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Polychlorinated naphthalenes (PCNs) in sub-Arctic and Arctic marine mammals, 1986–2009

Anna Rotander [a,*], Bert van Bavel [a], Frank Rigét [b], Guðjón Atli Auðunsson [c], Anuschka Polder [d], Geir Wing Gabrielsen [e], Gísli Víkingsson [f], Bjarni Mikkelsen [g], Maria Dam [h]

[a] Man-Technology-Environment (MTM) Research Centre, Örebro University, SE-701 82 Örebro, Sweden
[b] Department of Bioscience, University of Aarhus, Box 358 DK-4000 Roskilde, Denmark
[c] Innovation Center Iceland, Dept. of Analytical Chemistry, Keldnaholti, 112 Reykjavik, Iceland
[d] Norwegian School of Veterinary Science, Department of Food Safety and Infection Biology, P.O. Box 8146 Dep, NO-0033 Oslo, Norway
[e] Norwegian Polar Institute, FRAM Centre, NO-9296 Tromsø, Norway
[f] Marine Research Institute, Skúlagata 4, 101 Reykjavík, Iceland
[g] Museum of Natural History, Fútalág 40, FO-100 Torshavn, Faroe Islands
[h] Environment Agency, Pob 2048, FO-165 Argir, Faroe Islands

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ABSTRACT

A selection of PCN congeners was analyzed in pooled blubber samples of pilot whale (*Globicephala melas*), ringed seal (*Phoca hispida*), minke whale (*Balaenoptera acutorostrata*), fin whale (*Balaenoptera physalus*), harbour 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). A large geographical area of the North Atlantic and Arctic areas was covered. PCN congeners 48, 52, 53, 66 and 69 were found in the blubber samples between 0.03 and 5.9 ng/g lw. Also PCBs were analyzed in minke whales and fin whales from Iceland and the total PCN content accounted for 0.2% or less of the total non-planar PCB content. No statistically significant trend in contaminant levels could be established for the studied areas. However, in all species except minke whales caught off Norway the lowest ∑PCN concentrations were found in samples from the latest sampling period.

1. Introduction

Polychlorinated naphthalenes (PCNs) are a group of 75 compounds that have been manufactured as complex technical mixtures with more or less the same industrial applications as those of polychlorinated biphenyls (PCBs). PCNs have for example been used in cable insulation, wood preservation and as engine oil additives. The largest volume products used in Europe were Nibren waxes made by the German company Bayer. Bayer was producing between 100 and 200 t/y at the beginning of the 80s but ceased its production in 1983 (NICNAS, 2002). In the United States, the production of the largest volume PCN technical product, commercially marketed as Halowax, was ceased in 1980. The total PCN production has been estimated to be no less than 10% of the global PCB production, i.e. roughly 150,000 tonnes in total (Brinkman and De Kok, 1980). PCNs occurred as impurities in commercial PCB formulations, but not in distinct patterns since the ratio varied among the various PCNs in such blends (Yamashita et al., 2000). Yamashita and co-workers estimated that the PCN load emitted from the use of PCBs was less than 1% of the total production of PCN in Halowax.

Environmental sources consist of evaporation from products containing PCNs and PCBs, and release during combustion (Falandysz et al., 1997). Release of PCNs also occurs due to combustion from a range of industrial and waste incineration processes (Abad et al., 1999; Takasuga et al., 2004). PCNs have potential for long-range transport and are, like other persistent organic pollutants (POPs), globally distributed. PCN congeners, dominated by the tri- and tetrachlorinated homologues, have been detected in arctic air (Harner et al., 1998). Several PCN congeners have shown a tendency to bioaccumulate in blubber of beluga whales and ringed seals in the Canadian Arctic (Helm et al., 2002), in fish and mussels in the Baltic Sea region (Falandysz et al., 1997), seabirds from the Norwegian Arctic (Verreault et al., 2005; Gabrielsen and Pusch, unpublished), and in killer whales from the Northeastern Pacific Ocean (Rayne et al., 2004). Total PCNs previously reported in Arctic marine mammals ranged between 0.02

* Corresponding author.
E-mail address: anna.rotander@oru.se (A. Rotander).

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and 27 ng/g lw and followed the order: harbour seal ~ pilot whale ≥ polar bear > beluga > ringed seal ~ Weddell seal, (Bidleman et al., 2010).

Although examinations of the presence of PCNs in polar environments have increased in recent years, there are still relatively few studies. In the current study, we monitor the occurrence of PCNs in blubber samples of whales, seals and dolphins sampled over a 23-year period (1986–2009). A broad range of marine species is covered from Iceland, Faroe Islands, Norway, West Ice and Greenland.

2. Materials and methods

2.1. Sampling

Information on the chemicals and analytical standards used can be found in the supporting information. Blubber samples of pilot whales (*Globicephala melas*), ringed seals (*Phoca hispida*), minke whales (*Balaenoptera acutorostrata*), fin whales (*Balaenoptera physalus*), harbour porpoises (*Phocoena phocoena*), hooded seals (*Cystophora cristata*), and Atlantic white-sided dolphins (*Lagenorhynchus acutus*) were obtained from different specimen banks in Faroe Islands, Greenland (Denmark), Norway and Iceland (Fig. 1 and Table 1). Details of catching and the institutions providing the samples were described earlier (Rotander et al., 2012). The samples were pooled based on a strict protocol (Dam et al., 2011). 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. 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.

2.2. Sample preparation and analytical determination

Pooled blubber samples (10–20 g) made up from 3 to 6 individuals (Table 1) were homogenized in a mortar with anhydrous sodium sulphate (5*blubber mass), 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 multi-layer 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-labelled PCB-mix used as recovery standards were added. The analysis of individual PCN congeners (IUPAC numbers) 13 (Tri), 28/36, 27, 48, 46 (Tetra), 52, 50, 53 (Penta), 66, 69, 72 (Hexa), 73 (Hepta) and 75 (Octa) was performed by HRGC/HRMS using a Micromass Autospec Ultima (Waters Corporation, Milford) operating at 10,000 resolution. The analysis was performed in the selective ion recording (SIR) mode, monitoring the two most abundant ions of the molecular chlorine cluster. 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 the analytes was a 30 m SGE DB-5 (0.25 mm, 25 μm). The temperature program was set to an initial temperature of 180 °C for 2 min, ramped 15 °C/min to 205 °C (held for 5 min), and then ramped 3.7 °C/min to 300 °C.

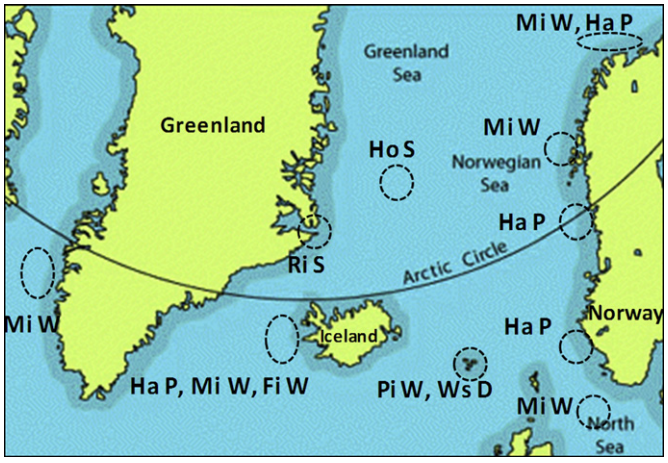

Fig. 1. Indicative map showing sampling locations of minke whales (Mi W), ringed seals (Ri S), hooded seals (Ho S), harbour porpoises (Ha P), fin whales (Fi W), pilot whales (Pi W), and white-sided dolphins (Ws D). Picture adopted from the North Atlantic Research Unit (NARU) at the University of Bradford, UK. http://www.brad.ac.uk/archenvi/research/NARU/Home.php. Copyright© 2008 NARU.

2.3. Quality assurance

PCN congeners 13 (Tri), 28/36, 27, 48, 46 (Tetra), 52, 50, 53 (Penta), 66, 69, 72 (Hexa), 73 (Hepta) and 75 (Octa) were quantified against ^{13}C-labelled PCBs. With every batch of 6–9 samples extracted, an extraction blank was also prepared and analyzed as well as monitoring instrumental blanks of toluene. The identification of PCNs was based on accurate isotope ratio and retention time, and detection limits were set to three times the signal to noise (S/N). Repeatability was assessed by spiking experiments of three replicates of a pilot whale blubber sample on two separate days, resulting in a relative standard deviation (RSD) between 18% and 33%. In each batch two unspiked samples were included, resulting in an RSD between 11% and 30% for the PCN congeners that were found above the limit of detection (LOD). Reproducibility was calculated from three individual analyses, resulting in an RSD between 9% and 37%. The PCN-52 data had an RSD of 51% due to that the levels in the reference sample were close to LOD. The analysis of PCN-48 showed an RSD of 80%. Separation (and thus quantification) of PCN-50 was not possible due to a large co-eluting peak. This is shown in Fig. 2 from spiking a whale blubber sample with 1250 pg of a native PCN mixture. PCN-50 could not be separated from the peak eluting at retention time 8.32 min.

2.4. Data treatment

The data were log-transformed prior to statistical tests since contaminant concentrations were found to more likely be log-normal distributed than normal distributed, even though samples were pooled. Values below LOD included in ∑PCN were divided by two prior to statistical tests. One-sided analyses of variance (ANOVA) followed by Tukey comparisons of means was 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. Occurrence of PCNs

PCN congeners 48, 52, 53, 66 and 69 were found in the blubber samples between 0.03 and 5.9 ng/g lw (Fig. 3 and Table 2). The concentrations for the rest of the targeted congeners were below LOD (<0.02–<0.3). Co-elution has been noted for PCN-48/35, PCN-52/60, PCN-53/55 and PCN-66/67, and therefore contribution to the calculated compound levels of co-eluting PCNs not pre-sent in the native standard mixture cannot be ruled out (Falandysz et al., 2000). The highest PCN levels were found in the three toothed whale species pilot whale, white-sided dolphin and harbour porpoise. Compared to baleen whale species, toothed whales commonly have higher contaminant levels since they feed at a higher trophic level (Fossi et al., 2003). Although PCNs in general are not found to biomagnify (Lundgren et al., 2002; Nfon et al., 2008), some congeners may do so as found for instance in freshwater food webs at Bjørnøya and Lake Ontario (Evenset et al., 2005; Helm et al., 2008). PCN-48 was the congener measured at highest concentrations (0.06–5.9 ng/g lw). However, in 23% of the samples PCN-48 was found below detection limits, including most of the ringed seal samples from East Greenland and all minke whale samples from Norway. Due to the poor QA/QC performance the reported levels of PCN-48 should be viewed as indicative (Table 2). For the same reason PCN-48 was excluded from the evaluation of PCN congener distribution and temporal changes in ∑PCN concentrations. The highest PCN concentrations (∑PCN) were found in the pilot whales and the white-sided dolphins captured around Faroe Islands (1.6–5.2 ng/g lw). The lowest ∑PCN levels were found in the ringed seals from central East Greenland and in the fin whales from Iceland (0.2–0.4 ng/g lw). The levels in the ringed seals, range 0.2–0.8 ng/g lw, were comparable or slightly higher compared to previous studies on ringed seal from Alaska caught between 1993 and 2002, in which the range of ∑PCN reported was 0.04–0.5 ng/g lw (Helm

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

Species	Geographic location	Sampling years	Sex	Tissue	N°/pool	N° 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
Harbour 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
Harbour porpoise	W Iceland	1992, 1997	Male	Blubber	5	3	6

et al., 2002; Muir et al., 2004). In comparison to levels measured in blubber of juvenile male and female harbour seals from the northern Gulf of Alaska (0.9–5.0 ng/g lw) (Wang et al., 2007), the levels in both ringed seals and hooded seals were lower in the present study. Comparing ∑PCN concentrations between different studies should be done with caution since different native standard mixtures commonly have been used in the quantification of PCNs. For example, in the case of the Harbour seals from Alaska ∑PCN was reported as the sum of the concentrations of individual congeners identified in Halowax 1014 referring to PCN congener compositions

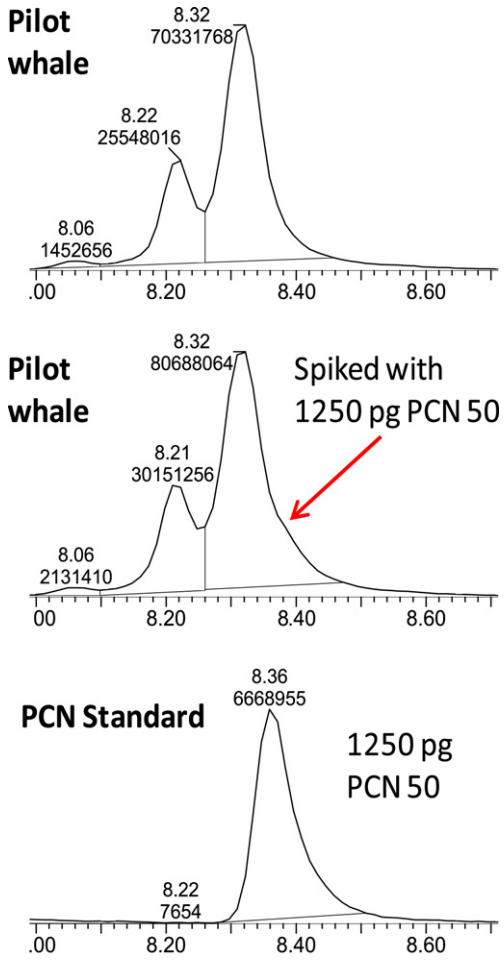

Fig. 2. A pilot whale blubber sample (top HRGC/HRMS chromatogram) and the same blubber sample spiked with 1250 pg of a native PCN mixture (middle chromatogram) compared to a standard injection of a native PCN mixture (bottom chromatogram). The peak in the standard chromatogram at retention time 8.36 shows the PCN-50 congener.

in accordance with the study by Harner and Bidleman (1997). Since fewer congeners were included in the pre-sent study it may be that the total PCN levels in the present study are underestimated compared to the levels reported in the study of harbour seals. PCN-50 could not be separated from a co-eluting compound in any of the samples and was therefore not quantified. The compound corresponding to the interfering peak could possibly be a chlordane of similar molecular mass, which has been shown to co-elute with PCN-50 (Herbert et al., 2005). Large contributions of PCN-50 were found in killer whales from the North Eastern Pacific Ocean and no other studies have presented a similar pattern (Rayne et al., 2004). This indicates that PCN exposure may be highly dependent on species and geographic location.

Comparing contaminant concentrations between different sites is complicated because of variations in environmental conditions. There were no significant differences in ∑PCN concentrations between the minke whales from Norway, Iceland and West Greenland (ANOVA, $p = 0.24$), or between the harbour porpoises samples from Iceland and Norway (ANOVA, $p = 0.55$). In four pooled samples of fin whales and three pooled samples of minke whales from Iceland 25 out of 38 analyzed non-planar PCBs were quantified (Table S1, Supporting Information). In all samples, the PCN content accounted for 0.2% of the non-planar PCB content or less. Similar ratios between PCB and PCN content were found in beluga whales and ringed seal from Baffin Island in the Canadian Arctic sampled in 1993 and 1994, where the maximum concentrations of PCN found in blubber tissue (0.4 ng/g lw in beluga and 0.07 ng/g lw in ringed seal) on average accounted for less than 1% of the sum of eight individual PCBs (Helm et al., 2002).

3.2. PCN congener distribution

There were considerable differences in congener proportions among the whale and seal species. The PCN congener occurring in largest proportions differed both between species and sampling localities (Fig. 4). In general, hexachlorinated PCNs (PCN-66 and PCN-69) were more dominant in baleen and toothed whale species compared to the two seal species. However, this was not the case for harbour porpoises and minke whales sampled off Iceland. Similar differences in congener distribution between seals and whales were also observed in the belugas and ringed seal from Baffin Island, where tetrachlorinated congeners dominated in the ringed seals while the higher chlorinated congeners were more common in the belugas (Helm et al., 2002). In those two species, sampled within short distances from each other and both mainly foraging on arctic cod, the difference in congener profiles was assumed to originate from metabolic differences. Although data on stomach content was not available for this study's species, it may be assumed that they have a more varying diet compared to the ringed seals and belugas both originating from Baffin Island. Hence, the

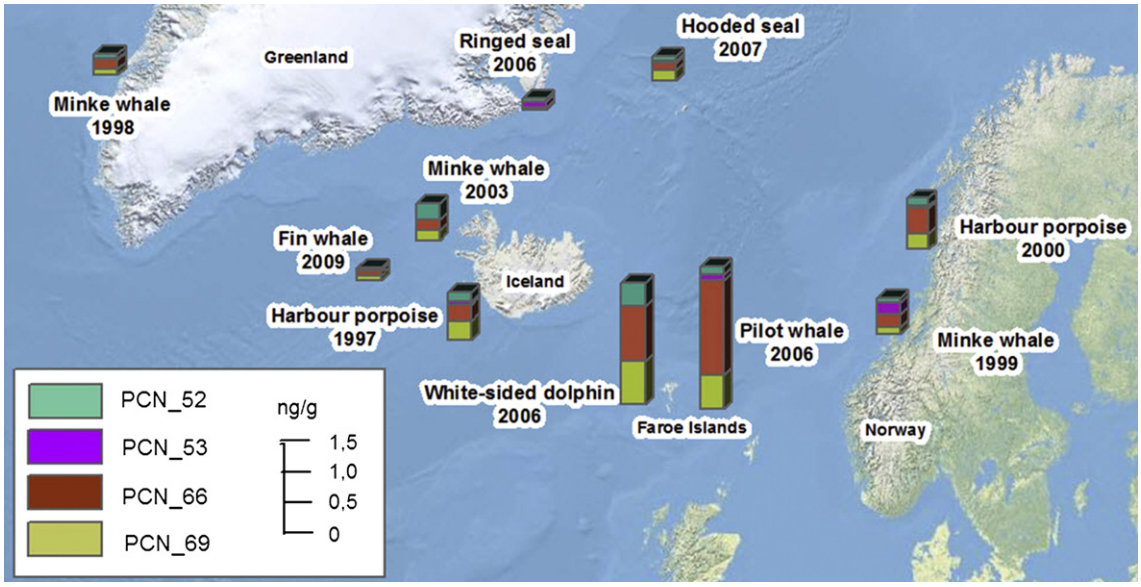

Fig. 3. Concentrations of four congeners of PCN occurring in overall highest concentrations are shown. The figure represents the most recent samples of the various species analyzed for each geographic sampling location. Image credit: Sanna í Túni Nielsen.

differences in congener profile between species are probably related to differences in feeding biology as well as species-dependent metabolic capacities. In the ringed seals from East Greenland and in the minke whales from Norway, PCN-53 constituted the largest proportion of PCNs. This was not species-dependent since samples of minke whales from Greenland and Iceland contained insignificant amounts of the PCN-53 congener. Also, no measurable levels of PCN-53 were detected in the Icelandic fin whales. Although the sampling took place in different years, the different patterns seen in the minke whale samples from Iceland (2003–2006), West Greenland (1998) and Norway (1993 and 1999) are likely explained for the most part by factors associated with geographic variations. For the female minke whales from West Greenland, as well as the female hooded seals, also sex-related differences in metabolism of contaminants may have influenced the PCN accumulation pattern.

3.3. Temporal variations in concentration

Temporal variations in PCN concentrations were examined in samples of pilot whale, ringed seal, fin whale, hooded seal, and Atlantic white-sided dolphin, covering a time period of 21 years (1986–2007). In general, no consistent temporal trend of ∑PCN was found for all species and geographical areas. However, in all species except minke whales from Norway the lowest ∑PCN concentrations were found in samples from the latest sampling period (Fig. 5). The relatively small data set and high data variability within species renders it difficult to determine a statistically significant trend in the contaminant levels in the studied areas. The variability can be attributed to different migratory paths of animals included in some of the pools. A significant decrease in ∑PCN concentration was only found in the hooded seals from West Ice for the period 1990 to 2007 (ANOVA, $p = 0.03$). In the ringed seals from East Greenland the

Table 2
Min and max IUPAC PCN congener concentrations (ng/g lw) in three pooled blubber samples ($n = 3–6$).

	Years	∑PCN[a]	PCN-48/35[b]	PCN-52/60	PCN-53/55	PCN-66/67	PCN-69
Pilot whale	86	2.9–3.7	<0.2–1.1	0.1–0.5	0.3–0.9	1.3–2.3	0.3–0.8
	97	2.5–5.2	0.8–0.9	0.2–0.4	0.1–0.8	1.6–3.9	0.5–0.7
	06/07	1.8–4.1	0.3–0.6	0.1–0.2	0.1–0.2	1.0–3.1	0.6–0.7
W-s dolphin	97	2.3–3.4	1.3–2.7	0.4–0.7	0.1	1.1–1.3	0.6–1.3
	01/02	3.2–5.1	2.8–4.6	0.5–0.7	0.1–0.2	1.5–1.9	1.2–2.3
	06	1.6–3.0	1.3–2.4	0.4–0.5	0.1	1.0–1.1	0.1–1.4
Hooded seal	90	0.6–0.8	0.8–1.4	0.2–0.3	0.1	0.1–0.3	0.1–0.2
	97	0.6–0.7	0.7–0.9	0.2	0.1	0.2	0.2
	07	0.4–0.5	0.3–0.4	0.1	0.04	0.1–0.2	0.2
Ringed seal	86	0.3–0.4	<0.3–0.6	0.2	0.1–0.2	<0.1	0.1
	00	0.2–0.8	<0.2	0.1–0.2	0.1–0.4	<0.1	0.1
	06	0.2–0.3	<0.2	0.1	0.1	<0.1	0.1
Harbour porpoise	92 I	0.8–1.6	0.5–5.9	0.1–0.6	0.1	0.4–0.6	0.3–0.4
	97 I	0.6–1.2	1.2–3.4	<0.3–0.3	<0.1–0.1	0.3	0.2–0.5
	00 N	1.0–1.2	0.5–2.3	0.1–0.2	0.04–0.1	0.5	0.3–0.4
Minke whale	93 N	0.5–0.7	<0.2	<0.1–0.1	0.2	0.2–0.4	0.1
	99 N	0.6–0.9	<0.3	<0.1–0.3	0.2–0.3	0.2–0.3	0.1–0.2
	98 G	0.3–0.6	0.2–0.4	0.1	<0.2	0.1–0.3	0.1
	03/06 I	0.5–0.9	1.3–3.1	0.2–0.5	<0.2	0.1–0.2	0.1–0.2
Fin whale	86/89	0.1–0.4	0.2–0.7	<0.1–0.1	<0.1	<0.1–0.3	<0.02–0.02
	06/09	0.1–0.3	0.1–0.5	<0.02–0.03	<0.1	0.04–0.1	0.1

[a] ∑PCN = PCN-52, 53, 66 and 69. Values below LOD included in ∑PCN were divided by two.
[b] Indicative values due to poor method reproducibility.

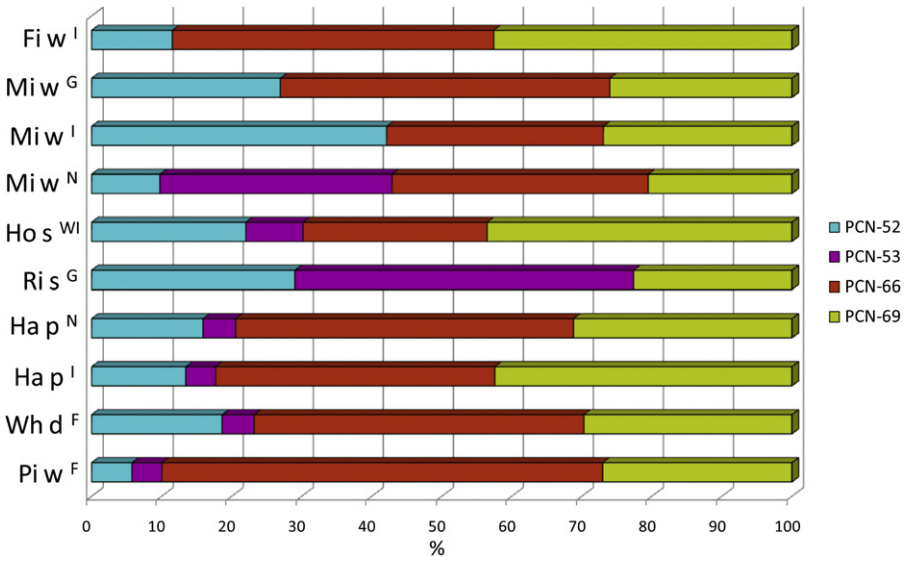

Fig. 4. Indicative distribution of PCN congeners in percentage of ∑PCN for PCN-52 (blue), 53 (purple), 66 (red) and 69 (green) indicated by colours. F = Faroe Islands, N = Norway, I=Iceland, WI = West Ice and G = Greenland. The bars represent average percentage based on three pooled samples from the latest sampling period (Fig. 3). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

median concentration decreased from 1986 to 2006, but not significantly (ANOVA, $p = 0.07$). Other studies have shown decreasing PCN levels, as well as diminished human exposure to these compounds. A notable decrease was seen in the concentrations of PCNs in samples of Swedish breast milk from 1972 to 1992 (Lundén and Norén, 1998), and based on consumption of sardine, hake and mussel, the intakes of PCNs by the general population of Catalonia, Spain decreased between 2000 and 2005, from 3.63 ng/day to 0.33 ng/day (Domingo et al., 2003; Llobet et al., 2007). The production of technical mixtures of PCNs ceased in most countries in the 1980s. However, since illegal use and export of PCNs were recently discovered, continued emissions of PCNs may be anticipated (Yamashita et al., 2003). Changes in the congener distribution over time seen within species may be due to changes in sources of exposure caused by changes in global emissions of PCNs and/or feeding behaviour.

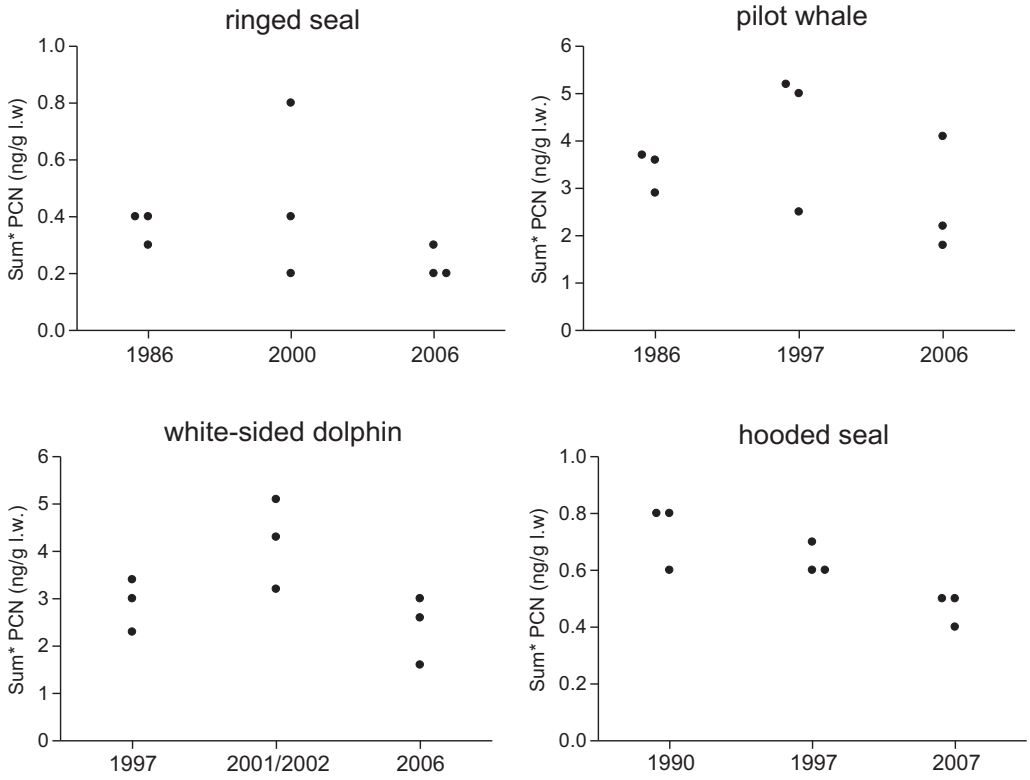

Fig. 5. PCN concentrations in 3 pooled blubber samples of ringed seals from central East Greenland, pilot whales and white-sided dolphins from the Faroe Islands and hooded seals from the West Ice ($n = 4–5$). *Sum of PCN congeners 52, 53, 66 and 69. Values below LOD included in ∑PCN were divided by two.

3.4. Total toxic equivalents (TEQs)

Some PCNs exhibit toxic activity similar to 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and co-planar PCBs by binding to the Ah-receptor, and consequently PCNs have been assigned relative effect potencies (REPs) similar to the co-planar PCBs (Blankenship et al., 2000; Van den Berg et al., 2006; Villeneuve et al., 2000). In Arctic and Antarctic marine mammals, PCNs were found to account for <1% of total toxic equivalents (TEQs) in ringed seal, Weddell seal, seabirds and polar bear, and 6–15% in beluga and pilot whale (Bidleman et al., 2010). Of the detected congeners in the current study, only PCN-66 has been shown to elicit a dioxin-like response and hence has a REP value. The highest levels of PCN-66 were found in the pilot whales, and the calculated TEQ-concentrations ranged between 4 and 17 pg/g lw. Similar TEQs derived from PCNs were also found in a previous study on pilot whales from the Faroe Islands (8–13 pg/g TEQ) (Vorkamp et al., 2004). These TEQ values are lower than those considered to elicit toxicological effects in birds and marine mammals (Kannan et al., 2001). Nevertheless, in the study by Vorkamp and co-workers, where the TEQs, provided by only dioxins and co-planar PCBs, ranged from 46 pg/g lw to 71 pg/g lw it was concluded that the contribution of TEQ from PCNs might be important in samples with dioxin TEQ-concentrations close to regulatory limits. Therefore, since many of the marine mammal species analyzed in the present study are part of the traditional local diet, they contribute partially to the human exposure to TEQs.

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Appendix. Supplementary material

Supplementary data related to this article can be found online at doi:10.1016/j.envpol.2012.01.035.

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