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Review

Polychlorinated naphthalenes in polar environments — A review ☆

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ABSTRACT

Polychlorinated naphthalenes (PCNs) consist of naphthalene substituted with 1–8 chlorines, yielding 75 possible congeners. They were formerly used in industry, occur at trace levels in commercial PCB mixtures, and have current sources in combustion processes. PCNs are widespread in arctic air with higher levels in the European Arctic. Concentrations were higher during the cold months in arctic Canada and Russia, but no seasonality was noted in subarctic Canada and Greenland. "Marker" congeners indicative of combustion were evident at some sites. Total toxic equivalents (TEQ) in air due to PCNs + dioxin-like PCBs were dominated by PCNs in arctic Canada and Russia, but not in subarctic Canada. Deposition of PCNs in snow was measured in northern Norway and Svalbard. Surveys of PCNs in the lower food web are limited to the northern Baltic Sea and lakes/rivers of northern Scandinavia. PCNs showed little or no biomagnification in lower food webs of the northern Baltic and discrimination among congeners suggested preferential metabolism. There are no reports of PCNs in fish and invertebrates from the Arctic Ocean, and only one from Antarctica. Total PCNs in marine mammals followed the order: harbour seal ~ pilot whale ≥ polar bear > beluga > ringed seal ~ Weddell seal. Total PCNs in seabirds varied over 100-fold, with higher concentrations in glaucous gull eggs and plasma from Bear Island, and livers of northern fulmar from the eastern Canadian Arctic. Lower concentrations occurred in eggs of glaucous gull from Svalbard and black-backed gull from the Faroe Islands. PCNs accounted for <1% of total TEQ in ringed seal, Weddell seal, seabirds and polar bear, but up to 6–15% in beluga and pilot whale. TEQ due to PCNs were generally low in harbour seal, but up to 9% of total TEQ in some animals.

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☆ This paper is a contribution to the AMAP POPs assessment.

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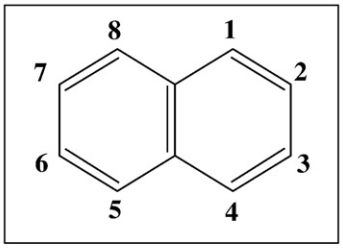
1. Introduction

Polychlorinated naphthalenes (PCNs) are a group of industrial chemicals consisting of naphthalene substituted with 1–8 chlorine atoms, yielding 75 possible congeners. These are listed by structure and number in Fig. 1 and by several authors (Falandysz, 1998; Helm et al., 2006; Jakobsson and Asplund, 2000; Kucklick and Helm, 2006; Lundgren et al., 2003; Noma et al., 2004). PCN mixtures were produced in several countries under the tradenames Halowax, Nibren, Clonaicre and Seekay waxes and Cerifal Materials (Falandysz, 1998). PCNs were first patented as flame retardants and dielectric fluids for capacitors in the early 1900s and found use in a variety of industrial applications, some of which were dye-making, fungicides in the wood, textile and paper industries, plasticizers, oil additives, casting materials for alloys and lubricants for graphite electrodes (Crooks and Howe, 1993; Falandysz, 1998; Helm et al., 2006; World Health Organization, 2001). PCNs also occur as trace contaminants in commercial polychlorinated biphenyl (PCB) mixtures (Falandysz, 1998; Taniyasu et al., 2003; Yamashita et al., 2000a). Production figures for PCNs are not well known, but have been estimated to be ~150 000 tonnes, ~10% of the global PCB production (Crooks and Howe, 1993; Falandysz, 1998). Production and usage history in the U.S.A. has been summarized by the



Monochloro-	Positions	Tetrachloro-	Positions	Pentachloro-	Positions
1	1	27	1,2,3,4	55	1,2,3,6,8
2	2	28	1,2,3,5	56	1,2,3,7,8
		29	1,2,3,6	57	1,2,4,5,6
Dichloro-		30	1,2,3,7	58	1,2,4,5,7
3	1,2	31	1,2,3,8	59	1,2,4,5,8
4	1,3	32	1,2,4,5	60	1,2,4,6,7
5	1,4	33	1,2,4,6	61	1,2,4,6,8
6	1,5	34	1,2,4,7	62	1,2,4,7,8
7	1,6	35	1,2,4,8		
8	1,7	36	1,2,5,6	**Hexachloro-**	
9	1,8	37	1,2,5,7	63	1,2,3,4,5,6
10	2,3	38	1,2,5,8	64	1,2,3,4,5,7
11	2,6	39	1,2,6,7	65	1,2,3,4,5,8
12	2,7	40	1,2,6,8	66	1,2,3,4,6,7
		41	1,2,7,8	67	1,2,3,5,6,7
Trichloro-		42	1,3,5,7	68	1,2,3,5,6,8
13	1,2,3	43	1,3,5,8	69	1,2,3,5,7,8
14	1,2,4	44	1,3,6,7	70	1,2,3,6,7,8
15	1,2,5	45	1,3,6,8	71	1,2,4,5,6,8
16	1,2,6	46	1,4,5,8	72	1,2,4,5,7,8
17	1,2,7	47	1,4,6,7		
18	1,2,8	48	2,3,6,7	**Heptachloro-**	
19	1,3,5			73	1,2,3,4,5,6,7
20	1,3,6	**Pentachloro-**		74	1,2,3,4,5,6,8
21	1,3,7	49	1,2,3,4,5		
22	1,3,8	50	1,2,3,4,6	**Octachloro-**	1,2,3,4,5,6,7,8
23	1,4,5	51	1,2,3,5,6	75	
24	1,4,6	52	1,2,3,5,7		
25	1,6,7	53	1,2,3,5,8		
26	2,3,6	54	1,2,3,6,7		

Fig. 1. Numbering and substitution positions of the 75 polychlorinated naphthalene congeners. The α-positions are 1, 4, 5 and 8; β-positions are 2, 3, 6 and 7.

World Health Organization (2001). Today, manufacture of PCNs is thought to have ended, although illegal importation of PCN-containing products into Japan was reported after 2000 (Falandysz et al., 2008; Yamashita et al., 2003). A technical review of the PCN dossier by the United Nations Economic Commission for Europe (UN-ECE) Task Force on Persistent Organic Pollutants (POPs) supported the dossier's conclusion that PCNs be considered POPs in the context of the Convention on Long-Range Transboundary Air Pollution (CLRTAP) (UN-ECE, 2006).

Sources of PCNs to the environment include evaporation from old or in-use products containing PCNs and PCBs, and release during combustion (Falandysz, 1998). Van der Gon et al. (2007) estimated that 1.0 tonne of PCNs was emitted in Europe in 2000, considering waste combustion, other combustion (residential, commercial, agricultural), industrial sources (PCNs as impurities in PCB mixtures) and solvent and product usage. Combustion sources accounted for over 80% of estimated PCN emissions.

Like other POPs, PCNs are globally distributed in air, sediments and biota. Early reviews summarized PCN production and usage, sources, occurrence in the environment and toxicology (Falandysz, 1998; Jakobsson and Asplund, 2000). More recent, specialized reviews have been published on PCN analytical chemistry (Kucklick and Helm, 2006), physicochemical properties (Puzyn and Falandysz, 2007), occurrence in aquatic animals, the food chain and human exposure (Domingo, 2005; Falandysz, 2003) and occurrence in the Laurentian Great Lakes ecosystem (Helm et al., 2006). Investigations of PCNs in polar environments have increased in recent years, but are still relatively few. This review begins with an overview of PCN properties, analysis and characteristics of CN congener distributions in source types, then focuses on PCN contamination of arctic and subarctic air, snow, sediment and biota. The single report of PCNs in antarctic biota is also included. Although the review generally does not cover temperate regions, some studies in the southern Baltic Sea and Laurentian Great Lakes of North America are briefly discussed to provide context.

2. Physicochemical properties

Physicochemical properties of PCN congeners have been measured for some congeners but not others. Liquid-phase saturation vapour pressures (VP, Pa) have been determined for all 75 congeners using gas chromatographic (GC) methods (Lei et al., 1999) or extensions of the method using the published retention data of Järnberg et al. (1994). Octanol–air partition coefficients (K_{OA}, dimensionless) have been determined for 56 congeners by a generator column technique (Harner and Bidleman, 1998a) or GC correlations (Su et al., 2002), but octanol–water partition coefficients (K_{OW}, dimensionless) and water solubilities (WS, μg L^{-1}) have been measured for only 21 and 16 congeners, respectively (reviewed by Helm et al., 2006, and Puzyn and Falandysz, 2007). No air–water partition coefficients (K_{AW}, dimensionless) have been directly measured, but have been estimated for 17 congeners from the ratio of K_{OW}/K_{OA} (Helm et al., 2006). Puzyn and Falandysz (2007) predicted K_{OW}, K_{OA} and K_{AW} for all congeners using quantitative structure–property relationships (QSPR). Based on comparisons between predicted and measured properties, the most successful modelling approach was described as "neural networks with variable selection using a genetic algorithm (GA-NN)". Table 1 gives a summary of PCN physicochemical properties by homologue.

Table 1
Physicochemical properties (logarithms) of PCNs, by homologue.

Chlorines	Water solubility[a]	Liquid phase VP[a]	K_{OW}	K_{OA}	K_{AW}
	μg L^{-1}	Pa			
1	924 to 2870	0.402 to 0.747	3.93 to 3.97	5.93 to 6.02	−2.05 to −2.01
2	137 to 474	−0.521 to −0.453	4.20 to 4.63	6.55 to 7.02	−2.83 to −1.98
3	17 to 65	−1.169 to −0.943	5.49 to 5.50	7.19 to 7.94	−3.35 to −2.01
4	3.7 to 8.2	−1.967 to −1.504	5.14 to 6.10	7.88 to 8.79	−3.54 to −2.02
5		−2.561 to −2.098	5.67 to 6.49	8.79 to 9.40	−3.73 to −2.30
6		−3.134 to −2.804	6.02 to 6.68	9.62 to 10.17	−4.13 to −3.04
7		−3.609 to −3.556	6.48 to 6.57	10.68 to 10.81	−4.34 to −4.11
8	0.08	−4.165	6.43	11.64	−5.21

Predicted dimensionless K_{OW}, K_{OA} and K_{AW} summarized from Puzyn and Falandysz, 2007.
[a] Experimental solubilities and vapour pressures summarized from original sources by Helm et al., 2006.

3. Analysis

Modern analysis of PCNs is usually carried out by capillary GC with detection by low- or high-resolution mass spectrometry in electron impact or electron capture negative ion modes (EI-MS, ECNI-MS). Following sample cleanup by conventional methods (e.g., florisil, alumina, silica, size exclusion), isolation of planar PCNs as well as coplanar PCBs, polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) from non-planar organochlorine compounds is often achieved by column chromatography on carbonaceous adsorbents or high performance liquid chromatography on a PYE (2-(1-pyrenyl)-ethyldimethylsilylated silica) column. A review of analytical methods, including a history of techniques, sample extraction methods, choice of GC columns, quantitation methods and potential interferences has been published by Kucklick and Helm (2006). Two interlaboratory studies (ILS) have been done for PCNs, the first involving distribution of a Halowax solution (Harner and Kucklick, 2002). Results were consistent for ΣPCNs, with all laboratories except one reporting values with a relative standard deviation (RSD) of 11%. Values reported for individual congeners were more variable, with RSDs of 20–40%. A second phase ILS was conducted using standard reference material (SRM) sediment and urban dust, as well as a Halowax solution (Kucklick and Helm, 2006). Preliminary results showed RSDs of 10% or lower for the ΣPCNs in the Halowax solution and SRMs. PCNs have also been reported in two fish SRMs (Kucklick et al., 2000), but these have not been subjected to interlaboratory comparisons.

Lack of single CN congeners for analytical standards has been a continuing problem, with fewer than half of the 75 congeners commercially available (Kucklick and Helm, 2006). Many studies have been done to characterize commercial PCN mixtures using the few congeners available and assuming the same GC–EI-MS or GC-flame ionization detection response factors for all congeners within a homologue These mixtures were then used as secondary standards in GC–ECNI-MS analysis (Kucklick and Helm, 2006; Noma et al., 2004, and references therein). Recent characterizations of the Halowax series have been done by Noma et al. (2004), Falandysz et al. (2000, 2006a,b) and Lukaszewicz et al. (2007). The congener profiles of CNs that occur as trace impurities in commercial PCB mixtures have been determined (Taniyasu et al., 2003; Yamashita et al., 2000a). A problem with using Halowax mixtures as analytical standards is that they are low in, or lack, some congeners which are useful markers of combustion sources (Section 4), particular CNs 29, 44, 54 and 70 (Noma et al., 2004).

4. Sources

PCN sources to the atmosphere can be broadly classified as "evaporation" and "combustion" (Helm and Bidleman, 2003). Evaporation sources include emission of PCNs by volatilization from in-use

or disposed products and from contaminated soil. Combustion release of PCNs occurs from various industrial and waste incineration processes and includes *de novo* synthesis as well as release of PCNs contained within incinerated waste. To a lesser extent, PCNs are also released during combustion of coal and wood (Lee et al., 2005a). Many studies have identified certain CN congeners that are absent or occur at low levels in commercial PCN and PCB mixtures, but are enriched in combustion sources. Various workers have associated enrichment of the following CN congeners with combustion processes: 13, 18, 17/25, 24, 26, 27, 29 (or coeluting 28/29/43), 35, 36, 39, 44, 36/45, 48, 50, 51, 52/60, 54, 62, 66/67, 70 and 73 (Abad et al., 1999; Baek et al., 2008; Harner et al., 2006; Helm and Bidleman, 2003; Helm et al., 2006; Iino et al., 2001; Jansson et al., 2008; Lee et al., 2007; Noma et al., 2004, 2006; Takasuga et al., 2004, and earlier studies summarized in Meijer et al., 2001).

Several approaches have been used to assess whether combustion processes contribute to PCN residues in environmental media. Such inputs are implied by the presence of PCNs prior to the industrial era in deep sediment layers from a U.K. lake (Gevao et al., 2000). Meijer et al. (2001) found increasing proportions of combustion-related congeners relative to their homologue totals in archived U.K. soils. The lack of a correlation of combustion congeners with other PCNs in air supports their use as indicators (Jaward et al., 2004). Lee et al. (2007) calculated the fraction of combustion congeners in the Halowaxes (<0.11) and compared this to observed fractions in air samples to estimate the influence of combustion sources. Similarly, Helm and Bidleman (2003) calculated the proportion of combustion contributions in air based on the enrichment of PCN indicator congeners over what was expected from evaporation alone.

Ratios or fractions of selected congeners have been used to indicate possible sources. Takasuga et al. (2004) found that the CNs 73/74 ratio was <1 in summer but >1 in winter for two air samples collected in urban Japan. The winter profile corresponded to the order of abundance in fly ash and flue gas and the enrichment of other combustion markers. However, CN 73/74 ratios vary in source samples, with values <1 in Halowaxes (Järnberg et al., 1997; Noma et al., 2004), >1 in combustion effluents (Abad et al., 1999; Jakobsson and Asplund, 2000; Jansson et al., 2008; Takasuga et al., 2004), but mixed ratios in PCB mixtures. In eighteen commercial PCB mixtures examined by Yamashita et al. (2000a), the CNs 73/74 ratio was >1 in twelve, <1 in four and ~1 in two. The ratio of CNs 73/74 was >1 in Delor PCB products (Taniyasu et al., 2003). Homologue profiles of samples impacted by chloralkali plants are dominated by hexa- and hepta-CNs (Järnberg et al., 1997; Kannan et al., 1998). The CNs 73/74 ratio in these has been reported to be >1 by Järnberg et al. (1997), but <1 by Kannan et al. (1998). Similarly, Helm and Bidleman (2003) determined isomer fractions, IF_{52-60} (CN52/[CNs52 + 60]) and IF_{66-67} in air, Halowax and PCB mixtures, and flyash samples. However, there was overlap in values among the various sources, limiting their use in interpretation of results.

While ratios can be useful, more diagnostic information about source contributions is obtained by examining relative proportions of many congeners using principal component analysis (PCA). Multivariate analysis was used to identify the source of PCNs in an illegally imported technical formulation (Falandysz et al., 2008). Järnberg et al. (1997) considered CN congener profiles for a number of different source types represented by Halowaxes, Aroclor and Clophen PCB mixtures, a municipal waste incinerator fly ash and graphite sludge from a chloralkali plant. An outcome of the PCA model was that Halowaxes and the fly ash appeared less important than CNs from technical PCB mixtures to the profiles seen in environmental samples from sites impacted by non-point source contamination. Helm and Bidleman (2003) applied PCA to Toronto air samples found a separation of loadings due to combustion CNs and non-ortho PCBs (which also occur in combustion effluents) and loadings due to CNs from evaporative sources. They concluded that evaporative PCN emissions dominated at both the downtown and suburban sites, but that combustion had a greater influence on the suburban site. Orlikowska et al. (2009) carried out a detailed examination of PCN homologue and congener profiles in Scots pine needles (*Pinus sylvestris* L) in Poland, collected in 2002. The ΣPCNs ranged from 70 to 1100 pg g^{-1} wet weight among the sites. Similarities, but also variations, in tri- to octa-CNs homologue and congener profiles were found. Tri- and tetra-CNs were major contributors at most locations, but hepta- and octa-CNs were also prominent and in many cases outweighted penta- and hexa-CNs. PCA was able to sort the congener profiles into factors related to molecular weight and specific substitution patterns, which could be related to source categories such as industrial emissions, evaporative losses from dumped products containing PCNs, technical PCB formulations, and combustion.

Studies have identified elevated levels of ΣPCNs in some European cities, which could be a source to the European Arctic. The ΣPCNs (3–8 Cl) averaged 85 pg m^{-3} in Chilton and 110 pg m^{-3} in Hazelrigg, U.K. in 2001, while a lower level of 15 pg m^{-3} was found at Mace Head, a remote station on the west coast of Ireland, in 2000 (Lee et al., 2005b). Average ΣPCNs (3–8 Cl) in Lancaster and Manchester, U.K. during 1994–1999 ranged from 66 to 152 pg m^{-3} (Harner et al., 2000; Lee et al., 2000). Lee et al. (2005b) reported no decline in ΣPCNs for air samples collected in the mid-1990s and 2001 at a site in southwestern England, while a decrease in ΣPCBs was found in the same samples. In 2000–2001, the ΣPCNs were approximately equal to the ΣPCBs at three U.K. locations. Jaward et al. (2004) reported 140–220 pg m^{-3} in London, Moscow and urban-industrialized regions of Poland by summing only 13 congeners, which may have underestimated actual ΣPCN levels. No significant correlations were found between PCNs and PAHs. Jaward et al. (2004) suggested the lack of correlation was related to different source types for the two compound classes; efficient, high temperature sources (e.g., incinerators) for PCNs and inefficient, low temperature sources for PAHs. Evaporative contributions of PCNs would also lead to low correlation with PAHs.

5. PCNs in air and snow

5.1. Total PCNs

Atmospheric half-lives with respect to gas-phase OH radical reaction were predicted by Puzyn et al. (2008) as: 2 days (mono-CNs), 5 days (di-CNs), 10 days (tri-CNs), 19 days (tetra-CNs), 39 days (penta-CNs), 79 days (hexa-CNs), 163 days (hepta-CNs) and 343 days (octa-CN). The Stockholm Convention (Annex D) (UNEP, 2001) recognizes an atmospheric half-life >2 days as a criterion for long-range transport potential. Several studies have measured PCNs in atmospheric samples from arctic and subarctic regions, but different numbers of homologues and congeners have been reported by various investigators, make comparison of concentrations difficult. Air concentrations of ΣPCNs, locations and homologues/congeners determined are summarized in Table 2. Spatial trends for locations where 37 or more CN congeners were measured are illustrated in Fig. 2.

PCNs was first quantified in arctic air by Harner et al. (1998), who collected shipboard samples in 1996 in the Barents Sea, Norwegian Sea and eastern Arctic Ocean, and in a few archived air sample extracts from 1993 to 1994. The latter were taken at land-based high arctic stations at Alert, Canada and Dunai, Russia, under Canada's Northern Contaminants Program. Site descriptions and methodology used in this monitoring program are given by Hung et al. (2005) and cited papers. Average concentrations of ΣPCNs (3–8 Cl) in air were 3.5 and 0.84 pg m^{-3} at the two land stations, while shipboard averages ranged from 7.1 to 40 pg m^{-3} in the three ocean regions.

A follow-up study at Dunai, Alert and a second subarctic Canadian station Tagish was done with archived air sample extracts covering a full year from 1994 to 95 (Helm et al., 2004). Annual mean ΣPCNs (3–8 Cl) were 0.66 pg m^{-3} at both Dunai and Alert stations and were

Table 2
ΣPCNs in air of arctic and subarctic regions, pg m^{-3}.

Location	Latitude °	Longitude °	Year	Mean ΣPCNs	Homologues	Congeners analysed	Reference
Alert, Canada	82.5N	62.3W	1993–1994	3.5	3–8	40	Harner et al. (1998)
Alert, Canada	82.5N	62.3W	1994–1995	0.66	3–8	40	Helm et al. (2004)
Alert, Canada	82.5N	62.3W	2004–2005	1.2	3–8	49	Lee et al. (2007)
Dunai, Russia	74.1N	124.5E	1993	0.84	3–8	40	Harner et al. (1998)
Dunai, Russia	74.1N	124.5E	1994–95	0.66	3–8	40	Helm et al. (2004)
Tagish, Canada	60.3N	134.2W	1994–95	0.37	3–8	40	Helm et al. (2004)
Resolute Bay, Canada	74.6N	94.9W	1999	4.2	3–8	40	Helm (2002)
Eastern Archipelago, Canada	63.7–74.6N	68.5–94.9W	1999	7.4	3–8	40	Helm (2002)
Norwegian Sea	63.4–74.4N	63–16.0E	1996	7.1	3–8	40	Harner et al. (1998)
Barents Sea	71.1–80.3N	21.6–65.7E	1996	40	3–8	40	Harner et al. (1998)
Eastern Arctic Ocean	74.6–89.9N	10.9–179.9E	1996	12	3–8	40	Harner et al. (1998)
Ny Ålesund, Norway	78.5N	11.5E	2001	35	3–8	44	Herbert et al. (2005a),b)
Ny Ålesund, Norway	78.5N	11.5E	2004–2005	7.6	3–8	49	Lee et al. (2007)
Tromsø, Norway	69.7N	17.0E	2003	25	3–8	44	Herbert et al. (2005a),b)
Ammernäs, Sweden	65.5N	43.7E	1990–1991	1.6	4–6	37	Egebäck et al. (2004)
Nuuk, Greenland	64.1N	51.4W	2004–2005	0.16	3–7	13	Bossi et al. (2008)
Storhofdi, Iceland	63.4N	20.3W	2004–2005	0.86	3–8	49	Lee et al. (2007)
Barrow, U.S.A.	71.3N	156.8W	2004–2005	2.3	3–8	49	Lee et al. (2007)

lower at Tagish, 0.37 pg m^{-3}. The ΣPCNs levels at Dunai and Alert were higher during winter, spring and fall compared to summer, while seasonal differences were not apparent at Tagish. The higher concentrations of ΣPCNs at Alert and Dunai during the cold months were in contrast to the trend for mono- and non-ortho PCBs, which showed no distinct seasonality. Helm et al. (2004) noted that the ΣPCNs appeared to follow the trend for arctic haze, which is a prominent feature of the high arctic air mass in winter–spring, and that the combustion products polycyclic aromatic hydrocarbons (PAHs) also peaked in the haze season (Halsall et al., 1997). In the summer of 1999, Helm (2002) conducted air sampling at Resolute Bay, Nunavut, Canada and from shipboard in the eastern Canadian Archipelago with mean ΣPCNs (3–8 Cl) concentrations of 4.2 pg m^{-3} and 7.4 pg m^{-3}, respectively.

Herbert et al. (2005a) sampled air in winter months at the Norwegian arctic research station at Ny Ålesund during April 2001 and in Tromsø, Norway during February to March 2003. Mean ΣPCNs (3–8 Cl) were relatively high at both stations, 35 and 25 pg m^{-3} respectively, similar to those found over the Barents Sea in 1996 (Harner et al., 1998).

Egebäck et al. (2004) collected air samples from September to March, 1990–1991 at Ammarnäs, a meteorological station in northern

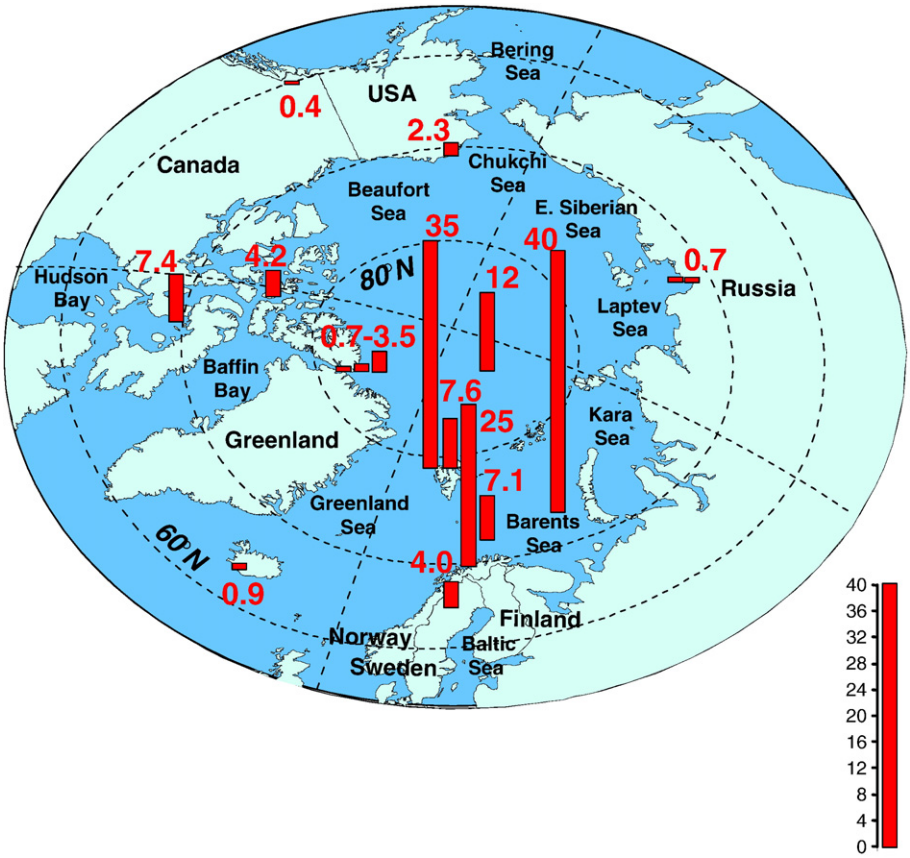


Fig. 2. ΣPCNs in air of arctic and subarctic regions, pg m^{-3}. Data are included for those studies which measured 37 or more congeners (Table 2). The ΣPCN concentration of 1.6 pg m^{-3} in northern Sweden, based on the analysis of the 4–6 Cl homologues (Egebäck et al., 2004), was increased to 4.0 pg m^{-3} by assuming that tri-CNs accounted for 60% of ΣPCNs.

Sweden. The mean ΣPCNs was 1.6 pg m^{-3}, though only the 4–6 Cl CNs were determined. Parallel sampling was done at Hoburgen, a station in southern Sweden, where ΣPCNs averaged 5.1 pg m^{-3}. At a third station in southern Sweden, tri-CNs were also measured in air and deposition and accounted for ~60% of ΣPCNs. Similarly high proportions of tri-CNs were found at other arctic stations (see below). With this consideration, the ΣPCNs at Ammarnäs and Hoburgen might be raised to ~4 and ~13 pg m^{-3}, respectively, to include the tri-CNs for comparison to other studies.

Bossi et al. (2008) carried out air sampling at Nuuk, Greenland during 2004–2005. The annual mean ΣPCNs (3–7 Cl) was 0.16 pg m^{-3}. Similarly to Tagish (Helm et al., 2004), no seasonality in ΣPCN concentrations was observed over this time frame. Since only 13 congeners were quantified, including one tri-CN, it is difficult to compare their ΣPCN levels with those in other studies. Relationships were explored between atmospheric concentrations of semivolatile compounds and carbon monoxide, considered to be an indicator of anthropogenic influence. Although significant correlations were obtained for some compounds: positive for chlordanes, *p,p′*-DDE, trifluralin, negative for γ-hexachlorocyclohexane (γ-HCH), correlations for other organochlorine pesticides (dieldrin, endosulfan, α-HCH) and ΣPCNs were not significant.

POPs are being surveyed around the world under the Global Atmospheric Passive Sampling (GAPS) study (Pozo et al., 2006). The ΣPCNs (3–8 Cl) in pg m^{-3} at arctic–subarctic sites in the first phase of the GAPS study were: Alert 1.2, Ny Ålesund 7.6, Storhofdi, Iceland 0.86 and Barrow, Alaska, U.S.A. 2.3 (Lee et al., 2007). PCNs were below the limits of detection, 0.0007–0.23 pg m^{-3} per congener, at the Italian base in Antarctica (Lee et al., 2007).

Farrar et al. (2006) and Jaward et al. (2004) deployed passive air samplers across Europe to determine spatial trends of PCNs and other POPs in air. Farrar et al. (2006) used polymer coated glass slides which were deployed during seven days in June 2002. Jaward et al. (2004) deployed GAPS-type samplers from June to July, 2002. Both groups included two arctic–subarctic stations in Ny Ålesund and Iceland. Although these papers show relative ΣPCN concentrations (based on analysis of 10–17 congeners) on bar graphs, neither report numerical values for most sites. Jaward et al. (2004) found no correlation between CN congeners and PAH congeners in European air survey. Farrar et al. (2006) found a significant correlation of PCNs with only one PAH, benzo[ghi]perylene, but PCNs were significantly correlated with most PCB congeners.

As shown in Fig. 2, the ΣPCN concentrations in air are much higher at most European arctic–subarctic locations than at sites in Siberia, Iceland, Alaska and the Canadian Arctic. Harner et al. (1998) noted that high concentrations in the Barents Sea were associated with five-day air parcel trajectories from Europe, while samples with lowest ΣPCNs had air mass origins over the open ocean, northern Scandinavia and Greenland. Herbert et al. (2005a) measured elevated ΣPCNs at Ny Ålesund during a period when air trajectories originated from western Europe, the U.K. and Scandinavia. However, the authors could not rule out the possibility of PCN contamination from the local scientific facility at Ny Ålesund or the city of Tromsø.

Although the ΣPCNs at Dunai, Alert and Tagish were much lower than those in the European Arctic, Helm et al. (2004) were able to make inferences about source quadrants based on five-day air trajectories. Levels at Dunai were positively correlated ($p<0.05$) with the fraction of time that air parcels originated over eastern and western Russia. At Alert, ΣPCNs were weakly correlated ($p<0.1$) with the time over the North Atlantic and Europe and negatively correlated ($p<0.05$) with the percent of air masses originating from the North American sector. Tagish is a high elevation site (2200 m) and air arrived from the North Pacific for most of the time. No relationships were found for ΣPCN concentrations at Resolute Bay or the eastern Canadian Archipelago and air transport direction (Helm, 2002).

Fig. 2 shows 1–2 orders of magnitude difference in ΣPCN concentrations among circumpolar arctic–subarctic stations. By comparison, the variability in median concentrations of legacy organochlorine pesticides (chlordanes, hexachlorocyclohexanes, dieldrin, *p,p′*-DDE) among circumpolar air monitoring stations is only about a factor of 2–3 (Su et al., 2006, 2008). Concentrations of Σ_{37-49} PCNs in arctic air (0.37–40 pg m^{-3}, median 3.5 pg m^{-3}, Table 2) can be compared to those of ΣPCBs. A survey of 102 PCB congeners at Alert, Tagish and Dunai in 1992–1994 showed similar Σ_{102}PCB levels at Dunai and Alert (27 and 34 pg m^{-3}) and lower concentrations at Tagish (17 pg m^{-3}). A heavier PCB congener profile characterized the Dunai site. The Σ_{10} AMAP PCBs at the same sites were 5.8, 8.1 and 3.7 pg m^{-3}, respectively (Hung et al., 2001; Stern et al., 1997). Reports of PCBs in 2000–2005 were only for the Σ_{10} AMAP congeners: 2.8, 8.5 and 7.4 pg m^{-3} at Alert, Zeppelin Mountain (Svalbard) and Storhofdi (Iceland), respectively, and 6.1 pg m^{-3} for the Σ_7 congeners at Pallas (Finland) (Hung et al., 2010-this issue). Temporal trends of ΣPCNs in arctic air have not been assessed, but there are no indications of decline in U.K. air between the mid-1990s and 2001 (Lee et al., 2005b; Section 4).

5.2. Homologue and congener profiles

PCN homologues at Alert and Dunai were dominated by tri-CNs, which accounted for 60% or more of ΣPCNs, followed by 20–35% contribution of tetra-CNs, with the remainder consisting of penta-, hexa-, hepta- and octa-CNs. (Harner et al., 1998; Helm et al., 2004). Tri-CNs and tetra-CNs accounted for 55% and 35–40% of ΣPCNs at Tagish. Penta-CNs made up ~10% of, and heavier CNs<2% of ΣPCNs (Helm et al., 2004). A similar distribution was found at Resolute and in the Canadian Archipelago: 48–65% tri-CNs, 26–47% tetra-CNs, 8–10% penta-CNs and 1–2% of the heavier CNs (Helm, 2002). Homologue profiles in the Norwegian Sea and eastern Arctic Ocean showed 45–50% and 35% contributions of tri-CNs and tetra-CNs, respectively, while tetra-CNs were slightly higher than tri-CNs in the Barents Sea, each homologue accounting for 40–45% of ΣPCNs (Harner et al., 1998).

Herbert et al. (2005a) found that tri-CNs contributed 65–70% and tetra-CNs 28–30% of ΣPCNs in air at Ny Ålesund and Tromsø. The tri-CNs in snow accounted for 84% of ΣPCNs at Ny Ålesund, but only 40% of ΣPCNs at Tromsø, where tetra-CNs dominated (54% of ΣPCNs).

Lee et al. (2007) reported that tri-CNs accounted for 70–80% of ΣPCNs at Alert and Barrow, Alaska, but 55% of ΣPCNs at Ny Ålesund. Tetra-CNs made up 15–20% of ΣPCNs at Alert and Barrow, and 40% at Ny Ålesund.

Several workers have noted the occurrence of combustion-indicator CNs in arctic air. Helm et al. (2004) found small peaks for CNs 29, 44 and a prominent peak for CN 54 in the cold period air samples from Dunai. Although these CNs are absent in Halowax mixtures (Noma et al., 2004), they occur at low levels in some PCB mixtures (Yamashita et al., 2000a) and so may not be unequivocal indicators of combustion CNs. Nonetheless, the abundance of CN 54 in proportion to other penta-CNs was suggestive of combustion input. The average IF_{66-67} was 0.54 in Dunai and Alert air samples from the cold period, which was closer to the IF_{66-67} for fly ashes (0.45–0.62) and Aroclors (0.5–0.62) than to IF_{66-67} of Halowaxes (0.6–0.94) (Helm et al., 2004).

Jaward et al. (2004) used a GC peak containing CNs 28/29/43 to speciate evaporative and combustion PCN sources in air. After subtracting evaporative contributions, combustion-derived PCNs were still indicated in air samples from Ny Ålesund and Iceland.

Lee et al. (2007) used a suite of indicator CNs to speciate combustion and evaporative PCNs at Ny Ålesund. Based on the fraction of combustion PCNs/ΣPCNs present in the Halowax series (<0.11, Section 3), the PCNs at Alert appeared to be derived from mostly evaporation sources, while combustion influence was more evident at Ny Ålesund. Over the three-month period of the GAPS

sampling campaign, combustion PCNs amounted to 4.8% of ΣPCNs at Alert and 19% of ΣPCNs at Ny Ålesund.

5.3. Atmospheric processes

Atmospheric deposition of POPs takes place by precipitation scavenging of particulate and gaseous compounds and the two-way exchange of gaseous compounds between water, ice and snow and soil. The relative loadings of PCNs to arctic ecosystems by these processes depend on their distribution between the particle and gas phases in the atmosphere. The apparent phase distribution of PCNs in arctic air was investigated by Harner et al. (1998) and Helm et al. (2004), using the concentrations of PCNs trapped on a glass fiber filter and a backup polyurethane foam adsorbent as measures of particle-bound (C_p, pg m^{-3}) and gaseous (C_g, pg m^{-3}) compounds. Heavier CN congeners with lower liquid-phase vapour pressures (P_L, Pa) and higher K_{OA} were preferentially associated with particles. Helm and Bidleman (2005) measured the apparent particle–gas distribution for PCNs, non-ortho PCBs and mono-ortho PCBs in a larger set of winter–spring air samples from Alert and Dunai. Percentages of CN homologues on particles ranged from <5% for tri-CNs and tetra-CNs, 20–30% for penta-CNs, 75% for hexa-CNs to >90% for hepta-CNs and octa-CN. About 50–60% of CBs 77, 114, 118 and 105 were associated with particles, increasing to 75–85% for CBs 126 and 156.

Harner et al. (1998) correlated the log C_p/C_g ratio to log K_{OA} ($r^2 = 0.837$) for a single winter sample taken at Alert. Egebäck et al. (2004) found similar correlations ($r^2 = 0.854$–0.896) for air samples collected at Hoburgen and Ammarnäs in southern and northern Sweden. Helm and Bidleman (2005) used sulphate as a surrogate for total suspended particles (TSP, μg m^{-3}) and calculated a particle–gas partition coefficient, $K_p = C_p/(C_g \times \text{TSP})$, for a suite of PCN and mono-non-ortho PCB congeners which was correlated to K_{OA} or P_L. Comparisons were made of measured and predicted K_p, using three models, the Junge-Pankow adsorption model (Pankow, 1987), which uses P_L and aerosol specific surface area as correlating parameters, an absorption model (Finizio et al., 1997; Harner and Bidleman, 1998b), which is based on K_{OA} and the fraction of aerosol organic matter, and a dual model which considers adsorption to black carbon and absorption to organic matter (Dachs and Eisenreich, 2000). The Junge-Pankow model using P_L overestimated observed K_p, while good agreement was found using the K_{OA} absorption model and aerosol organic matter fractions of 7–12%. Surprisingly, inclusion of black carbon generally did not improve the estimates of K_p, suggesting that partitioning of PCNs to arctic aerosols occurs mainly to the organic fraction. Other studies have found that planar chlorinated aromatic compounds are strongly associated with black carbon in water and sediments (Jonker and Koelmans, 2002; Lohmann, 2003; Persson et al., 2002, 2005).

Herbert et al. (2005a) sampled air (Section 5.1) and snow at Ny Ålesund during April 2001 and in Tromsø, Norway during February to March 2003. The ΣPCN concentrations in snow meltwater spanned a wide range from 60 to 1100 pg L^{-1} with means of 350 and 240 pg L^{-1} at Ny Ålesund and Tromsø, respectively. Bulk densities ranged from 0.01 to 0.3 kg L^{-1} for snows of different types. The highest meltwater concentration was found in an event of fresh snow with dendritic crystals and low density. Scavenging of gaseous POPs by snow is favoured by high specific surface area (SSA) (Burniston et al., 2007; Herbert et al., 2005b; Lei and Wania, 2004). An empirical relationship in which snow bulk density was inversely proportional to SSA was found by Legagneux et al. (2002). Herbert et al. (2005a) found that the PCN concentrations in meltwater were correlated inversely to snow density at $p<0.01$ for tetra-CNs and penta-CNs and <0.1 for tri-CNs.

Atmospheric deposition of gaseous POPs to water surfaces can be estimated from concentrations in air, but a description of two-way gas exchange requires that dissolved phase concentrations in water be known. Very few measurements of dissolved PCNs in water have been made, and none in arctic–subarctic waters. Persson et al. (2005) measured dissolved and particulate PCNs in the Grenlandsfjords, southern Norway, but the data were only presented in the form of the particle/dissolved ratio. Measurement of gaseous PCNs in air and dissolved PCNs in water allowed air–water exchange to be assessed for Lake Ontario, with the result that trichloronaphthalenes were undergoing net volatilization whereas tetrachloronaphthalenes were close to air–water equilibrium (Helm et al., 2003a). PCNs have not been reported in the Arctic Ocean; however, reports of dissolved and particulate PCBs in the Arctic Ocean (Sobek and Gustafsson, 2004) suggest that PCNs would also be present. The occurrence of dissolved PCNs at some level in Bothnian Bay is implied, as they were found on suspended particles (Lundgren et al., 2002, 2003).

Helm (2002) observed a decrease in the ΣPCNs and the tri-, tetra- and penta-CN homologues in air with decreasing temperature at Resolute Bay. Plots of the Clausius–Clapeyron equation (ln partial pressure vs. $1/T$) yielded apparent enthalpies of air–surface exchange (ΔH_{EX}) of 68 kJ mol^{-1} for the ΣPCNs and 44–91 kJ mol^{-1} for the homologues. These are similar to enthalpies of vapourization and octanol–air partitioning, which implies regional air–surface exchange, as has been noted for PCNs in temperate regions (Lee et al., 2000).

5.4. Potential toxicity

Some studies report tetrachlorodibenzo-*p*-dioxin (TCDD) toxic equivalents (TEQ) in arctic air due to PCNs and "dioxin-like" PCBs (DL-PCBs). Assessments of PCNs TEQ in the studies cited below were based on those penta-, hexa- and hepta-CNs for which relative potencies (H4IIE-REPs) have been estimated (CNs 52, 54, 56, 57, 60, 61, 63, 64, 66, 67, 68, 69, 70, 71, and 73) (Blankenship et al., 2000; Villeneuve et al., 2000) and which were found in samples. The TEQ due to DL-PCBs were based on non-ortho CBs 77, 81, 126, and mono-ortho PCBs 105, 114, 118, and 156.

Harner et al. (1998) reported ΣTEQ (PCNs + DL-PCBs, fg m^{-3}) for air samples collected in the Barents Sea (2.0), eastern Arctic Ocean (0.81), Norwegian Sea (≤0.36), Alert (0.41) and Dunai (0.60). These can be compared to the much higher ΣTEQ in Chicago (11.6). The PCNs TEQ at arctic–subarctic sites accounted for 13–67% of ΣTEQ due to PCNs + DL-PCBs. Helm et al. (2004) reported that the ΣTEQ (PCNs + DL-PCBs) at Alert, Dunai and Tagish ranged from 0.006 fg m^{-3} during the warm period to 0.061 fg m^{-3} during the cold period. During winter, PCNs contributed 71–75% of ΣTEQ at Alert and Dunai, but only 30% at Tagish. In summer, DL-PCBs accounted for 65–98% of the ΣTEQ at all three sites.

No studies have been done in which PCNs, DL-PCBs and PCDD/Fs have all been measured in arctic air at the same time. Harner et al. (1998) noted that the TEQ due to PCDD/Fs (1.7 fg m^{-3}) were higher than for DL-PCBs (0.019 fg m^{-3}) in two air samples collected at Ny Ålesund by Schlabach et al. (1996). During winter 2000–2001, PCDD/Fs TEQ at Alert averaged 0.4 fg m^{-3} (Hung et al., 2002), which Helm et al. (2004) recalculated to 0.8 fg m^{-3} using HII4E-REPs. Thus, the various studies indicate that PCDD/Fs dominate ΣTEQ in some cases and in others are rather similar to the TEQ contributions from the other two compound classes.

6. PCNs in sediments

Few measurements have been made of PCNs in sediments from subarctic regions and there appear to be no measurements in arctic sediments. Reported concentrations on a dry weight basis are summarized below. The ΣPCNs (4–7 Cl) in surface sediment (0–2 cm) of Lake Storvindeln, Sweden (65.7°N, 17.1°E, sampled in the late 1980s) was 0.23 ng g^{-1} (Järnberg et al., 1997). Average concentrations ΣPCNs (4–7 Cl) were 0.088–1.9 ng g^{-1} in multiple samples of surface sediment (0–1 cm) collected at six stations in Bothnian Bay and Bothnian Sea, northern Baltic Sea (59.6–65.3°N, 17.5–22.5°E,

sampled 1991–1992), and 0.3–1.4 ng g^{-1} in suspended particulate matter from the same region. The ΣPCNs fluxes ranged from 0.49 to 0.93 μg m^{-2} y^{-1} (Lundgren et al. 2002, 2003). For comparison, ΣPCNs (4–7 Cl) in sediments of lakes and rivers in southern Sweden ranged from 0.6 to 252 ng g^{-1}, representing background and polluted sites (Järnberg et al., 1993, 1997), 6.7 ng g^{-1} in the Gdañsk Basin of the southern Baltic (Falandysz et al., 1996), and ~0.5–100 ng g^{-1} in settled particulate matter from Lake Mälaren and the Stockholm Archipelago (Ishaq et al., 2003). The homologue distribution in Lake Storvindeln sediment was: tetra-CNs 33%, penta-CNs 41%, hexa-CNs 15% and hepta-CNs 11%. Distributions in northern Baltic sediments were: tetra-CNs 44–72%, penta-CNs 24–32%, hexa-CNs 3–10% and hepta-CNs 0.6–8%.

No vertical profiles for PCNs are available for sediments in arctic–subarctic regions, so the historical record of PCN sedimentation cannot be assessed. Dated cores from a lake in the U.K. indicated that ΣPCNs deposition peaked in 1960, about 20 years ahead of ΣPCBs (Gevao et al., 2000). Profiles in two cores from a Swiss lake showed peak ΣPCNs deposition occurred in 1958–1961, while a third core showed an earlier maximum in 1928 (Bogdal et al., 2008). The maximum concentration of ΣPCNs in a core from Tokyo Bay occurred in 1980 (Yamashita et al., 2000b). Fluxes (pg cm^{-2}) of PCNs to Lake Kitaura in Japan were 0.073–0.31 before 1926, 5.5–14 in 1963–1970, 50–107 in 1971–1985 and 17 in 1997–2000. The relative contribution of combustion to evaporative marker congeners increased after the early 1980s (Horii et al., 2005). The ΣPCNs in archived samples of U.K. agricultural soils peaked in 1956, preceding the peak of ΣPCBs in 1970 (Meijer et al., 2001).

7. PCNs in biota

7.1. Total PCNs

The ΣPCN concentrations in arctic and subarctic biota are summarized in Table 3 for invertebrates and fish, and Table 4 for marine mammals and seabirds. Fig. 3 shows the ranges of arithmetic mean (AM) values regardless of location, and the geometric means of the AMs. Most studies give lipid-normalized results. Those that report on a wet weight basis also supply lipid percentages, and these were used to normalize ΣPCN concentrations to lipid content in the tables. The ΣPCNs reported here are on a lipid weight basis, unless stated otherwise. As for air, a problem with interpreting the ΣPCN concentrations in biota is that they are frequently based on different numbers of congeners, even in studies which report the same homologues.

7.1.1. Invertebrates

Few measurements of PCNs have been made in polar invertebrates. Evenset et al. (2005) determined PCNs in zooplankton (>90% *Cyclops abyssorum*, collected 1999) from Ellasjøen, a lake on Bear Island in the Barents Sea where high accumulation of other POPs had been found in biota and sediments (Evenset et al., 2004). The ΣPCNs averaged 0.15 ng g^{-1}, but only four congeners were measured, CNs 42, 52 and 66 + 67. The ΣPCNs (3–8 Cl) in krill (*Euphausia superba*) from the Ross Sea, Antarctica (collected 1994–1996) was 0.1 ng g^{-1} (Corsolini et al., 2002).

The remainder of PCN data for invertebrates comes from the subarctic Bothnian Bay/Sea, the northernmost basin of the Baltic Sea. Lundgren et al. (2002) measured ΣPCNs (4–7 Cl) in amphipods (*Monoporeia affinis*) and isopods (*Saduria entomon*) at five sites (collected 1991–1993) in the ranges of 12–69 ng g^{-1} and 3.9–16 ng g^{-1}, respectively.

Nfon et al. (2008) measured PCNs (4–6 Cl) in pelagic and benthic food chains of the Baltic Sea, from 1991–1993 surveys. Five of the stations were at the subarctic Baltic locations sampled by Lundgren et al. (2002) in the Bothnian Bay/Sea, while two others were in the central Baltic. The pelagic chain consisted of phytoplankton, zooplankton, mysids (*Mysis sp.*), and herring (*Clupea harengus*). Components of the benthic chain were amphipods, isopods and fourhorned sculpin, as in the Lundgren et al. (2002) study. The ΣPCN concentrations (ng g^{-1}) in the invertebrates were in the order: amphipods (28.5) > phytoplankton (17.1) > isopods (8.4) > zooplankton (4.0) = mysids (4.0). Detailed investigation was made of CN congener biomagnification in these food chains (Section 7.2).

7.1.2. Fish

Arctic char (*Salvelinus alpinus*) from two lakes on Bear Island (Ellasjøen and Øyangen, sampled 1999–2001, 3–7 fish in each lake) contained ΣPCNs averaging 0.85 and 0.43 ng g^{-1}. As for zooplankton in this study, only four congeners were measured (Evenset et al., 2005).

Livers of arctic cod (*Cadus callarias*) from Vestertana Fjord, Norway were monitored for PCNs from 1987 to 1998 (Sinkonnen and Paasivirta, 2000). The ΣPCNs (5–6 Cl) ranged from 0.13 to 1.06 ng g^{-1} and averaged 0.42 ng g^{-1}. No significant trends in ΣPCN concentration were found over the decade. In 1985–1989, salmon muscle from the Tana River in arctic Finland and cod livers from the Vestertana Fjord contained 0.052 and 0.17 ng g^{-1}, respectively, of hexachloronaphthalenes (Paasivirta and Rantio, 1991).

Liver of shorthorned sculpin (*Myoxocephalus scorpius*) from eastern and western Greenland (four pools of 5 fish each, collected 2002) were analysed for PCNs (3–7 Cl), but all congeners were below the LOQ (limit of quantification). An upper limit to ΣPCNs was estimated at 0.34 ng g^{-1}, obtained by substituting one half the LOQ for each monitored congener, (Vorkamp et al., 2004).

PCNs were determined in three species of antarctic fish: sharp spined notothen (*Trematomus pennelli*), crocodile icefish (*Chionodraco hamatus*) and silverfish (*Pleuragramma antarcticum*) (collected 1994–1996, 2–3 fish of each species). The ΣPCNs (3–8 Cl) in these species ranged from 0.11 to 0.91 ng g^{-1}, with the highest concentration in the silverfish (Corsolini et al., 2002).

As for invertebrates, the remainder of PCN data for fish comes from the northern Baltic Sea. The ΣPCNs (4–7 Cl) in fourhorned sculpin in the Bothnian Bay/Sea (sampled 1991–1993, five sites, 1–5 animals at each) ranged from 0.54 to 1.5 ng g^{-1} (Lundgren et al., 2002), the upper end being similar to the average 1.9 ng g^{-1} found by Nfon et al. (2008) at the same stations and within the same years.

Muscle of whitefish (*Coregonus* sp., 35, collected 1986) and pike (*Esox lucius*, pooled samples, collected 1988) and from Lake Storvindeln, Sweden each contained an average of 2.6 ng g^{-1} ΣPCNs (4–6 Cl) (Jansson et al., 1993; Järnberg et al., 1993). Liver and muscle of burbot (*Lota lota*, pooled samples, collected 1988) from the Bothnian Bay and the Torne River, Finland contained 1.0–4.9 ng g^{-1} ΣPCNs (4–6 Cl) (Järnberg et al., 1993).

Koistinen et al. (2008) collected herring (*C. harengus*) in the northern Baltic Sea and Gulf of Finland during 1999 and analysed them for a suite of POPs, including PCNs (4–8 Cl). Concentrations were reported per wet weight, but the lipid content was given, so the results are adjusted to a lipid weight basis here. Median ΣPCNs (ng g^{-1}) for small and large herring classes, respectively, were: Bothnian Bay (3.2, 5.6), Bothnian Sea (2.6, 4.8), Gulf of Finland (5.1, 3.3). PCNs (4–8 Cl) were determined in cleaned herring (head and guts removed) from the Bothnian Sea (90, collected 2002; Parmanne et al., 2006). Fish ranged in age from 1 to 18 years (mean 6.4 years) and results were sorted by yearly age classes, ranging from 2 to ≥10 years. Means within each age class ranged from 0.3 to 2.9 ng g^{-1} with an overall mean of 1.7 ng g^{-1}, similar to 1.9 ng g^{-1}, reported by Nfon et al. (2008). The ΣPCNs on a fresh weight basis were related to age (years), length (cm) and weight (g). The best fits to length and weight were expressed by $\Sigma PCNs = 0.521 \times \ln(\text{length}) - 1.354$ ($r^2 = 0.614$) and $\Sigma PCNs = 0.176 \times \ln(\text{weight}) - 0.475$ ($r^2 = 0.631$), while a power relationship was strongest for age: $\Sigma PCNs = 0.0108 \times \text{age}^{1.284}$ ($r^2 = 0.671$).

Table 3
ΣPCNs in invertebrate animals and fish, ng g^{-1} lipid.

Species	Location	Year	*N*	Tissue[a]	Sex	Range	Mean	Homologues	Reference[b]
Arctic									
Zooplankton[c]	Bear Island, Ellasjøen, Norway	1999	Pool[d]	W		0.14–0.16	0.15	4–6	1
Arctic char (*S. alpinus*), small	Bear Island, Ellasjøen, Norway	1999–2001	1	W			0.81	4–6	1
Arctic char (*S. alpinus*), large	Bear Island, Ellasjøen, Norway	1999–2001	6	W		0.45–1.4	0.85	4–6	1
Arctic char (*S. alpinus*), small	Bear Island, Øyangen, Norway	1999–2001	1	W			0.29	4–6	1
Arctic char (*S. alpinus*), large	Bear Island, Øyangen, Norway	1999–2001	2	W		0.32–0.68	0.50	4–6	1
Phytoplankton	Central/northern Baltic	1991–1993	Pool	W			17.1	4–6	2
Zooplankton	Central/northern Baltic	1991–1993	Pool	W			4.0	4–6	2
Amphipod (*M. affinis*)	Central/northern Baltic	1991–1993	Pool	W			28.5	4–6	2
Mysis (*Mysis* sp.)	Central/northern Baltic	1991–1993	Pool	W			4.0	4–6	2
Isopod (*S. entomon*)	Central/northern Baltic	1991–1993	Pool	W			8.4	4–6	2
Herring (*C. harengus*)	Central/northern Baltic	1991–1993	Pool	C			1.90	4–6	2
Fourhorned sculpin (*O. quadricornis*)	Central/northern Baltic	1991–1993	Pool	W			1.90	4–6	2
Amphipod (*M. affinis*)	Bothnian Bay/Sea	1991–1993	Pool	W		12–69	28	4–7	3
Isopod (*S. entomon*)	Bothnian Bay/Sea	1991–1993	Pool	W		3.9–16	9.2	4–7	3
Fourhorned sculpin (*O. quadricornis*)	Bothnian Bay/Sea	1991–1993	Pool	W		0.54–1.5	1.10	4–7	3
Whitefish (*Coregonus* sp.)	Lake Storvindeln, Sweden	1986	35	M			2.6	4–6	4
Arctic cod (*C. callarias*)	Vestertana Fjord, Norway	1987–1998	5 per year	L		0.13–1.06	0.42	5–6	5
Herring (*C. harengus*)	Bothnian Bay/Sea, Gulf of Finland	1999	Pool	C	M, F	2.6–5.6		4–8	6
Herring (*C. harengus*)	Bothnian Bay/Sea	2001–2003	Pool	C	M, F	0.53–3.4	2.4	4–8	7
Salmon (*S. salar*)	Bothnian Bay/Sea	2001–2003	Pool	C	M, F	1.6–2.5	2.0	4–8	7
Burbot (*L. lota*)	Bothnian Bay/Sea	2001–2003	Pool	C	M, F	1.6–1.8	1.8	4–8	7
Pike (*E. lucius*),	Bothnian Bay/Sea	2001–2003	Pool	C	M, F	2.3–5.0	3.6	4–8	7
Perch (*P. fluviatilis*)	Bothnian Bay/Sea	2001–2003	Pool	C	M, F	1.0–3.0	1.7	4–8	7
Whitefish (*C. lavaretus*)	Bothnian Bay/Sea	2001–2003	Pool	C	M, F	1.0–4.6	1.9	4–8	7
Vendace (*C. albula*)	Bothnian Bay	2001–2003	Pool	C	M, F	0.16–0.32	0.24	4–8	7
River lamprey (L. fluviatilis)	Bothnian Bay	2001–2003	Pool	C	M, F	0.88–1.0	0.93	4–8	7
Herring (*C. harengus*)	Bothnian Sea	2002	90	C	M, F	0.3–2.9	1.7	4–8	8
Burbot (*L. lota*)	Pajala, Finland	1988	Pool	M			2.9	4–8	9
Burbot (*L. lota*)	Pajala, Finland	1988	Pool	L			2.0	4–8	9
Burbot (*L. lota*)	Etukrunni, Finland	1988	Pool	M			4.9	4–8	9
Burbot (*L. lota*)	Etukrunni, Finland	1988	Pool	L			2.0	4–8	9
Burbot (*L. lota*)	Seskarø, Sweden	1988	Pool	M			4.4	4–8	9
Burbot (*L. lota*)	Seskarø, Sweden	1988	Pool	L			0.98	4–8	9
Pike (*E. lucius*)	Lake Storvindeln, Sweden	1988	Pool	M			2.6	4–8	9
Burbot (*L. lota*)	Lake Oulujärvi, Finland	2001–2003	Pool	M + S	M		1.3	4–8	7
Pike (*E. lucius*)	Lake Oulujärvi, Finland	2001–2003	Pool	M + S	M, F	1.0–1.2	1.1	4–8	7
Perch (*P. fluviatilis*)	Lake Oulujärvi, Finland	2001–2003	Pool	M + S	M, F	0.6–0.6	0.60	4–8	7
Pike-perch (*S. lucioperca*)	Lake Oulujärvi, Finland	2001–2003	Pool	M + S	M	0.16–0.24	0.20	4–8	7
Whitefish (*C. lavaretus*)	Lake Oulujärvi, Finland	2001–2003	Pool	M + S		0.29–0.33	0.31	4–8	7
Vendace (*C. albula*)	Lake Oulujärvi, Finland	2001–2003	Pool	C	M, F	0.53–0.68	0.60	4–8	7
Bream (*A. brama*)	Lake Oulujärvi, Finland	2001–2003	Pool	M + S	M, F	0.24–0.73	0.49	4–8	7
Antarctica									
Krill (*E. superba*)	Ross Sea	1994	Pool	W			0.10	3–8	10
Sharp spined notothen (*T. pennelli*)	Terra Nova Bay	1995	2	W		0.081–0.13	0.10	3–8	10
Crocodile fish (*C. hamatus*)	Terra Nova Bay	1995	2	W		0.12–0.15	0.12	3–8	10
Silverfish (*P. antarcticum*),	Ross Sea	1994	3	M			0.91	3–8	10

[a] W = whole, C = whole cleaned (see study), M = muscle, M + S = muscle + skin, L = liver.
[b] 1. Evenset et al. (2005). 2. Nfon et al. (2008). 3. Lundgren et al. (2002). 4. Jansson et al. (1993), 5. Sinkonnen and Paasivirta (2000). 6. Koistinen et al. (2008); 7. Isosaari et al. (2006); 8. Parmanne et al. (2006). 9. Järnberg et al. (1993). 10. Corsolini et al. (2002).
[c] *Cyclops abyssorum* (>90%), *Daphnia umbra* (<10%).
[d] Pooled multiple samples.

Other fish (pooled samples collected 2001–2003) from the Bothnian Bay/Sea and Lake Oulujärvi, a subarctic lake in Finland, included salmon (*Salmo salar*), burbot (*Lota lota*), pike, perch (*Perca fluviatilis*), pike-perch (*Stizostedion lucioperca*), whitefish (*Coregonus lavaretus*), vendace (*Coregonus albula*), river lamprey (*Lampetra fluviatilis*), and bream (*Abramis brama*) (Isosaari et al., 2006). Vendace were analysed whole after cleaning (head and guts removed), while a slice from behind the dorsal fin (skin on) was taken for other fishes. The ranges of average ΣPCNs (3–8 Cl) concentrations in these species were 0.24–3.6 ng g^{-1} in the Bothnian Bay/Sea and 0.20–1.3 ng g^{-1} in the lake. In both the Parmanne et al. (2006) and Isosaari et al. (2006) studies, ΣPCNs were reported as "upperbound" concentrations, where nondetectable congeners were replaced by LOQ values. LOQs were 0.002–0.01 ng g^{-1} for single congeners. Upperbound concentrations in 90% of the samples were ≤30% higher than lowerbound concentrations, calculated by assuming zero for nondetectable congeners.

7.1.3. Marine mammals

PCNs have been determined in blubber of ringed seal (*Phoca hispida*) from the Canadian Arctic, Svalbard and Greenland. Average ΣPCNs in eastern Canadian Arctic seals were 0.047 and 0.051 ng g^{-1} for 3 male (M) and 3 female (F) animals collected in 1993 (Helm et al., 2002). A subsequent study of eastern Canadian Arctic seals in 1999–2003 showed higher levels, averaging 0.33 ng g^{-1} in 2 M and 0.25 ng g^{-1} in 19 F, while the average ΣPCNs in 10 F seals from the western Canadian Arctic was 0.072 ng g^{-1} (Muir et al., 2004). The ΣPCNs in 7 F seals at Svalbard, collected in 1981, averaged 0.038 ng g^{-1} (Jansson et al., 1993). The ΣPCN averages of all ringed seal measurements span a fairly narrow range considering the locations, years and differences in reported homologues: 3–7 or 3–8 Cl in Canada and 4–6 Cl in Svalbard. Vorkamp et al. (2004) reported only an upper limit 0.13 ng g^{-1} for ΣPCNs (3–7 Cl) in east and west Greenland seals (M, four pools of 4–5 animals each, collected in 2002), as noted above for shorthorned sculpin.

Table 4
ΣPCNs in marine mammals and birds, ng g^{-1} lipid.

Species	Location	Year	*N*	Tissue[a]	Sex	Range	Mean	Homologues	Reference[b]
Arctic									
Ringed seal (*P. hispida*)	Pangnirtung, Canada	1993	3	B	F	0.035–0.071	0.051	3–7	1
Ringed seal (*P. hispida*)	Pangnirtung, Canada	1993	3	B	M	0.045–0.049	0.047	3–7	1
Ringed seal (*P. hispida*)	Pangnirtung, Canada	2002	2	B	M	0.12–0.54	0.32	3–8	2
Ringed seal (*P. hispida*)	Pangnirtung, Canada	1999–2002	12	B	F	0.09–0.42	0.23	3–8	2
Ringed seal (*P. hispida*)	Grise Fiord, Canada	2003	7	B	F	0.15–0.59	0.27	3–8	2
Ringed seal (*P. hispida*)	Sachs Harbour, Canada	2001	10	B	F	0.023–0.18	0.072	3–8	2
Ringed seal (*P. hispida*)	Kongsfjorden, Svalbard	1981	7	B	F		0.038	4–6	3
Ringed seal (*P. hispida*)	Ittoqqortoormiit, Greenland	2002	Pool[c]	B	M	All ND[d]	0.13[d]	3–7	4
Ringed seal (*P. hispida*)	Qeqertarsuaq, Greenland	2002	Pool	B	M	All ND[d]	0.12[d]	3–7	4
Harbour seal (*P. vitulina*)	Gulf of Alaska, U.S.A.	2000–2001	39	B	M, F	0.92–27	4.8	3–8	5
Harbour seal (*P. vitulina*)	Gulf of Alaska, U.S.A.	2000–2001	39	L	M, F	0.38–1.4	1.1	3–8	5
Harbour seal (*P. vitulina*)	Gulf of Alaska, U.S.A.	2000–2001	39	K	M, F	0.31–0.90	0.59	3–8	5
Beluga (*D. leucas*)	Kimmirut, Canada	1994	3	B	F	0.036–0.26	0.18	3–7	1
Beluga (*D. leucas*)	Kimmirut, Canada	1994	3	B	M	0.30–0.38	0.33	3–7	1
Beluga (*D. leucas*)	Hudson Strait, Canada	1999	8	B	M	0.20–0.89	0.42	3–8	2
Beluga (*D. leucas*)	Hudson Strait, Canada	1999	1	B	F		0.56	3–8	2
Beluga (*D. leucas*)	Nastapoka, Canada	2000	4	B	M	0.11–0.21	0.16	3–8	2
Beluga (*D. leucas*)	Nastapoka, Canada	2000	6	B	F	0.081–0.33	0.16	3–8	2
Minke whale (*B. acutorostrata*)	Greenland	1998	Pool	B	F	All ND[d]	0.18[d]	4–7	4
Longfinned pilot whale (*G. melas*)	Faroe Islands	2001	Pool	B	Juvenile		3.66	4–7	4
Longfinned pilot whale (*G. melas*)	Faroe Islands	2001	Pool	B	F		0.99	4–7	4
Longfinned pilot whale (*G. melas*)	Faroe Islands	2001	Pool	B	M		2.22	4–7	4
Polar bear (*U. maritimus*)	Alaska, U.S.A.	1997–1999	5	L			3.2	3–8	6
Polar bear (*U. maritimus*)	Ittoqqortoormiit, Greenland	1999–2002	Pool	F	M, F	0.49–0.53	0.51	4–7	
Glaucous gull (*L. hypoboreus*)	Bear Island, Norway	2002–2004	10	E		1.8–162	49.0	4–7	7
Glaucous gull (*L. hypoboreus*)	Bear Island, Norway	2002–2004	10	P	M	1.34–126	74.0	4–7	7
Glaucous gull (*L. hypoboreus*)	Bear Island, Norway	2002–2004	10	P	F	1.34–119	62.8	4–7	7
Glaucous gull (*L. hypoboreus*)	Ny Ålesund, Svalbard	2002	6–7	E			0.88	3–7	8
Glaucous gull (*L. hypoboreus*)	Festningisfjord, Svalbard	2002	Pool	E			0.97	3–7	8
Glaucous gull (*L. hypoboreus*)	Northwater Polynya, Canada	1998		L		4.1–17.1	10.6[e]	3–6	9
Black legged kittewake (*R. tridactyla*)	Northwater Polynya, Canada	1998		L		3.1–9.1	6.1[e]	3–6	9
Black guillemot (*C. grylle*)	Northwater Polynya, Canada	1998		L		8.0–29.5	18.8[e]	3–6	9
Thick–billed murre (*U. lomvia*)	Northwater Polynya, Canada	1998		L		1.7–8.7	5.2[e]	3–6	9
Thick-billed murre (*U. lomvia*)	Prince Leopold Island, Canada	2005	Pool	E		1.27–2.12	1.68	4–7	10
Great black-backed gull (*L. marinus*)	Vardø, Norway	2001	20	E			See *L.argentatus*	3–7	8
Great black-backed gull (*L. marinus*)	Kongsfjord, Norway	2001	20	E			2.16	3–7	8
Great black-backed gull (*L. marinus*)	Rolvsøya in Valfjord, Norway	2002	Pool[c]	E			1.35	3–7	8
Great black-backed gull (*L. marinus*)	Alta, Norway	2001	20	E			See *L.argentatus*	3–7	8
Great black-backed gull (*L. marinus*)	Sommarøy, Norway	2001	20	E			2.45	3–7	8
Great black-backed gull (*L. marinus*)	Lyngøyan in Øksnes, Norway	2002	20	E			See *L.argentatus*	3–7	8
Great black-backed gull (*L. marinus*)	Kirkjubøreyni, Faroe Islands	2002	20	E			0.96	3–7	8
Lesser black-backed gull (*L. fuscus*)	Góðadular, Faroe Islands	2002	20	E			0.50	3–7	8
Herring gull (*L. argentatus*)+*L.marinus*	Vardø, Norway	2001	20	E			1.39	3–7	8
Herring gull (*L. argentatus*)	Rolvsøya in Valfjord, Norway	2002	Pool	E			2.06	3–7	8
Herring gull (*L. argentatus*)+*L.marinus*	Alta, Norway	2001	20	E			1.06	3–7	8
Herring gull (*L. argentatus*)	Lyngøyan in Øksnes, Norway	2002	20	E			1.60	3–7	8
Herring gull (*L. argentatus*)	Saltstraumen, Norway	2002	15	E			2.44	3–7	8
Herring gull (*L. argentatus*)	Givaer, Norway	2002	10	E			4.65	3–7	8
Northern fulmar (*F. glacialis*)	Faroe Islands	1998–1999	Pool	F	F		2.55	4–7	4
Northern fulmar (*F. glacialis*)	Faroe Islands	1998–1999	Pool	F	M		3.75	4–7	4
Northern fulmar (*F. glacialis*)	Prince Leopold Island, Canada	2005	Pool	E		1.64–2.56	2.0	4–7	10
Northern fulmar (*F. glacialis*)	Prince Leopold Island, Canada	2003	Pool	E			1.3	3–8	2
Northern fulmar (*F. glacialis*)	Cape Vera, Canada	2003	Pool	E			1.4	3–8	2
Northern fulmar (*F. glacialis*)	Northwater Polynya, Canada	1998		L		17.6–48.2	32.9[e]	3–6	9
Antarctica									
South polar skua (*C. maccormicki*)	Terra Nova Bay	1994	1	L			6.07	3–8	6
South polar skua (*C. maccormicki*)	Terra Nova Bay	1994	1	M			5.71	3–8	6
Weddell seal (*L. weddelli*)	Terra Nova Bay	1996	1	B			0.077	3–8	6
Weddell seal (*L. weddelli*)	Terra Nova Bay	1997	1	L			1.6	3–8	6

[a] B = blubber, L = liver, K = kidney, M = muscle, E = egg, P = plasma, F = fat.
[b] 1. Helm et al., 2002. 2. Muir et al., 2004. 3. Jansson et al., 1993. 4. Vorkamp et al., 2004, 5. Wang et al., 2007. 6. Corsolini et al., 2002. 7. Verreault et al., 2005, 8. Pusch and Gabrielsen, unpublished, 9. Helm et al., 2003b, 10. Braune et.al., unpublished.
[c] Pooled multiple samples.
[d] All congeners not detectable (below LOQs), upper limit estimated by substituting 1/2 LOQ values.
[e] Midpoint of range.

Substantially higher ΣPCNs (3–8 Cl) were reported for 39 harbour seal (*Phoca vitulina*) from the Gulf of Alaska (Prince William Sound and Kodiak Island, collected 2000–2001). Blubber, liver and kidney were analysed and results were grouped by age and sex (Wang et al., 2007). Overall averages for blubber, liver and kidney were 4.8 ng g^{-1}, 1.1 ng g^{-1} and 0.59 ng g^{-1}, respectively. The animals ranged in age from pups (<1 year), juveniles (1–4 years) and adults (7–15 years). The ΣPCN concentrations in blubber were positively and significantly

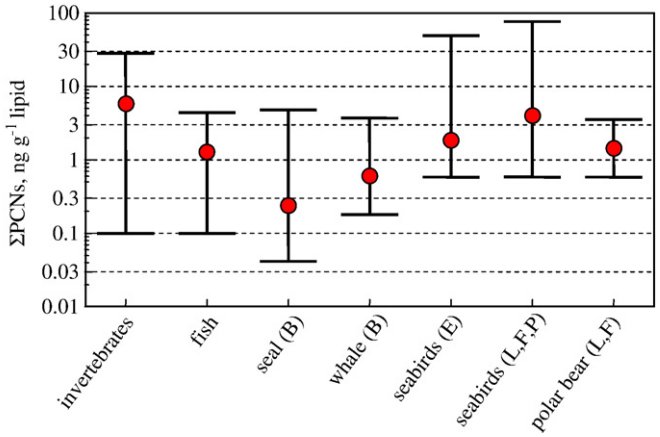


Fig. 3. Range of arithmetic mean (AM) values, and geometric means of AMs, for ΣPCNs (ng g^{-1} lipid weight) reported for invertebrates, fish, seal, whale, seabirds and polar bear. See Tables 3 and 4 for summarized data. B = blubber, F = fat, L = liver, E = egg, P = plasma.

($p<0.05$) correlated with age ($r^2=0.478$) and blubber thickness ($r^2=0.490$) in M, but not in F.

Weddell seal (*Leptonichotes weddelli*) from Terra Nova Bay, Antarctica (blubber of one seal collected 1996, liver of another collected in 1997) were analysed, with the resulting ΣPCN (3–8 Cl) concentrations of 0.077 ng g^{-1} in blubber and 1.63 ng g^{-1} in liver. Reported lipid contents were 100% in blubber and 2.7% in liver, so on a wet basis the ΣPCN concentration in liver were lower (0.044 ng g^{-1}) (Corsolini et al., 2002).

The ΣPCNs (3–7 Cl) in blubber of eastern Canadian Arctic beluga (*Delphinapterus leucas*) collected in 1994 averaged 0.33 ng g^{-1} in 3 M and 0.18 ng g^{-1} in 3 F (Helm et al., 2002). Similar levels were found in 1999–2000 (0.33 ng g^{-1} in 12 M, 0.19 ng g^{-1} in 9 F) (Muir et al., 2004). The upper limit (see above) for the ΣPCNs (3–7 Cl) in blubber of minke whale (*Balaenoptera acutorostrata*) from Greenland (one pool of 5 F, one pool of 5 M + F, collected in 1998) was estimated at 0.18 ng g^{-1} (Vorkamp et al., 2004).

PCNs were reported in blubber of longfinned pilot whale (*Globicephala melas*) from the Faroe Islands (one pool of 14 juveniles M + F, one pool of 34 adult F, one pool of 5 adult M, collected in 2001). The highest ΣPCNs (3–7 Cl) was found in the juveniles (3.7 ng g^{-1}), followed by M (2.2 ng g^{-1}) and F (0.99 ng g^{-1}). Although the Faroe Island pilot whales were from the same study as the Greenland ringed seal and minke whale, the ΣPCNs in pilot whale are not upper limits because they were obtained by summing quantifiable hexa- and hepta-CNs (Vorkamp et al., 2004).

For comparison, the ΣPCNs in blubber of killer whale (*Orcinus orca*) from the northeastern Pacific Ocean averaged 21.6 ± 6.7 and 20.4 ± 14.6 ng g^{-1} for northern and southern resident whales and 167 ± 131 ng g^{-1} in transient whales (Rayne et al., 2004).

Livers of five polar bear (*Ursus maritimus*) from Alaska (collected in 1997–1998) contained ΣPCNs (3–8 Cl)<0.0001–0.94 (mean 0.37) ng g^{-1} ww (Corsolini et al., 2002). The percent lipid ranged from 7.8 to 14.9% (mean 11.4%), but since lipid percentages were not associated with individual animals, an average lipid-normalized concentration of 3.2 ng g^{-1} ΣPCNs is calculated here. Fat samples from two Greenland polar bear contained 0.49–0.53 ng g^{-1} ΣPCNs, represented by a single quantifiable congener, CN 68. Other congeners were below the LOQ (Vorkamp et al., 2004).

7.1.4. Seabirds

PCNs were determined in eggs (10) and plasma (10 M, 10 F) of glaucous gull (*Larus hypoboreus*) from Bear Island, Norway (Verreault et al., 2005). The ΣPCNs (3–7 Cl) averaged 49, 74 and 63 ng g^{-1} in eggs, M plasma and F plasma, respectively.

Pusch and Gabrielsen (unpublished) sampled eggs of four gull species (herring gull, *Larus argentatus*; great black-backed gull, *Larus marinus*; lesser black-backed gull, *Larus fuscus*; and glaucous gull) from Svalbard, coastal locations in northern Norway and the Faroe Islands, between 2001–2002. Eggs were pooled for some species and locations, and analysed individually for others. The ΣPCNs (4–7 Cl) ranged from 0.50 ng g^{-1} at Góðadular, Faroe Islands to 4.7 ng g^{-1} at Givaer, Norway, with a mean of 1.7 ng g^{-1} for all birds at all locations.

Northern fulmar (*Fulmaris glacialis*) from the Faroe Islands (collected 1998–1999, two pools of fat samples from 6 juvenile M and 8 juvenile F) contained 3.8 ng g^{-1} (M) and 2.6 ng g^{-1} (F) of ΣPCNs (3–7 Cl). These are not upper limits, but obtained from summing congeners above the LOQ (Vorkamp et al., 2004).

The ΣPCNs in the livers of seabirds from the Northwater Polynya in the eastern Canadian Archipelago (glaucous gull; black-legged kittiwake, *Rissa tridacthyla*; thick-billed murre — also known as Brünnich's guillemot, *Uria lomvia*; black guillemot, *Cepphus grylle*; and northern fulmar) ranged from 1.7 to 48.2 ng g^{-1}, with highest concentrations in fulmar (Helm et al., 2003b). A study of POPs in seabird eggs collected in 1975 and 1993 from Prince Leopold Island in the Canadian Arctic found no detectable PCNs; however detection limits were high (2 ng g^{-1} wet weight) (Braune and Simon, 2004). A 2005 study of northern fulmar and thick-billed murre eggs from Prince Leopold Island (five pools of 3 eggs from each species) showed ΣPCNs (4–7 Cl) averaging 2.0 and 1.7 ng g^{-1} on a lipid weight basis (Braune et al., unpublished). Northern fulmar eggs from Prince Leopold Island and Cape Vera, Canada, contained ΣPCNs (3–8 Cl) in the range 1.3–1.4 ng g^{-1} (Muir et al., 2004).

The ΣPCNs (3–8 Cl) in south polar skua (*Catharacta maccormicki*) from Terra Nova Bay, Antarctica (collected 1994) was 6.1 ng g^{-1} in the liver of one bird and 5.7 ng g^{-1} in the muscle of a second bird (Corsolini et al., 2002).

7.2. Homologue and congener profiles, and biomagnification in food webs

The homologues included in ΣPCNs are listed in Tables 3 and 4. PCNs sought in biota are typically the tri- to octa- homologues. Log K_{OW} values for these homologues increase with the number of chlorines, ranging from about 5.5 for tri-CNs, 5.1–6.1 for tetra-CNs, 5.7–6.5 for penta-CNs and 6.0–6.7 for heavier homologues (Table 1), indicating that each of these homologues is subject to bioaccumulation. Bioaccumulation processes tend to alter homolog and congener profiles in biota relative to source signatures in abiotic compartments. Although bioaccumulation and biomagnification favour the heavier PCNs, other factors governing profiles in biota are differing congener distributions in sources (Järnberg et al., 1997) and selective metabolism, which eliminates certain congeners during trophic transfer. Congeners having vicinal carbons which are not substituted with chlorine are more readily metabolized or excreted by vertebrates due to hydroxylation via arene oxide intermediates, hydroxylation–dechlorination and via the mercapturic acid pathway (Chu et al., 1977; Falandysz, 1998; Jakobsson and Asplund, 2000). Congeners most amenable to biomagnification are those that do not contain vicinal carbons unsubstituted with chlorines (NVC-Cl). There are 15 of these NVC-Cl congeners, CNs 42, 52, 58, 60, 61, 64, 66, 67, 68, 69, 71, 72, 73, 74 and 75 (Falandysz, 1998; Helm et al., 2008; Jakobsson and Asplund, 2000). Generally, tetra-CNs with vicinal hydrogen atoms in $\beta\beta$-positions are more easily metabolized than tetra-CNs with vicinal hydrogen atoms in $\alpha\beta$-positions (Falandysz, 1998; Jakobssen and Asplund, 2000).

Biomagnification can be expressed between two species in a predator–prey relationship using a biomagnification factor (BMF), or in food webs using a food chain magnification factor (FCMF) or trophic magnification factor (Jardine et al., 2006; Nfon et al., 2008). BMFs are expressed as the lipid-adjusted concentration in a predator

divided by the lipid-adjusted concentration in its prey. BMFs can be adjusted for the trophic level of the predator and prey as determined from stable nitrogen isotope (δ^{15}N) measurements by:

$$BMF_{TL} = (concentrations\ in\ predator\ /\ prey)\ /\ (TL_{predator}\ /\ TL_{prey}).$$

Food chain magnification factors (FCMFs) are determined by first identifying the relationship between concentration and δ^{15}N by:

$$\ln(CN\ concentration) = A + B{*}\delta^{15}N$$

then, utilizing the slope of this relation, FCMF = e^B.

Homologue profiles of PCNs in arctic biota are summarized in Fig. 4. The result of selective metabolism and biomagnification is that the proportion of higher PCN homologues tends to increase with trophic position, although there are strong species differences in homologue and congener accumulation patterns. Such discrimination has been reported in food webs of the Laurentian Great Lakes (Hanari et al., 2004; Helm et al., 2006, 2008) and the southern Baltic Sea (Falandysz, 1998; Falandysz and Rappe, 1996; Falandysz et al., 1996; Järnberg et al., 1993).

Among the four congeners analysed in the two lakes on Bear Island, CN 52 was the most abundant in Øyangen and 66/67 in Ellasjøen (Evenset et al., 2005). In the Ellasjøen food chain, ΣPCN increased significantly ($p = 0.006$) with δ^{15}N from zooplankton to small and large arctic char. BMFs between small char/zooplankton were: CN 42 = 9.5, CN 52 = 5.3, and CN 66/67 = 3.6, while BMFs between large char/small char were 0.6, 6.4 and 0.8 for these congeners.

The ΣPCNs in antarctic krill comprised 55% tri-CNs, 14% tetra-CNs, and 31% penta-CNs. No heavier CNs were found (Corsolini et al., 2002). Two fish species (sharp-spined notothen and silverfish) also contained a predominance of tri-CNs (50–81%) followed by tetra-CNs (19–27%). No heavier CNs were found in notothen. Heavier PCNs in silverfish were penta-CNs (7.5%) and hepta-CNs (16%), but no hexa-CNs. Crocodile icefish was unusual in having a low proportion of tri-CNs (0–17%), mainly penta-CNs (83–100%), and no other homologues.

Within the benthic food chain in the Bothnian Bay/Sea, the proportion of tetra-CNs in ΣPCNs decreased from amphipod (69%) to isopod (41%) to fourhorned sculpin (31%) (Lundgren et al., 2002). Penta-CNs accounted for 22%, 44%, and 27% of ΣPCNs in these animals. Proportions of the heavier CNs increased within the food chain: hexa-CNs (7.6%, 13% and 37%) and hepta-CNs (1.5%, 2.6% and 5.3%). Tri-CNs were not measured. In general, the profiles of CN congeners in amphipods resembled those in sediments, while shifts away from the sediment profiles were found in isopods and sculpin. The three species showed differences in congener profiles within a homologue

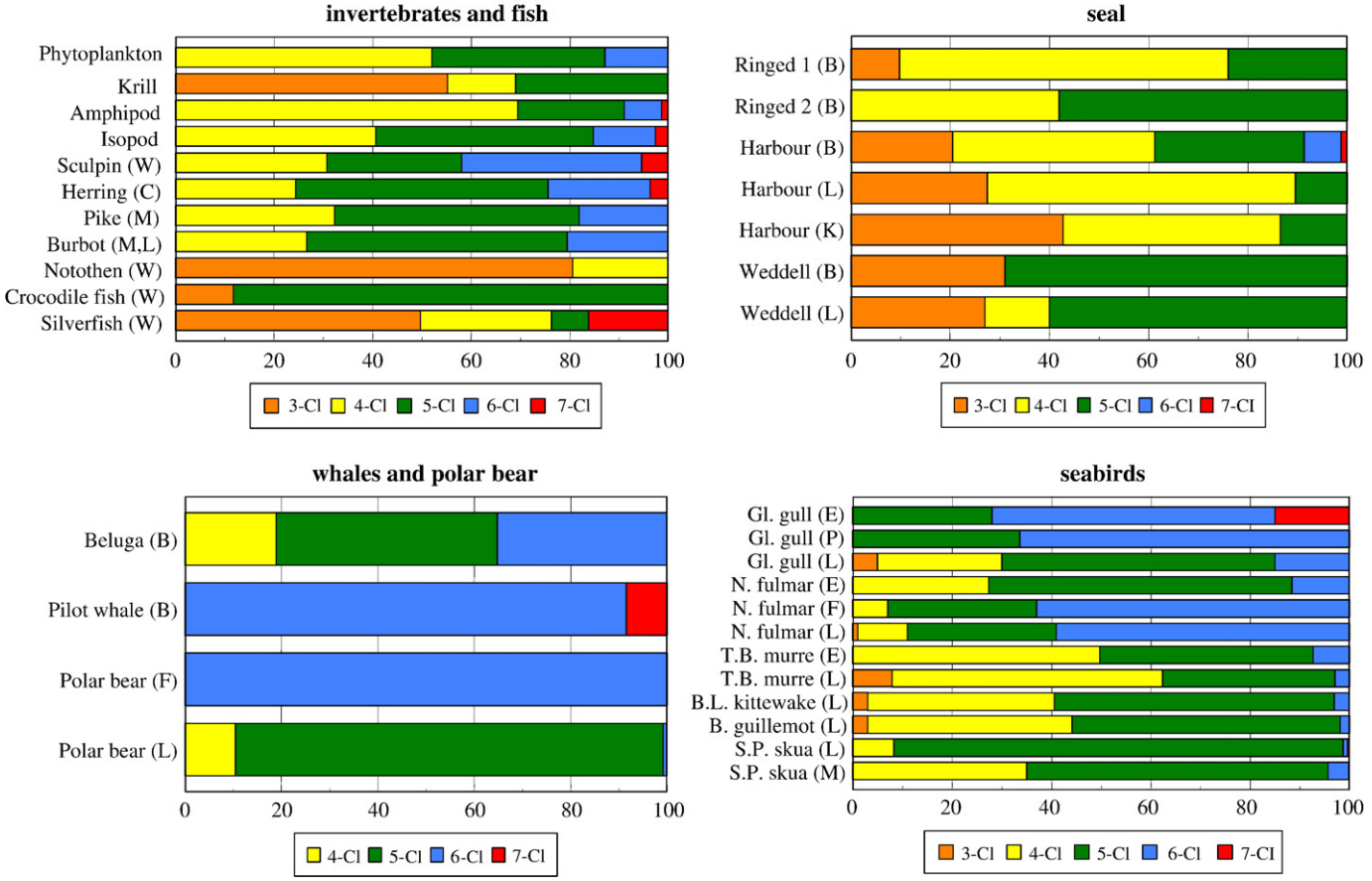


Fig. 4. PCN homologues (% of ΣPCNs) in arctic, subarctic and antarctic biota. Symbols in parentheses: W = whole, M = muscle, L = liver, C = cleaned (head and guts removed), B = blubber, F = fat, P = plasma. Reports containing three or more homologues were included; those containing only upper limits were excluded Lack of a homologue may indicate not found or not measured, see original study. When ranges were given, the midpoints of the range were taken, resulting in central values for homologue percentages which did not always sum to 100%. In these cases, figure data were normalized to 100%. Organisms studied (homologues analysed) and data sources are listed. Invertebrates and fish: phytoplankton (4–6) (Nfon et al., 2008); krill (3–8) (Corsolini et al., 2002); isopod, fourhorned sculpin (4–7) (Lundgren et al., 2002); herring (4–8) (Parmanne et al., 2006); pike, burbot (4–8) (Järnberg et al., 1993); notothen, crocodile fish, silverfish (3–8) (Corsolini et al., 2002). Seal: ringed seal 1 (3–7) (Helm et al., 2002), ringed seal 2 (4–6) (Jansson et al., 1993); harbour seal (3–8) (Wang et al., 2007); Weddell seal (3–8) (Corsolini et al., 2002). Whales and polar bear: beluga whale (3–7) (Helm et al., 2002); longfinned pilot whale (4–7) (Vorkamp et al., 2004); polar bear (3–8) (Corsolini et al., 2002). Seabirds: glaucous gull (4–7) (Verreault et al., 2005), (3–6) (Helm et al., 2003b); northern fulmar (4–7) (Vorkamp et al., 2004), (3–8) (Helm et al., 2003b; Muir et al., 2004); black-legged kittewake (3–8) (Helm et al., 2003b); black gullemot (3–8) (Helm et al., 2003b); thick-billed murre (4–6) (Braune et al., unpublished), (3–8) (Helm et al., 2003b); south polar skua (3–8) (Corsolini et al., 2002).

which suggested congener-specific absorption, metabolic transformation and excretion. Biota-sediment accumulation factors (BSAFs) for amphipods averaged 2.9, 1.4, 0.88 and 0.90 for tetra-, penta-, hexa- and hepta-CNs. BMFs for the four homologues between isopod/amphipod averaged 0.27, 0.92. 1.3 and 0.69, while those for sculpin/isopod averaged 0.13, 0.092, 0.69 and 0.22. Mean BMFs for sculpin/amphipod were 0.04, 0.10, 0.65 and 0.23. Within each homologue, BSAFs for amphipods varied little among congeners, ranging over a factor of three for tetra-CNs and factors of two for penta- and hexa-CNs. Greater variability was found for BMFs, especially for sculpin/isopod and sculpin/amphipod, for which congener BMFs spanned an order of magnitude or more within each homologue. Only congeners CN 66/67 and CN 69 biomagnified between sculpin/amphipod (BMFs of 1.1 and 1.4, respectively). Lundgren et al. (2002) noted higher sculpin/amphipod BMFs for penta-CNs congeners with NVC-Cl structures (52, 58, 60 and 61). Penta-CN without NVC-Cl structures (49, 51, 53, 56, 57, 59, 62) exhibited lower BMFs. Among the penta-CNs, the highest BMF was for CN 54 (1,2,3,6,7-PeCN), which has a lateral substitution similar to 2,3,7,8-TCDD.

Nfon et al. (2008) studied biomagnification of PCNs in a pelagic and benthic food chain in the Baltic Sea (Section 7.1.1), taking into account trophic levels. BMFs were reported for 28 congeners in each step of the food chain. Congener-specific FCMFs ranged from 0.66–1.29 in the pelagic food chain to 0.36–1.52 in the benthic food chain. FCMFs increased with average log K_{OW} values, tetra-CNs<penta-CNs<hexa-CNs, but great differences were found among congeners within a homologue. The highest two FCMFs were for CNs 68 and 54 (1.29, 1.28) in the pelagic chain and CNs 63 and 72 (1.52, 1.50) in the benthic chain. By comparison, FCMFs for tetra- to heptachlorinated PCB congeners ranged from 1.22–1.48 in the pelagic chain to 0.60–1.45 in the benthic chain. Nfon et al. (2008) concluded that FCMFs for PCNs generally indicated trophic dilution, especially for the lower homologues.

The Nfon et al. (2008) FCMF data are examined further here. Large differences were not seen in FCMFs as related to NVC-Cl substitution for the lower homologues. Within the pelagic chain, the FCMF for the single NVC-Cl congener CN 42 was 0.76, compared to 0.66–0.89 (mean 0.75) for ten other tetra-CNs without the NVC-Cl feature. Three NVC-Cl penta-CNs (58, 60, 61) had FCMFs ranging from 0.87 to 1.16 (mean 0.99), while eight other penta-CNs that were not NVC-Cl congeners had FCMFs in the range 0.84–1.28 (mean 1.01). Four NVC-Cl hexa-CNs (64, 66, 68 and 72) showed slightly higher FCMFs (1.06–1.29, mean 1.16) compared to two hexa-CNs that were not NVC-Cl congeners (63 and 65, FCMFs 1.09 and 0.94, respectively). Note: methods used by Nfon et al. (2008) probably did not resolve CN 52 from 60, or 66 from 67, so the FCMFs above include these congeners. In the benthic chain, the FCMF for CN 42 (0.38) was actually low compared to 0.41–0.89 (mean 0.60) for tetra-CNs without NVC-Cl. FCMFs for NVC-Cl and non- NVC-Cl penta-CNs were 0.43–0.74 (mean 0.55) and 0.36–1.11 (mean 0.72), respectively. FCMFs for NVC-Cl hexa-CNs ranged from 0.83 to 1.50 (mean 1.18) vs. 1.38–1.52 for the two non-NVC-Cl congeners.

Herring from the Bothnian Sea were dominated by penta-CNs (51% of ΣPCN), followed by tetra-CNs (24%), hexa-CNs (21%) and hepta-CNs (3.7%) (Parmanne et al., 2006). Penta-CNs were represented largely by one congener CN 52, (45.9% of ΣPCNs) with CNs 53 and 54 accounting for 3.0 and 2.3% of ΣPCNs. Other prominent congeners were CN 42 (19.6%) and CN 66/67 (11.9%). Tri-CNs were not measured. The ΣPCN concentrations increased significantly with age (Section 7.1.2) and some congeners (CNs 48, 66/67, 68 and 73) accumulated more strongly with age than ΣPCNs. Major congeners in the survey of herring in the Northern Baltic Sea by Koistinen et al. (2008) were CNs 52, 42, 66/67, 68 and 73.

Homologue profiles in burbot liver and muscle from Bothnian Bay and the Torne River, Finland, and pike muscle from Lake Storvindeln, Sweden contained mainly penta-CNs (47–57%), followed by tetra-CNs (22–36%). Hexa-CNs were also prominent (7.3–30%). Tri-CNs were not measured (Järnberg et al., 1997).

Tetra-CNs were the main components (70%) of ΣPCNs in ringed seal from the Canadian Arctic, followed by penta-CNs (25%) and tri-CNs (10%) (Helm et al., 2002). Hexa- and hepta-CNs were not found. Penta-CNs (58% of ΣPCNs) outweighed tetra-CNs (42%) in ringed seal from Svalbard (Jansson et al., 1993). No hexa-CNs were detected; tri- and hepta-CNs were not measured.

Homologues as percentages of ΣPCNs in the blubber of harbour seal from Alaska were: tetra-CNs 46%, penta-CNs 25%, tri-CNs 23%, hexa-CNs 8% and hepta-CNs 1%. By comparison, lower PCNs predominated in kidney (tri-CNs 45%, tetra-CNs 46%, penta-CNs 14%, no hexa- or hepta-CNs) and liver (tetra-CNs 65%, tri-CNs 29%, penta-CNs 11%, no hexa- or hepta-CNs) (Wang et al., 2007).

Penta-CNs were the main group of ΣPCNs in blubber and liver of Weddell seal (60% and 69%, respectively). Tri-CNs accounted for 31% and 27% of ΣPCNs in these organs. Tetra-CNs amounting to 13% of ΣPCNs were found in liver and none in blubber. No higher CNs were found. The only congeners found in the higher homologues were CN 57 in blubber and CNs 54 and 73 in liver (Corsolini et al., 2002). Overall, there was no strong evidence for biomagnification among the antarctic organisms. The ΣPCNs in krill (0.10 ng g^{-1}) were about the same as in two fish species (0.10–0.12 ng g^{-1}) and in Weddell seal (0.077 ng g^{-1}).

The ΣPCNs in Canadian Arctic beluga was dominated by penta-CNs (46%), followed by hexa-CNs (34%) and tetra-CNs (19%). No tri- or hepta-CNs were found (Helm et al., 2002). Only four CN congeners were quantifiable in longfinned pilot whale from the Faroe Islands, CNs 66/67 and 68 from the hexa-CN homologue (92%) and hepta-CN 74 (8% of ΣPCNs) (Vorkamp et al., 2004).

Hexa-CNs was the main homologue in eggs and plasma of glaucous gulls from Bear Island, accounting for 57 and 65% of ΣPCNs (Verreault et al., 2005). The contribution of penta-CNs was 28% and 33%. Hepta-CNs amounted to 15% of ΣPCNs in eggs, but were not found in plasma. No tetra-CNs were found, and tri-CNs were not measured. The most abundant congeners in eggs were CNs 66/67, 52/60, 71/72 and 73; while in plasma only CNs 66/67 and 52/60 were found.

The ΣPCNs in livers of black-legged kittewake, black guillemot and thick-billed murre from the Canadian Arctic were dominated by tetra- and penta-CNs, with tri-CNs and hexa-CNs accounting for 10% or less. Hexa-CNs were ~15% of ΣPCNs in glaucous gull liver (Helm et al., 2003b). Hexa-CNs contributed ~60% to ΣPCNs in northern fulmar liver (Helm et al., 2003b) but only 12% in fulmar eggs, where tetra- and penta-CNs made up 27% and 61% of ΣPCNs, respectively (hepta-CNs were not found and tri-CNs were not measured) (Braune et al., unpublished). The composition of ΣPCNs in thick-billed murre eggs from the Canadian Arctic was 50% tetra-CNs, 43% penta-CNs and 7.3% hexa-CNs. No hepta-CNs were found and tri-CNs were not measured.

Penta-CNs dominated in the liver of one south polar skua and in the muscle of another bird (90% and 61% of ΣPCNs, respectively). Tetra-CNs accounted for 35% of ΣPCNs in muscle, and 8.3% in liver. Hexa-CNs were 4.2% of ΣPCNs in muscle and hepta-CNs were not found. Other homologues were minor in skua liver (tri-CNs 0.1%, hexa-CNs 0.9%, hepta-CNs 0.3%). The most abundant congeners within the higher homologues were CNs 52/60, 54, 57 and 66/67 in liver, and 52/60, 73, and 66/67 in muscle (Corsolini et al., 2002).

Polar bear liver from Alaska contained homologues in the following percentages of ΣPCNs: penta-CNs 89%, tetra-CNs 10% and hexa-CNs 1%. Main congeners were CNs 54, 52/60, 57, 56 and 66/67 (Corsolini et al., 2002). Polar bear fat from Greenland contained a single quantifiable congener, CN 68 (Vorkamp et al., 2004).

Similar biomagnification trends have been found for PCNs in temperate ecosystems. Järnberg et al. (1993) observed that PCNs were very low in grey seal (*Halichoreus grypus*) and porpoise (*Phocaena phocaena*) in the Baltic Sea, with only one quantifiable tetra-CN peak. This result is in contrast to the study of Falandysz and Rappe (1996),

who found several congeners from the tetra- to hepta-CN homologues in porpoise from the southern Baltic. The ΣPCNs increased from 7.5–20 ng g^{-1} in plankton to 29 ng g^{-1} in herring, but then dropped to 1.7–2.8 ng g^{-1} in porpoise blubber. Large differences in BAFs and BMFs occurred on a congener-specific basis. BAFs from plankton to herring were <5 for most tetra-CNs except CN 42 (BAF ~7.5), but ranged from ~3–15 for penta-CNs and 10–23 for hexa-CNs. CNs 66/67 increased from 2% of ΣPCNs in herring to 20–30% in harbour porpoise, while CNs 52/60 and 61 decreased from 15% in herring to 4–6% in porpoise. The highest BMFs between herring and porpoise were for CNs 66/67 (BMF 1.2).

Log ΣPCNs increased with $\delta^{15}N$ in a Lake Ontario food web consisting of plankton, mysids (*Mysis relicta*) and amphipods (*Diporeia hoyi*), forage fish (alewife, *Alosa pseudoharengus*; rainbow smelt, *Osmerus mordax*; slimy sculpin, *Cottus cognatus*) and lake trout (*Salvelinus namaycush*). FCMFs were 1.25 for ΣPCNs and 1.42–1.43 for CNs 67 and 66, respectively (Helm et al., 2008). Principal component analysis was successful in discriminating congener distributions of congeners having differing numbers of vicinal carbons unsubstituted with chlorine. BMFs ranged from 5 to 10 for the NVC-Cl congeners in the tetra- to hexaCN homologs in lake trout relative to their prey. However BMFs varied by feeding relationship, as BMFs ranged from 0.5 to 3 for the same NVC-Cl congeners in slimy sculpin relative to their *Diporeia* diet (Helm et al., 2008).

BMFs for 4-Cl through 8-Cl congeners between algae/zebra mussels (*Dreissena polymorpha*) ranged from 3 to 10 in the Lake Erie–Detroit River–Lake St. Claire system, but there was no further increase for most congeners in round goby (*Neogobius melanostomus*), which prey on the zebra mussels. The exceptions were for CNs 52/60 and 66/67, which showed goby/mussel BMFs of 1.6 and 2.2, respectively (Hanari et al., 2004).

Järnberg et al. (1993) found that several CN congeners biomagnified from herring to guillemot (*Uria aalge*). BMFs were <5 for most tetra-, penta-, and hexa-CNs, but ~25 for CNs 66/67 and ~15 for CN-73.

Falandysz et al. (1997) found that PCNs biomagnified from fish to black cormorant (*Phalacrocorax carbo sinensis*) in the Gulf of Gdañsk, southern Baltic Sea. Dominating congeners in liver and breast muscle were 33/34/37, 42, 56/60, 58, 61, 62, and 66/67, with highest BMFs for the hexa-CNs (BMFs 4–14) and CN 42 (BMFs 4–10). Among the penta-CNs, congeners 50, 51, 52/60, 54, 57, 58, 61 and 62 had BMFs ranging from 1 to 4, while the penta-CNs 49, 53, 55, 56, 59 had BMFs <1.

Hexa-CNs (CNs 66/67) were the main homologue found in herring gull eggs from the Laurentian Great Lakes, while penta- (mainly CNs 52/60) and hexa-CNs were approximately equal in eggs of double crested cormorants (*Phalacrocorax auritus*) (Kannan et al., 2001).

Some studies which provided congener-specific data reported CN 54. This was the most abundant penta-CN in Alaskan polar bear liver, the only penta-CN found in silverfish and Weddell seal liver from the Ross Sea and a prominent penta-CN in liver of South Polar skua (Corsolini et al., 2002). CN 54 was found in the pelagic and benthic food webs in the Bothnian Bay/Sea (Lundgren et al., 2002; Nfon et al., 2008), herring from the Bothnian Sea (Koistinen et al., 2008; Parmanne et al., 2006) and seabird eggs from northern Norway, Svalbard and the Faroe Islands (Pusch and Gabrielsen unpublished). CN 54 also occurs in arctic air samples (Section 3) and in biota of temperate ecosystems (Falandysz, 1998; Falandysz et al., 1997; Falandysz and Rappe, 1996; Falandysz et al., 1996; Helm et al., 2008; Järnberg et al., 1993, 1997). CN 54 may be overlooked in studies that use only Halowax mixtures as standards, because Halowaxes do not contain or contain only trace amounts of CN 54, which is usually considered a combustion marker (Section 3).

7.3. Potential toxicity

Assessments of PCNs TEQ in the studies cited below were based on those congeners for which relative potencies (REP) have been estimated (Section 5.4) and which were found in samples. Authors differed greatly on the number of DL-PCB congeners reported. Some studies base their DL-PCBs TEQ on only 3–4 coplanar PCBs (i.e., CBs 77, 81, 126 and 169), while others include some or all of mono-ortho PCBs 105, 114, 118, 123, 156, 157, 167 and 189. Thus, comparison of DL-PCBs TEQ values among studies is difficult.

Reported PCNs TEQ in seals are generally low, with exceptions. The PCNs TEQ in ringed seal blubber from the eastern Canadian Arctic averaged 0.016 pg g^{-1} in F and 0.0028 pg g^{-1} in M (Helm et al., 2002; Muir et al., 2004). The DL-PCBs TEQ were much higher, 0.59 pg g^{-1} in F and 0.72 pg g^{-1} in M (Helm et al., 2002). PCNs TEQ averaged 0.0002 pg g^{-1} in blubber of F ringed seals from the western Canadian Arctic (Muir et al., 2004). Corsolini et al. (2002) noted that PCNs TEQ in Weddell seal was <0.0003% of ΣTEQ due to PCDD/Fs, + DL-PCBs + PCNs. On the other hand, Wang et al. (2007) reported the PCNs TEQ values in blubber of harbour seal which averaged 1.1% of DL-PCBs TEQ, but with a wide range of <0.01% to 9%.

Higher PCNs TEQ was reported in blubber of beluga collected in 1994 from the eastern Canadian Arctic, 0.12 pg g^{-1} in F and 0.30 pg g^{-1} in M, contributed almost exclusively (97%) by CNs 66/67 (Helm et al., 2002). The DL-PCBs TEQ averaged 1.5 pg g^{-1} in F and 1.8 pg g^{-1} in M. The contribution of PCNs TEQ to ΣTEQ due to PCNs + DL-PCBs was 7–19%, with an average of 11%. It can be noted that hexa-CNs 66/67, which typically coelute on most gas chromatographic columns and differ by about a factor of two in REPs by H4IIE-EROD assay (Villeneuve et al., 2000) and by a factor of four in REPs by H4IIE-luciferase assay (Blankenship et al., 2000), were resolved on a derivatized β-cyclodextrin column (Helm et al., 1999). Ratios of CNs 66/67 in six beluga ranged from 0.8 to 1.4, with an average of 1.1. The REPs of CNs 66 and 67 were calculated for these congeners individually. PCNs TEQ in blubber of eastern Canadian Arctic beluga collected in 1999–2000 averaged 0.25 (M) and 0.14 (F) pg g^{-1}. DL-PCB TEQ was not measured in these animals (Muir et al., 2004).

The PCNs TEQ in blubber of long-finned pilot whale from the Faroe Islands was 8.0 pg g^{-1} for juveniles, 2.9 pg g^{-1} in F and 4.3 pg g^{-1} in M. The main contributors were CNs 66/67 and CN68. The PCDD/Fs TEQ (8–13 pg g^{-1}) was on par with those due to CN congeners. The ΣTEQ from PCDD/Fs + DL-PCBs (non-ortho only) in different pilot whales which were not analysed for PCNs was 46–71 pg g^{-1} (Vorkamp et al., 2004). If the latter values are assumed for animals in which PCNs were determined, the PCNs TEQ is ~6–15% of ΣTEQ, which is on the order of the PCNs TEQ contribution in Canadian Arctic beluga.

Average PCNs TEQ in glaucous gull from Bear Island were 0.03 pg g^{-1} in eggs and 0.04 pg g^{-1} in plasma (M and F), due mainly to CNs 66/67. The DL-PCBs TEQ in eggs and plasma (M,F) averaged 12.5 and 6.1–8.8, respectively (Verreault et al., 2005). Thus, PCNs TEQ contributed ~0.2–0.7% of ΣTEQ due to PCNs + DL-PCBs. The PCNs TEQ in the liver of other birds (black-legged kittiwake, thick-billed murre, black guillemot, northern fulmar and glaucous gull) from the Northwater Polynya in the Canadian Arctic, collected in 1998, ranged from 1.6 to 54 pg g^{-1} (Helm et al., 2003b). PCBs were not determined in these samples, but comparisons were made to ΣTEQ (PCDD/Fs + DL-PCBs) in eggs of the same bird species from the same location, collected in 1993 (Braune and Simon, 2003). With this cross-comparison, the PCNs TEQ in bird livers amounted to 0.22–0.65% of ΣTEQ in eggs. The PCNs TEQ in eggs of northern fulmar from Prince Leopold Island, Canadian Arctic, averaged 0.56 pg g^{-1}, and amounted to 0.21% of ΣTEQ (PCDD/Fs + DL-PCBs + PCNs) (Braune et al., unpublished). Muir et al. (2004) found similar PCNs TEQ in northern fulmar eggs from Prince Leopold Island and Cape Vera, 0.52–0.56 pg g^{-1}, and noted that the PCNs TEQ were only ~0.05% of the ΣTEQ due to PCDD/Fs and DL-PCBs. PCNs (largely CNs 66/67) accounted for <0.1% of ΣTEQ (PCDD/Fs + DL-PCBs + PCNs) in south polar skua (Corsolini et al., 2002).

Liver of polar bear from Alaska contained an average of 6.5 pg g^{-1} PCNs TEQ, estimated from the mean PCN content of 0.37 ng g^{-1} wet

weight, an average lipid content of 11.4%, and the authors' statement that PCNs (CNs 66/67) contributed 0.2% of ΣTEQ (PCDD/Fs + DL-PCBs + PCNs) (Corsolini et al., 2002). Polar bear fat samples from Greenland contained an average of 0.9 pg g^{-1} PCNs TEQ, due to a single congener, CN 68. No CNs 66/67 were reported in the bear fat (Vorkamp et al., 2004).

8. Conclusions and recommendations

8.1. Atmospheric transport and deposition

PCNs are atmospherically transported to arctic and subarctic regions and accumulate in invertebrates, fish, seabirds and marine mammals. Although one survey reported that PCNs were below detection in Antarctica, atmospheric transport is implied by their occurrence in biota.

The ΣPCNs in arctic air show high spatial variation. Concentrations in the European Arctic are up to an order of magnitude higher than those at Iceland and North American stations. Transport from source regions in the U.K. and central Europe is implicated. The ΣPCNs at high arctic stations (Alert, Dunai) are higher in winter–spring and lower in summer, while subarctic stations (Tagish, Nuuk) do not show a seasonal trend. Further measurements of PCNs in arctic air from other locations would verify spatial distributions. Examination of archived air sample extracts from the circumpolar network of arctic air monitoring stations could be a cost-effective way to do this, and to assess seasonal and temporal variations at other locations.

PCNs have legacy sources due to evaporation from products and residues, and from combustion sources which are ongoing. Both source types contribute to air concentrations in temperate regions, and evidence from archived U.K. soil samples suggests that the proportion of combustion-derived CNs is increasing. Combustion sources contribute to the ΣPCN concentrations in arctic air, as indicated by the presence of marker congeners. Several combustion-related congeners (e.g., CNs 54, 52/60 and 66/67) have higher REPs relative to other PCNs. More detailed analyses involving marker PCN congeners are needed to enable apportionment of source categories between legacy evaporative emissions and ongoing combustion emissions. The proportion of combustion-related CN congeners in biota should be carefully examined to determine if their spatial/temporal trends differ from the evaporative congeners.

PCNs are atmospherically deposited in snowfall, through adsorption of gaseous PCNs to the snow surface and scavenging of particle-bound compounds. Snows with low bulk density (= high SSA) appear to favour higher PCN concentrations in the meltwater. This implies that PCNs would be lost by revolatilization as the snowpack ages, as found for other POPs. The presence of PCNs on arctic aerosols suggests that particle dry deposition would also contribute to PCN loadings. The role of the snow pack in contaminant fate has only begun to be explored for POPs in the arctic, and the fate of PCNs would be better understood if included in such studies.

Although PCNs are found on suspended sediments in subarctic regions, there have been no reports of dissolved PCN measurements in arctic waters and so air–water gas exchange cannot be assessed at the present time. There has been no assessment of the depositional history and flux of PCNs to marine and lake sediments in polar regions. Analysis of archived sediment cores (or extracts) and examining water sample extracts collected for PCBs would be effective means of making these assessments in water bodies.

CN congeners contributed 13–75% of the ΣTEQ in arctic air due to both PCNs and DL-PCBs. The highest PCN contributions were in the cold season at Alert and Dunai, while lower contributions were in summer at these two stations and at Tagish throughout the year. No studies have been done in which PCNs, DL-PCBs and PCDD/Fs have all been measured in the same air samples. Comparisons to samples in which only PCDD/Fs, or PCDD/Fs + DL-PCBs, were measured suggest that PCDD/Fs dominate ΣTEQ in some cases and in others are rather similar to the TEQ contributions from the other two compound classes.

8.2. Levels and trends in biota

The data base for PCNs in biota of arctic and subarctic regions is small, and extremely small for the Arctic Ocean and seas around Antarctica. A large portion of the data are from specimens collected in the 1990s and the early years of this decade. Most invertebrate and fish measurements come from the northern Baltic Sea and northern Scandinavian lakes, rivers and fjords. There are no PCN data for invertebrates and fish in the Arctic Ocean and only one study in Antarctic biota.

Spatial trends are poorly defined because the same species have not been sampled at several locations throughout the Arctic. PCN measurements in seal are restricted to ringed seal in Canada, Greenland and Svalbard, harbour seal in Alaska and Weddell seal in Antarctica. Very few data are available for PCNs in cetaceans and polar bear.

Seabirds collected along the Norwegian coast, Bear Island, Svalbard, the Faroe Islands and the Canadian Arctic have been examined for PCNs. However, not all species have been collected in these four regions and spatial trend information is poor for a particular species.

No temporal trends data are available for PCNs in any polar species, except for arctic cod in the Vestertana Fjord in Norway, where no change in concentration of ΣPCNs was reported from 1987 to 1998.

Available data suggest that, among marine mammals, blubber concentrations of ΣPCNs follow the order: harbour seal ~ pilot whale > beluga > ringed seal ~ Weddell seal. The single survey of harbour seal organs show ΣPCNs in the order blubber > liver > kidney, while data for Weddell seals suggests liver > blubber (although from two different animals). The ΣPCNs in birds vary over two orders of magnitude with highest concentrations in glaucous gull eggs and plasma from Bear Island and livers of northern fulmar from the eastern Canadian Arctic. Lowest concentrations occurred in eggs of glaucous gull from Svalbard and black-backed gull from the Faroe Islands. The large variation is likely due to a combination of concentration differences among species as well as among tissues.

Few food webs have been examined for PCNs, except in the subarctic Bothnian Bay/Sea, lakes on Bear Island and Antarctica. The former two studies were for the lower food web only (zooplankton to arctic char, and amphipods–isopods–sculpin, respectively). PCNs generally show low biomagnification within these food webs. Discrimination in congener profiles occurs due to preferential metabolism. Certain PCN congeners are biomagnified in arctic seabirds. There is a complete lack of PCN data for food webs in the Arctic Ocean.

Dioxin-type toxicity due to PCNs appears low in most arctic, subarctic and antarctic biota. Contributions of PCNs TEQ to ΣTEQ (DL-PCBs + PCNs, or DL-PCBs + PCDD/Fs + PCNs) is less than a few tenths of a percent for ringed and Weddell seals, seabirds and polar bear. The PCNs TEQ accounts for a greater share of ΣTEQ in cetaceans, averaging 11% in beluga blubber and up to 6–15% in longfinned pilot whale blubber. The single study on harbour seals shows PCNs TEQ which are generally low, but up to 9% of ΣTEQ in some animals. No PCN data are available for other marine mammals in polar regions. It is beneficial to have PCN measurements obtained in conjuction with DL-PCBs to enable the ΣTEQ due to both compound classes to be evaluated.

To improve the availability of data in polar biota, circumpolar monitoring programs should consider incorporating PCNs when measuring POPs to enable spatial differences and temporal trends to be established. There is especially a need for PCN data in cetaceans, since available data suggest that PCNs may be important contributors to the ΣTEQ in these animals.

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