this study were considerably lower (Law et al., 2010). The range of PBDE levels in minke whales caught off Norway, Greenland and Iceland were 82–389 ng/g lw, 50–170 ng/g lw and 64–111 ng/g lw, respectively. These levels were slightly lower compared to minke whales caught of the Korean coast (Moon et al., 2010).

3.2. PBDE congener distribution

The median, min and max lipid normalized PBDE concentrations based on three pooled blubber samples are shown for all individual PBDE congeners in Table 3. BDE congeners #47, #99, #100, #153 and #154 were found in all analyzed samples. BDE #28, #66 and #85 were found in 70–90% of the samples. BDE #138 and #183 were found close to the limit of detection in 30–50% of the samples. PBDE concentrations, as well as the congener distribution in relation to ΣPBDE, varied between the different species. The congener distribution is influenced by species-specific metabolic capacities as well as different feeding habits. BDE #47 was dominating in all species, with an exemption of two pools of hooded seals, followed by #99, #100, #153 and #154. In ringed seals BDE #47 accounted for 80% of total PBDE concentrations, whereas for only 30–40% in white-sided dolphins and hooded seals. In hooded seals BDE #99 was most prevalent in the samples from 1990 and 1997. However, in 2007 the distribution had changed in favor of BDE #47 (Fig. 4). In pilot whales and harbor porpoises caught off Iceland the relative exposure of BDE #154 decreased over the sampling period. However, in minke whales caught along the

Table 3
Median, min and max ∑ PBDE concentrations and min and max concentrations for 10 BDE congeners (ng/g lw) in pooled blubber samples (n = 3) of seven marine mammal species. I = Iceland, N = Norway, G = Greenland.

Species	Years	∑ PBDE	# 28	# 47	# 66	# 100	# 99	# 85	# 154	# 153	# 138	# 183
Pilot whale	86	51 (27–61)	1.4–4.2	11–37	0.3–1.4	1.7–5.8	1.9–4.2	0.5–1.0	7.2–9.5	0.7–1.1	<0.1	<0.1–0.2
	97	613 (568–2428)	11–42	304–1389	19–74	58–207	100–430	8.8–37	34–152	21–94	<0.1	0.7–3.3
	06/07	1041 (840–1307)	16–21	489–711	25–26	93–151	121–185	21–49a***	49–124a***	24–40a***	<0.1a***	0.6–1.2a***
WS dolphin	97	230 (172–325)	<0.04–0.1	70–110	1.6–2.6	39–60	41–73	1.2–2.5	4.2–53	15–23	0.1–1.9	NQ
	01/02	673 (570–710)	<0.1–0.2	204–221	3.0–5.7	118–160	111–160	2.7–5.4	88–108	38–64	0.1–2.1	NQ
	06	279 (249–308)	<0.04–<0.1	94–112	1.5–2.2	51–63	36–54	1.6–2.2	43–53	11–22	0.3–0.6	NQ
Hooded seal	90	54 (44–71)	0.3–0.4	15–19	0.2–0.3	3.2–4.8	16–28	<0.1b*	5.5–12b*	2.8–5.6b*	<0.2–0.4b*	0.2–0.3b*
	97	87 (78–148)	0.4–0.7	25–47	0.5–0.9	4.9–9.2	27–61	<0.1	11–15	9.1–14	<0.2–0.4	0.4
	07	59 (38–61)	0.2	19–21	0.2–0.3	1.9–3.7	7.2–15	<0.1–<0.3	5.4–12	4.3–10	<0.1–<0.4	0.1–0.3
Ringed seal	86	25 (25–34)	0.4–0.6a*	18–28a*	<0.04–0.1a*	1.0–2.1a*	2.5–3.1a*	<0.1–0.3	1.0–1.3	0.3–0.5	<0.1	<0.1–0.2
	00	49 (32–69)	1.2–1.6	23–55	0.3	2.3–3.7	3.2–5.5	0.3–0.8	0.6–2.8	0.6–2.1	<0.1	<0.1
	06	28 (21–33)	0.6–0.9	16–28	<0.1–0.2	1.5–2.0	1.5–2.9	<0.1–0.5	0.3–0.7	0.3–0.6	<0.1–0.1	<0.1
Harbor porpoise	92 I	94 (85–98)	0.9–1.1b*	38–52b*	0.7–1.2b*	8.0–11b*	9.2–19b*	<0.2–<0.8b*	8.9–16b*	2.8–8.4b*	<0.3–<0.6b*	<0.2–0.4b*
	97 I	75 (67–96)	0.8–0.9	43–59	0.6–0.7	6.4–9.3	9.1–13	<0.1	4.4–8.9	1.8–3.2	<0.2	<0.1–0.1
	00 N	161 (71–540)	0.8–1.2	53–301	0.4–4.2	7.3–56	4.5–87	<0.2	4.1–46	0.8–44	<0.1–<0.2	<0.1–0.5
Minke whale	93N	126 (82–153)	1.1–2.1	54–101	1.2–1.8	5.7–13	14–29	0.8–1.0	3.2–6.1	1.4–3.0	<0.1–0.1	<0.1
	99N	105 (97–389)	0.8–1.6	59–212	0.8–5.4	7.5–30	14–88	1.7–3.8	4.3–27	1.0–20	<0.1–0.9	<0.1–0.4
	98G	71 (50–170)	1.3–3.1	29–69	0.6–3.6	4.0–14	9.6–63	0.3–0.6b***	3.6–10b***	0.9–5.4b***	0.1b***	0.1–0.2b***
	03/06 I	99 (64–111)	0.7–1.4	41–68	0.1–0.5	3.9–8.0	7.1–17	2.5–5.7	6.3–13	1.5–2.9	<0.1–0.1	0.1–0.2
Fin whale	86/89	8.4 (5–12)	0.1–1.1	2.4–5.1	<0.1–0.1	0.3–0.8	1.0–2.4	<0.1–0.3	0.6–2.4	0.2–1.5	<0.2–0.2	<0.1–0.1
	06/09	22 (20–31)	0.2–4.0	7.6–13	0.1–0.2	0.7–1.9	1.3–5.5	0.2–0.6	2.7–5.7	0.9–1.5	0.1	0.2–0.3

Quantified with a higher uncertainty due to recovery [a]<50%, [b]>150%. * One pool. *** Three pools. NQ = Not quantified.

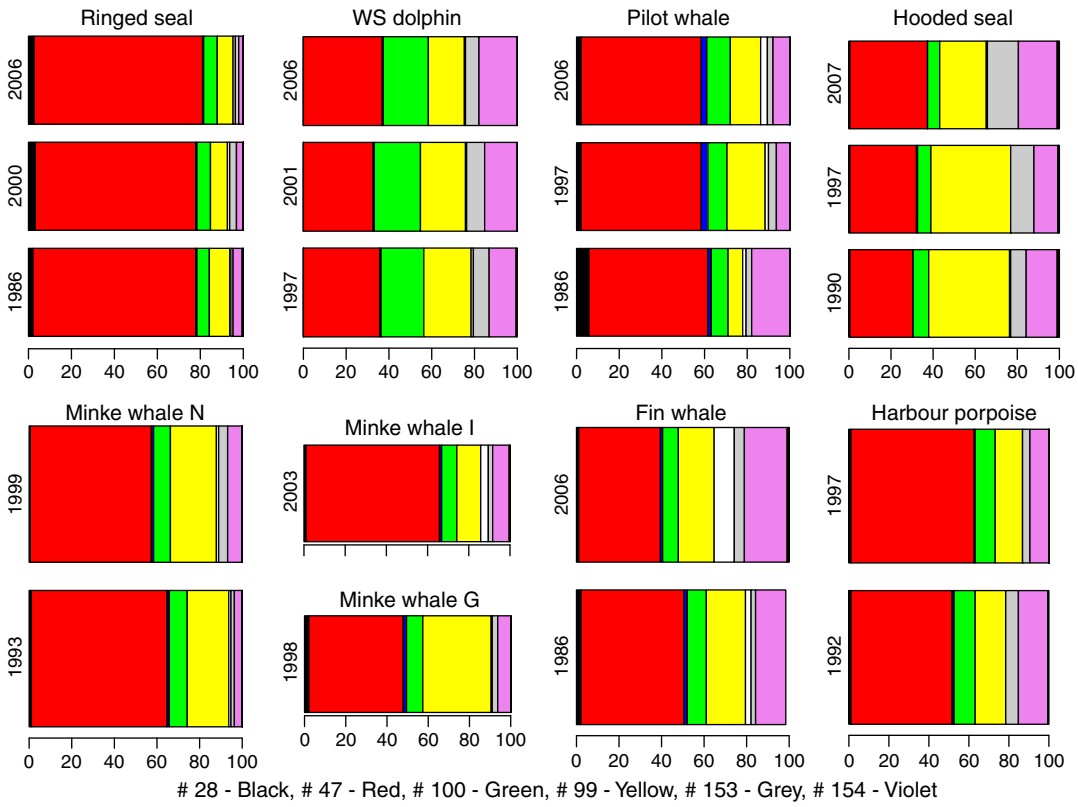

Fig. 4. Congener distribution in percentage of ∑ PBDE for BDE #28, #47, #100, #99, #153 and #154 indicated by colors. N = Norway, I = Iceland and G = Greenland.

Norwegian coast, fin whales and hooded seals there seemed to be an increase in the levels of BDE #153 and #154 relative to the total PBDE concentration in the most recent sampling period. A relative increase of BDE #154 was also indicated in the samples of white-sided dolphins from 2006, which may indicate an increased relative exposure to higher brominated congeners (Fig. 4). A more rapid increase of BDE #154 compared to BDE #47 has previously been observed in burbot from the Canadian Arctic between 1988 and 2006, and an increase of relative proportions of BDE #153 and #154 was observed in harbor porpoise between 1992 and 2008 (de Wit et al., 2010; Law et al., 2010). The difference in congener distribution observed in female minke whales from Greenland compared to male minke whales from Norway and Iceland may partly be attributed to gender and different sources of exposure. The use of the different commercial PBDE technical mixtures with different congener compositions have altered between countries as well as over time. In herring gull eggs from the Laurentian great lakes in Canada PBDE concentrations and congener pattern dramatically changed between 1995 and 2006. An increase of congeners deriving from mainly Penta- and OctaBDE mixtures was observed until the year 2000 but not beyond, while DecaBDE continuously increased, possibly reflecting the increasing regulation and phasing out of PentaBDE and OctaBDE mixtures and shift towards increasing use of the Deca-BDE mixture (Gauthier et al., 2008). Changes in diet over time, for example as a result of climate change or prey availability, may also reflect changes in contaminant load and congener profile (Fontaine et al., 2007; Law et al., 2010; Mckinney et al., 2009).

3.3. Temporal and spatial concentration differences

Differences in PBDE concentrations between three sampling periods, from 1986 to 2007, were studied in pooled blubber samples of pilot whales, ringed seals, white-sided dolphins and hooded seals (Fig. 2). The highest PBDE levels were found in samples from the late 1990s or beginning of 2000. Samples of fin whales were only available from the end of the 1980s and 2006–2009, and the highest PBDE levels were found in the pools from the most recent years. These findings possibly reflect the increase in the global production of technical PBDE mixtures in the 1990s. However, all the studied species undertake relatively extensive migrations except ringed seals, which are believed to be relatively sedentary (Born et al., 2004), hence the influence of multiple ecological factors make the interpretation of temporal as well as spatial patterns of variation complex (Aguilar et al., 2002). In ringed seals caught off West Greenland, the difference in PBDE concentration observed in the samples from 2000 compared to 1986 was significant for BDE #28, #66 and #153. No trend in PBDE concentrations was found for ringed seal from the same area sampled between 1984 and 2004 in a study by Rigét et al. (2010). However, a rapid increase in PBDE levels was seen in ringed seals between 1982 and 2005 from West Greenland and between 1981 and 2000 in ringed seals from the Canadian Arctic (Ikonomou et al., 2002; Vorkamp et al., 2008). In ringed seals from Svalbard the levels decreased between 1996 and 2004 (Wolkers et al., 2008). In hooded seals, the concentrations of BDE #28, #47, #66, #99, and #153 measured in the samples from the middle sampling period in 1997 were significantly different from the concentrations measured in the seals sampled in 2007. As oppose to ringed seals, hooded seals often migrate long distances between their breeding and molting periods. Satellite tagging programs have revealed that individual hooded seals made long excursions to areas around Faroe Islands, Iceland and North Norway, as well as being relatively sedentary in the drift ice waters of the Greenland Sea (Folkow et al., 1996; Haug et al., 2007). Thus contaminant concentrations in hooded seals are likely reflecting exposure to multiple sources in the North Atlantic. By comparing the median values, a 12–16 fold increase in PBDE levels was found in pilot whales between 1986 and the two later sampling points in 1997 and 2006. Pilot whales often form schools while migrating and this may potentially lead to large differences in chemical body burden

depending on the migratory paths. Differences in pollutant load can be attributed to different food availability and different trophic levels of the prey eaten by the various pods (Aguilar, 1987). High variability between POPs in various schools of pilot whales around the Faroe Islands have been described previously (Aguilar et al., 1993; Dam and Bloch, 2000). The 1986 and 2006/07 data represent three schools or more and therefore this high variability may be regarded as being part of the trend. However, the high data variability makes it difficult to conclude whether PBDE levels have increased, decreased or leveled off in recent years. In white-sided dolphins, the peak in PBDE levels observed in 2001 was significantly different from levels found in 1997 and 2006 for all congeners except #154. Although temporal trends of PBDE levels in biota published in recent years are not consistent, various contaminant studies indicate that increasing PBDE levels observed in the last decades have stabilized or leveled off in recent years (Table 2). No significant difference was found in total PBDE concentrations between the pools of minke whales from Iceland, Norway and Greenland (ANOVA, $p = 0.33$), or between harbor porpoises caught off Iceland and Norway (ANOVA, $p = 0.26$). However, given the differences in sampling years and in the case of minke whales also gender, only general comparisons were possible. The highest PBDE levels were found in minke whales and harbor porpoises caught off Norway at one of the more southern locations. Levels of POPs commonly decrease with latitude moving up from the North Sea toward the Arctic, which may result in higher exposure to animals that spend longer periods feeding at southern latitudes (Jasmin et al., 2010). Specific patterns can vary widely within wildlife populations as a result of differences in habitat, ranging patterns, and diet. Different stocks of marine mammals have been distinguished based on their contaminant load, for example minke whales (Hobbs et al., 2003), ringed seals (Weis and Muir, 1997) and long-finned pilot whales (Aguilar et al., 1993). Stomach content analyses indicate that the harbor porpoise diet varies between neighboring areas in Norwegian and adjacent coastal waters, and that this variation in diet composition could be related to local habitat characteristics (Aarefjord et al., 1995; Fontaine et al., 2007). The variation in diet has also been confirmed by that harbor porpoises from the southwest coast of Norway have significant differences in stable isotope compositions and cadmium concentrations compared to animals from more northern waters (Nordland and Finnmark), suggesting that harbor porpoises do not migrate extensively along the Norwegian coast (Fontaine et al., 2007). Habitats in proximity to point sources of pollution have been shown to contribute to variations in individual contaminant patterns in bottlenose dolphins (Yordy et al., 2010). Stomach content analyses of krill as well as cod show that minke whales feed on different trophic levels. Hence regional differences in availability of prey probably affect their accumulation of contaminants (Hobbs et al., 2003). Although the winter distribution of minke whales is not fully elucidated, they most likely migrate along the European coast in the autumn to southern latitudes (Glover et al., 2010). Animals have been spotted as far south as off the coast of Senegal, and such long-range movements may influence the exposure to contaminants (Van Waerebeek et al., 1999).

3.4. Toxicological implications

PBDEs have a potential to interfere with liver enzyme production and the thyroid hormone system as well as induce immunotoxicity (Darnerud, 2003; Frouin et al., 2010; Hall et al., 2003), However, pilot studies involving a small number ($n = 5$) of pilot whales with ΣPBDE in the range 63–217 ng/g lw in liver, whereof 11–77 ng/g lw were BDE #47, could not reveal any correlation between ΣPBDE and thyroid hormones analyzed; free and total tri- and tetraiodothyronine (Dam et al., 2002). It is unclear if the current levels will have any toxic effects on the studied populations, partly due to insufficient knowledge about other effect variables. In a recently published article on exposure and effects of POPs in arctic wildlife by Letcher et al. (2010), the general lack of basic ecological and physiological information is identified as one obstacle to assess potential changes caused by contaminants, as well as that most effect thresholds established for POPs have been generated using non-arctic animals. Arctic mammals are exposed to a complex mixture of both traditional and new emerging pollutants and are affected by other environmental stressors such as climate change, which makes it difficult to draw conclusions about exposure from biomarker studies.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.envint.2011.07.001.

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