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Persistent organochlorine pollutants in ringed seals and polar bears collected from northern Alaska[☆]

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Abstract

Blubber samples from ringed seal (*Phoca hispida*; $n = 8$) and polar bear subcutaneous fat (*Ursus maritimus*; $n = 5$) were collected near Barrow, Alaska in 1996 as part of the Alaska Marine Mammal Tissue Archival Project (AMMTAP) and retained in the National Biomonitoring Specimen Bank at the National Institute of Standards and Technology in Gaithersburg, Maryland (USA). The samples were analyzed for a variety of persistent organochlorine pollutants (POPs) including polychlorinated biphenyls (PCBs), hexachlorocyclohexanes (HCHs), chlordane and metabolites, hexachlorobenzene (HCB) and DDTs and metabolites. The geometric mean, on a wet mass basis, of Σ PCBs (sum of 29 congeners and congener groups) were 732 ± 282 ng/g (1 S.D.) in seals and 3395 ± 1442 ng/g in polar bears. The geometric mean of Σ DDTs, Σ HCHs (α -, β - and γ - HCH) and HCB concentrations (wet mass basis) in seals and bears were 562 ± 261 ng/g vs. 74.8 ± 39 ng/g, 380 ± 213 ng/g vs. 515 ng/g, and 17.4 ± 10.1 ng/g vs. 183 ± 153 ng/g, respectively. The geometric mean sum of chlordane (Σ chlordane, sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane and heptachlor epoxide) and dieldrin concentrations in ringed seals and polar bears were 753 ± 617 ng/g vs. 720 ± 315 ng/g and 38.6 ± 22.8 ng/g vs. 130 ± 65 ng/g, respectively. Apparent bioaccumulation factors (polar bear/ringed seal POP concentrations) were lower in the animals sampled near Barrow, Alaska than in those from locations in the Canadian Arctic. This suggests that polar bears are also preying

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on marine mammals from lower trophic levels than the ringed seals with correspondingly lower organochlorine levels, such as bowhead whale carcasses. PCB congener patterns in the samples demonstrated the metabolism of certain PCB congeners in the polar bear relative to the ringed seal in agreement with previous studies. Regional comparisons of animals collected in Alaska and Arctic Canada are presented. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Ringed seal; Polar bear; Alaska; Arctic; Persistent organochlorine pollutants; Organochlorines; Bioaccumulation

1. Introduction

Since 1987, the Alaska Marine Mammal Tissue Archival Project (AMMTAP), with the cooperation of Alaskan Native organizations and subsistence hunters, has obtained tissues from over 300 individual marine mammals representing 11 species (Becker et al., 1993, 1997). These samples were collected from a variety of locations along the Alaska coast and comprise the majority of marine mammal samples obtained from the US region of the Arctic and sub-Arctic. The AMMTAP samples have been analyzed for a variety of constituents including trace elements (Becker et al., 1997) and persistent organochlorine pollutants (POPs; Schantz et al., 1993; Becker et al., 1997; Krahn et al., 1997; O'Hara et al., 1999).

Two species that are sampled by the AMMTAP are the ringed seal (*Phoca hispida*) and the polar bear (*Ursus maritimus*). Both species are circum-Arctic in distribution, with the polar bear generally having a much larger home range than the ringed seal (Stirling and Øritsland, 1995). These two species are higher trophic level predators that have been used for examining the spatial distribution of POPs in the Canadian Arctic (Norstrom et al., 1988; Weis and Muir, 1997; Muir et al., 1999, 2000). For instance, Weis and Muir (1997) recently compiled data on 221 ringed seal blubber samples collected primarily from Canadian Arctic and sub-Arctic locations detecting spatial trends in POPs concentrations. There was a general trend of lower POP concentrations in the northern and western Arctic compared with the more eastern sub-Arctic Hudson Bay. Weis and Muir (1997) also noted an increasing contribution of

the more volatile congeners relative to total PCBs (Σ PCB) with increases in latitude, consistent with the 'cold condensation' hypothesis (e.g. Wania and Mackay, 1996). Norstrom et al. (1998) also found spatial differences in POPs measured in 320 polar bear adipose samples collected from 16 Arctic and sub-Arctic locations. The POP concentrations in polar bears were variable with an overall-declining trend in chlordane and dieldrin concentrations from east to west. The Σ PCBs and 4,4'-DDE concentrations were variable with no obvious north to south or east to west gradient. In both of these studies, there were relatively few values from the western Arctic including Alaska; POPs were measured in nine polar bears by Norstrom et al. (1998) and no ringed seal data were reported from this area by Weis and Muir (1997).

Polar bears are the top-level carnivore in the Arctic marine food chain and are opportunistic feeders consuming bearded seal, beluga whales, walrus and bowhead whale carcasses, with a large reliance on ringed seals, especially pups and juvenile animals (Hammill and Smith, 1991; Stirling and Øritsland, 1995). Mature bears often remove the blubber from a ringed seal to capitalize on its high caloric and vitamin content (Stirling and Øritsland, 1995). Since POPs preferentially accumulate in the blubber relative to other tissues due to their stability and lipophilic nature, polar bears ingest a large portion of the seal's body burden of POPs. Polar bears, like many other terrestrial animals, have an excellent ability to metabolize many POPs by the induction of hepatic mixed function oxidases (Tanabe et al., 1988; Letcher et al., 1996; Bandiera et al., 1997). Consequently,

there are far fewer measurable POPs in polar bears than in ringed seals (Norstrom et al., 1988). Many POP metabolites, such as methyl sulfones derived from PCB congeners and chlordan metabolites, are produced during metabolism (Norstrom et al., 1988; Letcher et al., 1998).

There have been comparatively few POP measurements on marine mammals from the Alaskan Arctic relative to the Canadian Arctic. Therefore, the main goal of this investigation is to provide baseline characterization data on POP concentrations in ringed seals and from polar bears collected from similar geographic locations in the Alaskan Arctic. A secondary goal is to examine the selective bioaccumulation of PCB congeners in polar bears relative to ringed seals. The samples of ringed seals and polar bears analyzed in this study are useful for this purpose, since they were obtained from the same location (Barrow, Alaska) during the same year and season (Spring, 1996) thereby reducing spatial and temporal variability.

2. Materials and methods

2.1. Sample collection

Duplicate 150 g samples of ringed seal blubber and polar bear subcutaneous fat (rump area) were collected within 24 h post mortem following Alaskan Native subsistence hunts near Point Barrow (71°19' N, 156°50' W, Table 1; Becker et al., 1991). Only male polar bears were analyzed for POPs to simplify the metabolic comparisons with ringed seals. Samples were frozen and transferred in liquid N₂ dry shippers to the National Biomonitoring Specimen Bank (NBSB) at the National Institute of Standards and Technology (NIST) in Gaithersburg, Maryland (Becker et al., 1991). Ringed seal ages were estimated by the enumeration of front claw growth bands. Information regarding the samples used in this investigation is summarized in Table 1. A total of nine ringed seals blubber samples were analyzed representing samples from three females and six males from the Point Barrow area. The ringed seals ranged in

Table 1
Sample information

Animal number	Age (years)	Gender	Sampling date	Length (cm)	Weight (kg)	Girth (cm)	Blubber thickness (cm)
<i>Ringed seals</i>							
RGSL-047	7	M	8 July, 1996	117	34.5	86	3.5
RGSL-048 ^a	7	F	8 July, 1996	108	34	78	2
RGSL-049 ^a	7	M	8 July, 1996	88	18.1	62	1.9
RGSL-050 ^a	6	M	8 July, 1996	113	30.4	78	2.9
RGSL-051 ^a	7	M	10 July, 1996	109	35.4	87	2.8
RGSL-052	5	M	10 July, 1996	101	26.8	71	2.9
RGSL-053	6	M	10 July, 1996	107	34.5	76	1.9
RGSL-054 ^a	5	F	12 July, 1996	95	29.5	76.5	1.8
RGSL-055 ^a	5	F	12 July, 1996	88.5	24.9	77.5	2
<i>Polar bears</i>							
PLBR-001 ^a	Adult	M	30 March, 1996	168.9	ND	ND	NA
PLBR-002 ^a	Adult	M	26 March, 1996	176.4	ND	ND	NA
PLBR-009 ^a	Adult	M	15 October, 1996	168	ND	ND	NA
PLBR-011 ^a	Adult	M	31 December, 1996	183	ND	ND	NA
PLBR-017	Adult	M	30 December, 1996	165	ND	ND	NA

Abbreviations: ND, not determined; NA, not applicable.

^aAlso analyzed for PCBs using the methods detailed in Kucklick et al. (1996).

age from 5 to 7 years (Table 1). Five adult polar bears were collected 1996 also from the Point Barrow region. Additional information including animal size and blubber thickness in the case of ringed seals is presented in Table 1.

2.2. Sample preparation

Aliquots of the cryohomogenized samples (Table 1; Zeisler et al., 1983) packaged in Teflon jars were shipped to the NIST Charleston Laboratory in a liquid N₂ dry shipper and stored at –80°C until analysis. Between 0.5 and 0.6 g of sample was removed from the jar and mixed with 30 g of Na₂SO₄ (dried at 700°C for 24 h). The mixture was then transferred to a 33-ml pressurized fluid extractor cell (PFE; Dionex). Five calibration solutions were prepared by weighing portions of Standard Reference Materials (SRMs) 2261 (Chlorinated Pesticides in Hexane), 2262 (Chlorinated Biphenyl Congeners in 2,2,4-trimethylpentane), 2274 (Chlorinated Biphenyl Congeners in Isooctane II) and 2275 (Chlorinated Pesticides in Hexane II) into a weighed portion of isooctane. The solutions along with a mixed internal standard solution containing 4,4'-DDT-*d*₈, 4,4'-DDE-*d*₈, 4,4'-DDD-*d*₈, endosulfan I-*d*₄, PCB 103 and PCB 198 were gravimetrically added to the PFE cells packed with Na₂SO₄.

The samples were extracted with CH₂Cl₂ using the PFE (Schantz et al., 1997). Samples were initially purified by size exclusion chromatography (SEC; Schantz et al., 1993; Kucklick et al., 1997). The extracts (in hexane) was then fractionated using a semi-preparative aminopropylsilane column (Waters) into relatively lower and higher polarity fractions (F1 and F2, respectively; Schantz et al., 1993). Compounds contained in the F1 included PCBs heptachlor, 2,4'-DDE, 4,4'-DDE, 2,4'-DDT, hexachlorobenzene (HCB), oxychlordan and mirex. Analytes in F2 included 4,4'-DDT, *cis*- and *trans*-chlordan, *cis*- and *trans*-nonachlor, α- and β- and γ- and hexachlorocyclohexane (HCH), heptachlor epoxide, 2,4'- and 4,4'-DDD, and dieldrin.

2.3. Sample analysis

Organochlorine compounds (see above) were determined by gas chromatography (GC) with a micro-electron capture detection (ECD) (Hewlett Packard 6890). Compounds were separated using a 60 m DB-5 capillary column (J&W Scientific) with 0.25-mm interior diameter and a 0.25-μm film thickness. The injector and detector temperatures were 220°C and 325°C, respectively; the carrier and makeup gases were H₂ (constant velocity of 25 cm/s) and N₂ (60 ml/min), respectively. Samples were injected into the GC (2 μl, splitless injection) and the oven was programmed from 100°C initially (1 min hold) to 170°C at 18°C/min, then 1°C/min to 260°C, and finally ramped to 300°C at 1.5°C/min (107 min run time). The amount of each compound was calculated using internal standards and the slope and intercept of a five-point calibration curve. An aliquot of SRM 1945 'Organics in Whale Blubber' was run with each sample set as a control.

GC with negative-chemical ionization (NCI) mass spectrometry (MS) was used to quantify oxychlordan if this compound split between F1 and F2. The recombined samples plus their respective calibrants were injected twice via splitless injection onto a Hewlett Packard 6890 GC coupled to a Hewlett Packard 5973 mass spectrometer operated in the NCI mode (Kucklick et al., 1996). The samples were quantified from a four-point calibration curve generated from the original calibrants. Endosulfan-*d*₄ was used as the internal standard by monitoring *m/z* 410.

A number of PCB congeners were also determined in all the samples and the aliquot of SRM 1945 using GC-MS with electron-impact ionization (EI) to verify GC-ECD results. The congeners were: PCBs 99, 128, 101 + 90, 110, 138, 156, 201, 180, 206 and 209. The instrument was the same as above, but used a 30 m × 0.25 μm × 0.25 mm HP-5 column (Hewlett Packard). Samples were injected twice with the cool on-column injector. The instrument was operated in the selected ion monitoring mode using the following ions: 256, 258, 292, 294, 326, 328, 360, 362, 394, 396, 428 and 430. The oven temperature program

was as follows: 60°C for 1 min, 25°C/min to 150°C, 1.5°C/min to 250°C, then 5°C/min to 300°C with a 5-min hold. The source and transfer line temperatures were 250°C and 300°C, respectively. The carrier gas was He at 32 cm/s (constant velocity mode). Lipid was gravimetrically determined as in Kucklick et al. (1996).

2.4. Analysis of samples for metabolic comparison

The polar bear adipose tissue samples (PBLR-001, -002, -009 and -011) and ringed seal blubber samples (RGSL-048, -049, -050, -051, -054 and -055) were also analyzed for additional PCB congeners (Table 2). This was done to provide more detailed information on congener profiles to assess the metabolic transformation of PCBs in polar bears relative to ringed seals. The method used a mixture of Aroclors with known congener contributions and provided values for 74 individual PCB congeners or congener groups (Kucklick et al., 1996). Concentrations were determined using a GC-ECD equipped with a 60 m × 0.25 μm

× 0.25 mm DB-5 column. Individual congeners or congener groups were placed in the metabolic categories first proposed by Boon et al. (1987) and later modified by Kannan et al. (1995) (Table 2).

2.5. Quality control

The PCB concentrations reported in Table 3 are the sum of the 29 congeners determined using the SRM solutions and a combination of GC-ECD and GC-MS (EI). If the same congeners were measured using both methods, the values were averaged if similar otherwise, the lower value was used. For instance the PCB 99 was higher by GC-ECD, likely due to the coelution of MC6 (nonachlor III), hence the GC-MS value was used. POP concentrations were also determined in three aliquots of SRM 1945 that were analyzed in conjunction with the ringed seal and polar bear samples. SRM 1945 has certified or reference values for 29 PCB congeners and 17 organochlorine pesticides, and the identity of these compounds

Table 2
PCB congener metabolic groups as suggested by Kannan et al. (1995)

Metabolic group	Group definition	P-450 induction type	IUPAC congeners ^a
I	No vicinal hydrogens (highly chlorinated, non-coplanar, low Ah receptor affinity)	Refractory congeners	146, 132 + <u>153</u> , 178, 175, <u>187</u> + 182, 183, <u>202</u> + 171, 172, 180, 193, 191, 201, 196 + <u>203</u> , 208 + 195, 207, 194, 205, 206, 209
II	Only <i>meta-para</i> vicinal hydrogens	Metabolized by phenobarbital (PB)-type enzymes (2B subfamily)	52, 95, 101, 151, 123 + <u>149</u> , 185, 174
III	Only <i>ortho-meta</i> vicinal hydrogens	Metabolized by methyl cholanthrene (MC)-type enzymes (1A subfamily)	28, 47, 42 + <u>37</u> , 63, 66, 99, 119, 107, 118, 105, 130, <u>163</u> + 138, 158, 128, 177, <u>156</u> , <u>170</u> + 190
IV	Both <i>meta-para</i> and <i>ortho-meta</i> vicinal hydrogens (not highly chlorinated, coplanar, high Ah receptor affinity)	Metabolized by both PB and MC-type enzymes	<u>4</u> + 10, <u>7</u> + 9, 6, <u>8</u> + 5, 19, 12, 18, 15 + <u>17</u> , 24 + <u>27</u> , <u>16</u> + 32, 26, 31, 33, 51, 22, 45, 46, 49, 44, 64, 91, <u>56</u> + 60, 97, 87, <u>110</u> + 77, 82

^aDominant congener of co-eluting pair is underlined.

Table 3
Polychlorinated biphenyls in ringed seal blubber and polar bear adipose tissue samples

Lipid (%) → Compound	Females			Males					Males				
	RGSL-054 84.9	RGSL-055 85.3	RGSL-048 86.3	RGSL-051 85.9	RGSL-047 83.2	RGSL-050 87.0	RGSL-052 84.6	RGSL-053 88.3	PLBR-001 81	PLBR-002 83.7	PLBR-009 82.5	PLBR-011 83.9	PLBR-017 84.7
PCB 18	5.94	4.66	7.18	4.55	7.89	6.55	9.58	9.45	7.62	7.11	6.14	6.40	12.0
PCB 31	8.00	5.68	8.98	9.71	12.0	7.30	11.41	8.32	4.65	4.55	3.48	4.18	8.93
PCB 28	13.0	16.2	16.3	24.6	35.5	23.4	19.8	17.7	4.31	5.40	4.76	5.92	10.4
PCB 52	17.0	18.5	16.6	48.5	35.6	64.0	19.4	63.7	4.13	5.63	9.17	6.27	6.37
PCB 49	6.27	5.19	4.69	18.6	7.56	14.3	6.82	11.1	4.82	4.17	8.24	4.03	2.94
PCB 44	3.29	2.60	3.18	5.25	4.08	4.94	3.62	4.27	5.54	5.97	8.60	8.70	4.19
PCB 66	9.02	9.27	9.93	21.8	44.9	12.3	16.6	8.62	8.33	15.0	27.0	27.6	9.81
PCB 95	5.33	4.23	5.44	14.5	6.43	22.0	6.82	15.4	4.10	4.63	4.62	4.24	2.87
PCB 101+90	47.5	51.3	45.7	92	72	114	39.3	63.7	14.6	14.7	13.9	13.3	4.35
PCB 99	20.0	39	17.5	67	104	148	53	132	554	361	393	329	258
PCB 87	6.18	6.75	5.24	14.5	12.3	10.5	6.72	6.13	< 1.3	< 1.3	< 1.3	< 1.3	2.17
PCB 110	7.2	6.0	7.2	9.5	15.8	9.3	10.0	8.5	< 3.1	< 3.1	< 3.1	< 3.1	< 3.1
PCB 151	4.39	3.44	3.07	< 1.2	3.40	< 1.2	1.72	10.5	< 1.2	< 1.2	< 1.2	< 1.2	< 1.2
PCB 149	10.1	11.4	10.5	14.0	13.8	22.3	6.05	13.6	< 1.4	2.53	2.81	< 1.4	< 1.4
PCB 118	37.2	30.5	36.7	55.4	112	55.9	59.0	37.0	30.6	43.4	59.1	80.3	26.7
PCB 153	138	120	124	160	245	273	135	274	2548	1614	1617	1310	1113
PCB 105	12.1	15.6	9.99	22.0	33.3	20.6	15.8	13.3	14.3	17.5	21.2	26.0	10.3
PCB 138 + 158 + 160	78.7	78.5	63.0	116	148	197	78.4	198	463	390	295	337	210
PCB 187	21.7	18.6	16.2	31.7	24.9	45.2	9.23	45.7	9.4	17.8	14.9	16.5	6.24
PCB 183	23.2	18.4	20.3	26.7	25.7	33.2	10.7	37.0	54.1	52.9	40.4	47.8	29.1
PCB 128	< 2.4	3.14	< 2.4	27.3	< 2.4	19.9	< 2.4	22.6	15.2	17.6	9.7	11.4	5.40
PCB 156	3.40	3.20	3.40	4.10	6.10	6.50	9.30	< 1.9	99.4	51.3	49.0	42.8	37.1
PCB 201	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 2.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9
PCB 180	42.9	28.9	43.9	37.6	37.8	42.4	15.9	44.5	1129	539	401	288	310
PCB 170	14.2	9.27	13.8	10.3	13.4	15.4	5.13	14.1	610	276	248	136	171
PCB 195	1.87	1.14	1.85	1.06	1.09	1.75	< 0.87	1.61	6.27	5.29	2.96	3.20	1.62
PCB 194	3.52	1.83	4.07	1.92	2.65	3.01	1.14	2.80	309	98.5	58.5	33.9	40.9
PCB 206	1.54	0.89	2.02	0.72	0.78	1.23	< 0.9	0.89	78.0	24.7	10.7	8.45	7.55
PCB 209	< 1.1	< 1.1	< 1.1	< 1.1	< 1.1	< 1.1	< 1.1	< 1.1	31.8	12.3	2.99	3.14	1.63
ΣPCB	542	514	501	840	1026	1173	550	1065	6010	3590	3312	2754	2292

All values are in ng/g wet mass.

overlaps with those measured in this study. The majority of the values determined in SRM 1945 were within 12% of the certified or reference values. The mean percent lipid measured in SRM 1945 was $70.6 \pm 0.8\%$ (95% CI; $n = 6$) vs. the certified value of $74.3 \pm 0.45\%$.

3. Results and discussion

3.1. Concentrations of POPs in ringed seal blubber and polar bear subcutaneous fat

3.1.1. Ringed seals

Concentrations of PCB congeners and organochlorine pesticides from ringed seal blubber samples are given in Tables 3 and 4. The geometric mean is reported as POP concentrations in ringed seals and polar bears have been found to be log normally distributed (e.g. Muir et al., 2000). The POP concentrations are reported on a wet mass basis unless otherwise noted. Concentrations of Σ PCB congeners (sum of the 29 PCB congeners in Table 3) in the ringed seal blubber samples ranged from 501 ng/g in RGSL-48 to 1173 ng/g in RGSL-050. The mean \pm 1 S.D. of Σ PCBs was 732 ± 282 ng/g wet mass ($n = 8$, Table 3). The Σ PCB concentrations measured in Alaskan ringed seal blubber were within the broad range of those observed in ringed seals from south of Baffin Island reported by Muir et al. (1995); sum of 58 congeners); 250 ng/g to 4570 ng/g for females and 467 ng/g to 1799 ng/g for males. The concentrations of Σ PCBs (geometric mean of 58 congeners; age and gender effects statistically removed) reported by Weis and Muir (1997) from Sachs Harbour and Tuktoyaktuk (south-eastern Beaufort Sea) were the two closest locations to Barrow, Alaska, were 588 ± 117 ng/g and 561 ± 302 ng/g, respectively. Ringed seal blubber samples have also been analyzed from Holman Island in the western Canadian Arctic (Cameron et al., 1997; Addison and Zinck, 1986). For example Σ PCBs (sum of eight congeners) in male seals ($n = 4$) collected in 1989 were 241 ± 44 ng/g (Cameron et al., 1997). Schantz et al. (1996) measured Σ PCB (15 congeners and congener groups) concentrations in two male ringed seals

from Barrow, Alaska, of 686 ng/g for both animals. Levels of Σ PCBs in two male seals collected from Nome, Alaska, measured by the same investigators were 371 ng/g and 415 ng/g. In this study, PCB concentrations from Barrow were higher than those observed in the Bering Sea by Krahn et al. (1997) ($n = 8$), when summing the same 17 congeners; 420 ± 21 ng/g vs. 346 ± 17 ng/g for females and 710 ± 182 ng/g vs. 249 ± 75 ng/g for males, respectively. In the present study, male seals had higher Σ PCB concentrations than females, 901 ± 244 ng/g vs. 519 ± 21 ng/g likely due to the transfer of POPs to offspring during lactation and gestation (Tanabe et al., 1994). Congener 153 is one of the most abundant congeners in most Aroclor mixtures (Schulz et al., 1989) and extremely resistant to metabolism (Boon et al., 1987). PCB 153 was the dominant congener contributing from 19% to the Σ PCBs in RGSL-51 to 26% in RGSL-53 (Table 3).

Organochlorine pesticides measured in ringed seal blubber are presented in Table 4. Wet mass sum HCH (Σ HCH, sum of α - β - and γ -HCH) values for samples that included β -HCH measurements ranged from 146 ng/g wet mass (RSGS-52) to 561 ng/g wet mass (RGSL-53). Muir et al. (1995) also measured variable Σ HCH concentrations in ringed seal samples; 246 ± 231 ng/g in females and 274 ± 123 ng/g in males. Schantz et al. (1996) reported γ -HCH values in ringed seals from Barrow and Nome, Alaska, from 2.1 ± 0.01 ng/g to 633 ± 4 ng/g. For the samples in which all three HCHs were measured, α -HCH contributed the most to the Σ HCHs, ranging from 59% of Σ HCH in RGSL-047 to 68% in RGSL-053.

The geometric mean sum of chlordane (Σ chlordane; sum of *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, oxychlordane and heptachlor epoxide) was 251 ± 109 ng/g in female ringed seals and 885 ± 612 ng/g in males (Table 4). Oxychlordane was the dominant chlordane-related compound comprising between 18% of Σ chlordane in RGSL-051 to 53% in RGSL-052. In contrast, the Σ chlordane values reported by Weis and Muir (1997) from Sachs Harbour and Tuktoyaktuk were 347 ± 23 ng/g wet mass and 364 ± 85 ng/g wet mass, respectively. Concentra-

Table 4
Chlorinated pesticides in ringed seal blubber and polar bear adipose samples

Compound	Females			Males					PLBR-001	PLBR-002	PLBR-009	PLBR-011	PLBR-017
	RGSL-054	RGSL-055	RGSL-048	RGSL-051	RGSL-047	RGSL-050	RGSL-052	RGSL-053					
Hexachlorobenzene	17.5	14.8	40.5	15.4	8.24	17.7	16.9	8.07	451	128	79.0	94.1	163
α -HCH	104	123	116	265	285	165	87.5	380	63.5	191	97.4	169	160
β -HCH	– ^a	–	–	–	122	–	53.8	137	–	–	–	–	355
γ -HCH	7.36	7.17	4.52	30.9	27.9	26.6	5.04	44.4	10.2	8.96	8.90	6.39	< 1.2
Σ HCH	112	130	121	296	435	191	146	561	73.7	200	106	175	515
Heptachlor	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	< 2.1	3.57
<i>trans</i> -Chlordane	4.90	4.13	3.65	7.90	2.66	6.70	3.05	7.62	3.62	3.27	4.77	5.15	3.28
<i>cis</i> -Chlordane	3.37	4.48	2.52	26.0	3.74	36.2	2.08	32.9	4.87	3.71	3.69	< 1.0	< 1.0
<i>trans</i> -Nonachlor	60.1	106	43.1	607	132	791	76.7	1048	62.6	205	179	177	96.1
<i>cis</i> -Nonachlor	< 1.0	9.16	4.51	26.1	11.9	50.4	4.58	34.8	3.37	6.42	< 1.0	4.34	2.12
Heptachlor epoxide	33.4	43.6	28.6	104	73.6	170	34.2	187	61.30	113	140	164	114
Oxychlordane	116	221	105	275	429 ^b	233	201 ^b	608 ^b	196	351	513	495	983
Σ Chlordane	218	389	187	1046	653	1287	322	1918	332	682	840	846	1202
Dieldrin	22.4	26.7	24.9	37.7	52.1	83.9	12.1	49.0	253	99.9	106	109	126
2,4'-DDE	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4	< 1.4
4,4'-DDE	251	304	212	508	644	731	327	744	17.8	81.9	88.9	101	18.3
2,4'-DDD	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9	< 1.9
4,4'-DDD	6.39	7.73	7.76	9.04	8.81	14.2	4.93	10.4	< 1.4	4.42	4.84	< 1.4	< 1.4
2,4'-DDT	7.84	8.45	7.53	13.9	9.62	13.5	3.50	8.76	25.5	< 2.2	< 2.2	< 2.2	6.98
4,4'-DDT	37.5	41.6	39.4	51.0	138	100	84.6	141	14.4	14.3	24.4	23.5	9.44
Σ DDT	302	361	267	582	800	859	420	904	57.7	101	118	124	34.8

All values are in ng/g wet mass.

^aNot measured.

^bGC–NCI–MS values.

tions found in Bering Sea ringed seal blubber were 255 ± 40 ng/g for females and 157 ± 67 ng/g for males ($n = 8$; Krahn et al., 1997). *Cis*-chlordane was especially variable in the Barrow, Alaska, ringed seals, with concentrations ranging from 2.08 ng/g wet mass to 36.2 ng/g wet mass. Weis and Muir (1997) also measured variable levels of *cis*-chlordane, which they found to be related to sample location. The highest level of *cis*-chlordane measured in the sample set from that study was 49.8 ± 2.9 ng/g determined in seals from Sachs Harbour in the western Canadian Arctic.

The geometric mean sum of DDTs (Σ DDT; sum of 2,4'- and 4,4'- DDD, DDE and DDT) in the Barrow, Alaska, seals ranged from 307 ± 47.5 ng/g in females to 685 ± 205 ng/g in males (Table 4). Σ DDTs concentrations determined in the Barrow, Alaska, seals were lower than those observed in seals from south of Baffin Island by Muir et al. (1995); 1140 ± 683 ng/g in males and 1006 ± 1164 ng/g in females. Concentrations determined in the Barrow, Alaska ringed seals were, however, higher than those found by Krahn et al. (1997) for seals in the Bering Sea ($n = 8$); 194 ± 27

ng/g for females and 188 ± 63 ng/g for males. 4,4'-DDE was the DDT-group compound at the highest concentration ranging from 253 ± 46 ng/g in females to 566 ± 175 ng/g in males (Table 4). Schantz et al. (1996) reported 4,4'-DDE ranging from 27 ± 1 ng/g to 350 ± 12 ng/g in ringed seals from Barrow, AK and from 173 ± 4 ng/g to 240 ± 9 ng/g in seals from Nome, Alaska.

3.1.2. Polar bears

POP concentrations in adult (> 4 years) male polar bear subcutaneous fat are also given in Tables 3 and 4. The comparisons made to other studies in this paragraph are done so on a lipid mass basis. The geometric mean of Σ PCBs in polar bears collected near Barrow, Alaska was 4080 ± 1730 ng/g. Concentrations of Σ PCBs in these bears were higher than those found in a recent study examining the influence of geography on organochlorine distribution in polar bears. Norstrom et al. (1998) reported that Σ PCBs (sum of 16 congeners) in male bears ($n = 3$) collected from the Alaskan Arctic ranged from 2104 ng/g to 2958 ng/g (mean = 2380 ng/g). Male polar bears collected east of Barrow, Alaska, from the

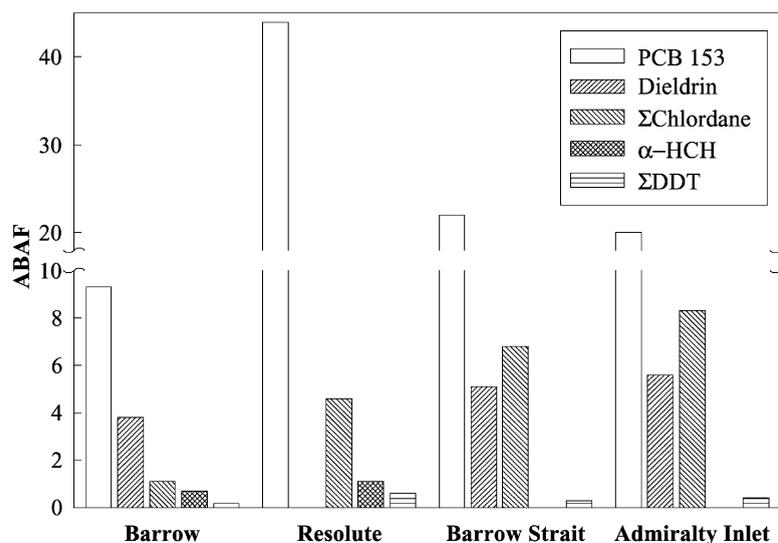


Fig. 1. ABAF vs. location. Data from Barrow Strait and from Admiralty Inlet are given in Muir et al. (1988); Σ DDT data from Resolute are from Letcher (1996); other POP data from Resolute bears are from Wiberg et al. (2000) derived from the same samples [the same samples also used in Letcher et al. (1998)]. Σ Chlordane is the sum of *cis*- and *trans*-chlordane, *trans*-nonachlor, oxychlordane and heptachlor epoxide.

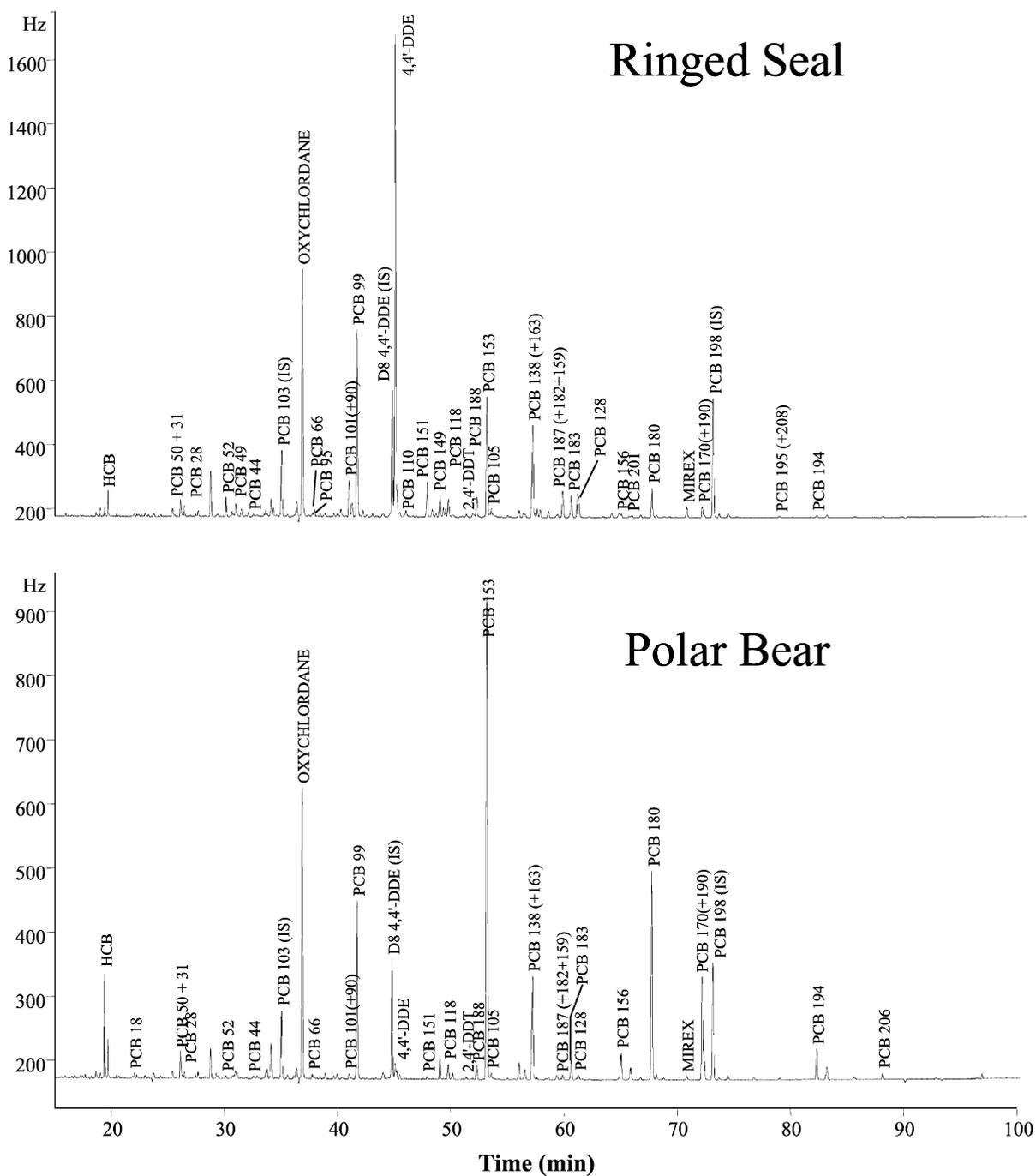


Fig. 2. Fraction 1 chromatograms of ringed seal and polar bear extracts obtained using GC-ECD equipped with a 60 m DB-5 column.

McClure Strait and adjacent Arctic Ocean ranged from 12 608 ng/g to 53 175 ng/g (mean = 22 391 ng/g). Age may affect POP concentrations in polar bears and discrete ages were not available for the animals. Norstrom et al. (1988) found that Σ chlordanes were significantly lower in adults than in sub adults (0–4 years), while other POP classes were not significantly different. POP concentrations in males tend to decrease slightly from age 5 to 6 then become constant with age (Bernhoft et al., 1997).

The number of measurable PCB congeners in polar bear subcutaneous fat was greatly reduced relative to ringed seal blubber (Figs. 1–3), and were dominated mainly by congeners 99, 153, 138, 180 and 170 + 190. These congeners comprised $89 \pm 1.0\%$ of the Σ PCB in polar bears. PCB 153 was the dominant congener, comprising $49 \pm 2.7\%$ of the Σ PCB. The reduction in the PCB congener pattern probably results from metabolism of PCBs by hepatic mixed function oxidases that attack less refractory congeners possessing adjacent hydrogen atoms mainly in the *meta-para* positions on the biphenyl ring (Letcher et al., 1998).

Wet mass organochlorine pesticide concentra-

tions in polar bears are shown in Table 4. The geometric mean Σ HCH concentrations were lower in polar bears and less variable, 134 ± 52 ng/g, than in ringed seals, 182 ± 121 ng/g. However, for the polar bear sample in which β -HCH was measured, PLBR-17, β -HCH was the dominant isomer unlike ringed seals (Table 4). The mean Σ chlordanes concentration in polar bears, 720 ± 315 ng/g, was similar to that of ringed seals, 552 ng/g (617 ng/g), but most of the chlordanes in the bears was in the form of the metabolite, oxychlordanes. Dieldrin was higher in polar bears, 130 ± 65 ng/g, than in ringed seals, 33.1 ± 23 ng/g. Σ DDTs were greatly reduced in polar bears relative to ringed seals; 78.4 ± 39 ng/g vs. 508 ± 261 ng/g in seals (Table 4). The Σ chlordanes, dieldrin and 4,4'-DDT concentrations were similar to those observed by Norstrom et al. (1998) from Alaskan bears.

3.2. Bioaccumulation of organochlorines in polar bears

Seals, especially ringed seals, are an important part of the polar bear diet (e.g. Hammill and

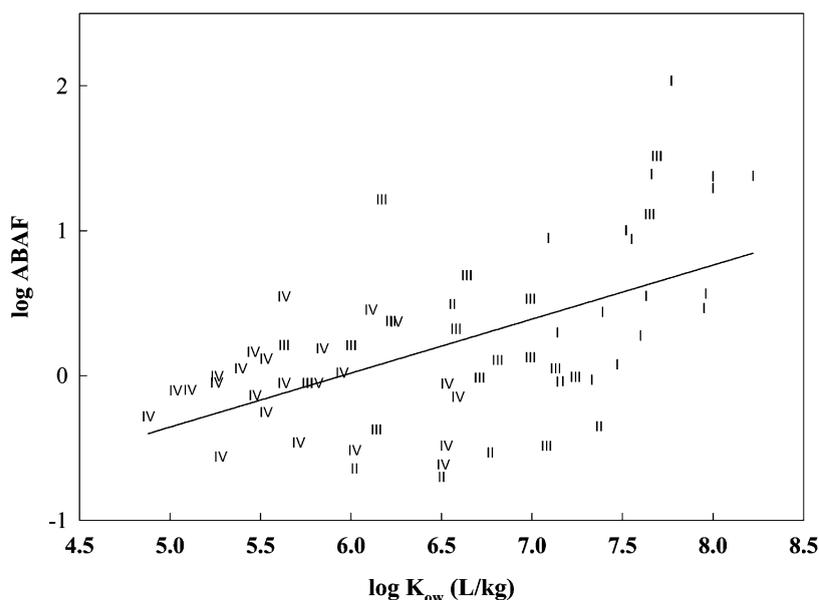


Fig. 3. Log ABAF for PCB congeners vs. log K_{ow} (l/kg). Log K_{ow} values are from Franz (1990). I, II, III and IV refer to the metabolic groups described by Kannan et al. (1995) and given in Table 2.

Smith, 1991). The POP concentrations determined in this study allowed for the examination of the bioaccumulation of POPs in polar bears relative to the ringed seal. The ratio of the lipid-based organochlorine concentrations in the polar bear divided by that in the ringed seal provides the 'apparent bioaccumulation factor' or ABAF. ABAFs for polar bears and ringed seals from several Arctic locations are compared in Fig. 1. An ABAF > 1 indicates that the organochlorines are biomagnified in the polar bear relative to the seal, whereas a value < 1 indicates that elimination dominates over accumulation. The data suggest that many POPs are biomagnified in the polar bear and that geographic differences in the ABAFs are probable. The ABAFs for PCB 153 and dieldrin were significantly greater than 1 (t -test, $P < 0.05$) in agreement with other work (Fig. 2). The ABAFs for α -HCH was not significantly different than 1, while Σ DDTs was significantly less than 1 (t -test, $P < 0.05$). α -HCHs is less hydrophobic, hence less bioaccumulative, than most other POPs examined in this work, with a log octanol/water partition coefficient (log K_{ow} ; l/kg) of approximately 4, while all other POPs measured have log $K_{ow} > 5$ (Suntio et al., 1988; Franz, 1990). Wiberg et al. (2000) showed that the enantiomeric ratio of α -HCH was different in ringed seals vs. polar bears suggesting either preferential accumulation of α -HCH + or selective removal of α -HCH -. Previous work (e.g. Letcher et al., 1998) has shown that DDTs are not accumulated above their concentrations in polar bear prey due to metabolism of 4,4'-DDE and other DDT compounds. The lower ABAFs in the Barrow Alaska, samples, for compounds that do substantially biomagnify in polar bear, support the observation (O'Hara, personal communication) that polar bears also prey on animals other than the ringed seals and the results shown in Fig. 1 suggest that this may vary by location. Polar bears are known to prey on other marine mammals utilizing different trophic pathways than the ringed seal. These include bearded seals, Pacific walrus and bowhead whales. Bearded seals and pacific walrus are top predators in benthic food webs whereas the bowhead whale is

principally a plankton feeder. Unlike ringed seals that prey mainly on fish, amphipods and euphausiids, bearded seals and walrus primarily feed on shrimp, crabs, mollusks, with only a seasonal reliance on fish, while the bowhead whale consumes euphausiids and copepods (Lowry et al., 1978, 1980). Polar bears have also been observed feeding on beluga whales, occupying a comparable trophic level to that of the ringed seal. Existing data indicate that other prey in the polar bear diet, especially bearded seals and walrus, have lower organochlorine levels in blubber than the ringed seal (Muir et al., 1995; Krahn et al., 1997). Bowhead whales sampled from Barrow, Alaska had Σ PCB concentrations in blubber that averaged only 350 ± 202 ng/g wet mass (O'Hara et al., 1999). Krahn et al. (1997) reported bearded seal POPs concentrations that are lower than ringed seal levels.

Polar bears have a high capacity to metabolize some organochlorines, including DDT-compounds and PCBs congeners, likely through the expression of CYP2B isozymes (Letcher et al., 1996, 1998). The result of PCB metabolism is readily apparent in the chromatograms of the ringed seals and polar bear PCB fraction (Fig. 2). Several PCB congeners are either missing or reduced in polar that are present in the ringed seal, particularly those congeners with adjacent hydrogens at the *meta-para* position (group II). Congeners shown in Fig. 2 with adjacent hydrogen atoms at the *meta-para* position include PCBs 52, 49, 95, 101, 110, 151, and 149. These congeners are generally present in the seal, but either greatly reduced or absent in the polar bear. Other congeners with no adjacent hydrogen atoms, such as PCBs 153, 138 and 180 are not metabolized by the polar bear and are strongly biomagnified (Fig. 2). Fig. 3 shows the log ABAF for PCB congeners vs. the log K_{ow} . The symbols on the plot represent the metabolic classifications in Table 3 based on Kannan et al. (1995), who classified chlorine positioning on the biphenyl ring in relationship to the induction of enzymes that may be responsible for biotransformation. The slope of the regression is significant ($P < 0.05$), however, it is only 0.37. Values close to unity are sometimes observed for

this BAF in foodwebs containing organisms that do not appreciably metabolize PCBs (e.g. Kucklick et al., 1996). Almost all the congeners detected in both the ringed seal and polar bear are from metabolic groups I, III and IV (Table 2).

To further explore the transformation of PCB congeners by polar bears, the congeners present in each metabolic group were normalized to the Σ PCBs. The bars in Fig. 4 contain PCB congeners grouped in the general structural-activity classifications described in Table 2. The percent of Σ PCBs among all of the groups was not significantly different between male and female ringed seals (one-way ANOVA, $P > 0.05$). The metabolic groups in both male and female seals, consequently, were pooled then compared with those in the polar bear. Seals had a significantly higher percentage of Groups IV and II congeners and significantly lower Group I congeners than the polar bears (one-way ANOVA $P < 0.05$). Group III congeners (MC or CYP1A type isozyme induction) were not significantly different between seals and polar bears. This suggests that CYP2B isozymes are correlated with the modification of PCB patterns from ringed seal to polar bear as suggested in other work (Letcher, 1996).

4. Conclusions

This investigation provides baseline data on ringed seals and polar bears, which are two key arctic species that have been suggested for arctic monitoring (Norstrom et al., 1988; Muir et al., 2000). The concentrations of POPs measured in ringed seals and polar bears from Barrow Alaska, are similar to those reported for these two species in the western Canadian Arctic. PCBs in polar bears were dominated by congeners that were not biotransformed by metabolism (e.g. congeners 99, 138 + 158 + 160, 180 and 170 + 190). The Σ HCH distributions in ringed seals were dominated by α -HCH, while β -HCH was the major isomer in polar bears. Concentrations of DDTs and chlordanes were reduced in polar bears compared to ringed seals suggesting that the bears have the ability to metabolize DDT and chlordane compounds as shown in previous studies (Letcher et al., 1996, Wiberg et al., 2000).

The biomagnification factors for POPs in ringed seal-polar bear relationships suggest the existence of regional differences. The ringed seal/polar bear ABAFs from the Barrow area were lower than those calculated from other regions in the

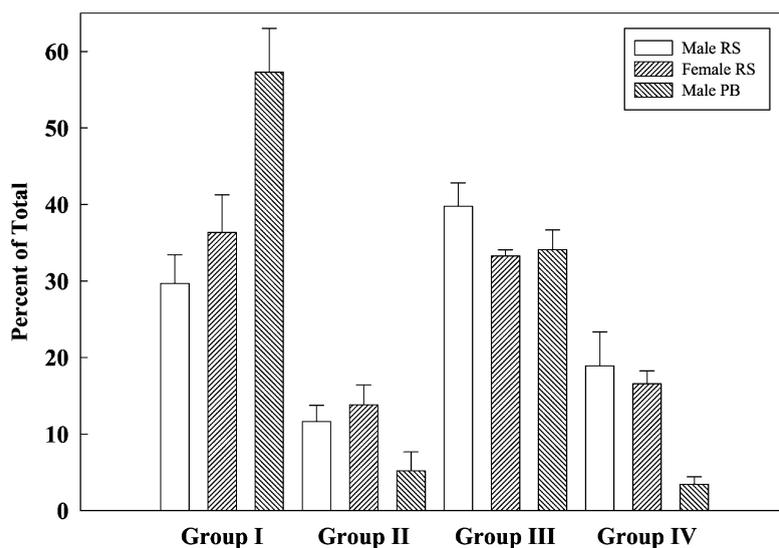


Fig. 4. PCB congener distributions in ringed seals (RS) and polar bears (PB) from Barrow, Alaska. Metabolic groups I–IV correspond to those in Table 2. Error bars are 1 S.D.

Canadian Arctic. This may reflect spatial and temporal variability in the bear diet across the North American Arctic. Recent population modeling suggest that approximately 80% of polar bears in the Barrow area originate from the Bering/Chukchi Sea stock rather than the Beaufort Sea stock (Amstrup, US Geological Survey/Biological Resources Division (USGS/BRD), personal communication). While ice seal population information is lacking, the Bering and Chukchi seas are believed to be more important bearded seal habitats than previously thought (York, US Geological Survey/Biological Resources Division, personal communication). Since polar bear stocks significantly overlap near Barrow, additional contaminant analysis of samples of polar bears and seals east and to the south of this area may provide additional insight into geographic differences in food habits, trophic pathways and population structure.

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