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Determination of levels of polychlorinated dibenzo-*p*-dioxins, dibenzofurans, biphenyls and pesticides in harp seals from the Greenland Sea

M. Oehme*[a], M. Schlabach[a], K. Hummert[b], B. Luckas[b], E.S. Nordøy[c]

[a] *Norwegian Institute for Air Research, P.O. Box 100, N-2007 Kjeller, Norway*
[b] *Institute of Food and Environment, Dornburgerstr. 25, D-07743 Jena, Germany*
[c] *Department of Arctic Biology, University of Tromsø, Breivika, N-9037 Tromsø, Norway*

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Abstract

The levels of polychlorinated dibenzo-*p*-dioxins (PCDD), dibenzofurans (PCDF), biphenyls (PCB) and compounds of the DDT group were determined in individual seal blubber and brain samples from ten male and one female harp seals caught in the Greenland Sea. No data from this region and from harp seals have been reported before. The age of the animals varied between 1 and 18 years. PCDD/PCDF concentrations in the blubber (4–10 pg/g TEQ wet weight (w.w.), Nordic model) were somewhat lower than in ringed seals from the Arctic. Non-ortho substituted PCB (CB 77, 126 and 169) showed TEQ levels in the blubber which were about 4–10-times higher. CB 77 dominated in most samples, but in a few cases CB 169 was the most abundant congener. p,p'-DDE (average 760 ng/g w.w.) and di-ortho substituted PCB concentrations (2560 ng/g w.w. expressed as ΣPCB) in the blubber were comparable to those found in ringed seals from the European and Canadian Arctic. Levels of PCDD/PCDF in brain correspond to the detection limit (about. 0.1–0.6 pg/g w.w.) for most congeners. With one exception, PCB amounts in brain (w.w.) were one to two orders of magnitude lower. A highly significant correlation was found between age, p,p-DDE and di-ortho PCB as well as between single PCDD and coplanar PCB congeners. No relationship was observed between levels of PCDD/PCDF, age and di-ortho-PCB.

Keywords: Arctic; Harp seals; Organochlorines; Dioxins

1. Introduction

In 1972 a paper had already been published about the presence of persistent organochlorines in ringed seals from the Canadian Arctic (Holden, 1972). Polychlorinated biphenyls (PCB) and compounds of the DDT-group (ΣDDT) could be detected. During the past decade a numerous studies confirmed that PCB and ΣDDT levels in Arctic seal blubber are around 1 μg/g wet weight and somewhat lower for hexachlorocyclohexanes (ΣHCH). The results were recently summarised in a review paper by Muir et al. (1992). Further-

* Corresponding author.

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more, it could be shown that long range atmospheric transport of these compounds to the Arctic is possible (Oehme, 1991, 1991a). Models developed by Wania and Mackay (1993, 1994) showed that this is probably the most important transfer route.

PCB and chlorinated pesticides are compound groups which have been synthesised and applied in large amounts. However, other polychlorinated substances are formed as undesired by-products in trace quantities by combustion processes (incineration, heating and traffic), by chemical synthesis of chlorinated compounds or by metal reclamation. Examples are the highly toxic polychlorinated dibenzo-*p*-dioxins (PCDD) and dibenzofurans (PCDF) (Safe and Hutzinger, 1990). PCDD and PCDF are now widely spread in the environment and could even be detected in marine mammals from the Canadian and European part of the Arctic (Oehme et al., 1988; Bignert et al., 1989; Oehme et al., 1990, Norstrom et al., 1990) However, only a very limited number of animals have been analysed so far (see Summary in Muir et al., 1992).

In most studies only a few PCDD/PCDF congeners could be detected due to levels close to the detection limits of the applied methods (Bignert et al., 1989; Norstrom et al., 1990). Furthermore, with one exception (Oehme et al., 1988) only results for PCDD/PCDF were reported and not for other organochlorines such as PCB and DDT. However, to study possible correlations between levels of PCDD/PCDF and other persistent polychlorinated compounds such data have to be available from the same samples.

PCB with no chlorine in the ortho-positions of the benzene rings show dioxin-like toxic properties (Ahlborg et al., 1992; Safe, 1990). Their toxicity is comparable with some of the more moderately toxic PCDD/PCDF congeners. The concentration of the most important coplanar (non-ortho substituted) congeners (IUPAC numbers CB-77, CB-126 and CB-169) is far below 1% in technical PCB mixtures (Duinker et al., 1988). The concentration of CB-77 in Chlophen A50 is, e.g., only 0.6% (Kihlström et al., 1992). However, levels which are one to two orders of magnitude higher than those of single PCDD/PCDF are found in marine mammals from the Arctic such as ringed seal (Daelemans et al., 1993). Concentrations of both PCDD/PCDF and coplanar PCB are often expressed as 2,3,7,8-tetrachloro dibenzo-*p*-dioxin toxicity equivalents (2,3,7,8-TEQ) according to the international (Safe, 1990) or Nordic model (Ahlborg et al., 1988), or using the toxicity equivalent factors (TEF) for coplanar PCB proposed by Safe (1990). Normally, the latter contributes more to the total 2,3,7,8-TEQ level in biota than PCDD/PCDF. Therefore, it is very important to determine coplanar PCB together with PCDD/PCDF in marine mammals.

The objectives of this study were to extend the very limited data base about levels of PCDD/PCDF in Arctic marine mammals and to quantify simultaneously other persistent organochlorines such as polychlorinated non-ortho substituted PCB with dioxin-like toxic properties, some mono-ortho and di-ortho substituted PCB which should be determined according to international recommendations (Duinker et al., 1988a) as well as polychlorinated pesticides. Concentrations of PCDD/PCDF in Arctic seals are only available for ringed seal blubber and one harp seal sample. Therefore, harp seals (*Phoca groenlandica*) were selected for this study to obtain information about levels in a different species. The animals were caught east of Greenland (about 72–75°N, 4–10°W) in the so-called West Ice region of the Greenland Sea. From this area levels of organochlorines in pinnipeds have not been published before.

The complex and time-consuming analysis of both PCDD/PCDF and other polychlorinated compounds restricted the number of samples which could be quantified. Therefore, mainly male harp seals with a large age distribution were selected. Males do not have the possibility to eliminate accumulated organochlorines by lactation and show in general higher level than females (Daelemans et al., 1993; Ronald et al., 1984; Miles et al., 1992). If correlations between levels of different organochlorines as well as age do exist, this preselection should increase the possibility to find them despite the limited number of samples. Furthermore, the obtained results

were compared with levels in seals reported from the Canadian and European Arctic.

2. Methods

2.1. Seal samples

The tissues examined in this study came from animals which were caught in the West Ice region (72–75°N and 5–10°W) of the Greenland Sea (see Fig. 1) by the department of Arctic biology in 1991. Two female and ten male individuals were sampled randomly and about 50 g of blubber from above the sternum and brain were collected, and wrapped immediately in aluminium foil and stored thereafter at −20°C. Ages were obtained from the cementum growth layers in canine teeth and was carried out at the Marine Research Institute in Bergen, Norway. Details about origin, age and sex are given in Table 1.

2.2. Determination of PCDD / PCDF and coplanar PCB

For quantification of PCDD/PCDF and coplanar PCB the same method was applied as recently described in detail for the determination of sub-ppt levels in Arctic sediments (Oehme et al., 1994) and in Antarctic seals (Oehme et al., 1994a). Therefore, only a brief description is given. Five g of seal blubber was homogenised with 100 g of water-free sodium sulphate in a household mixer (Braun Multiquick ZK3) with an insert made from polypropylene. The sample was dehydrated by spreading it as a thin layer on an aluminium foil which was kept in a clean fume hood overnight. The salt-like mixture was homogenised once more, and a mixture of ^{13}C-isotope-labelled compounds was added (totally 4–16 pg for each congener). The sample was transferred to the top of the first column of the multicolumn clean-up

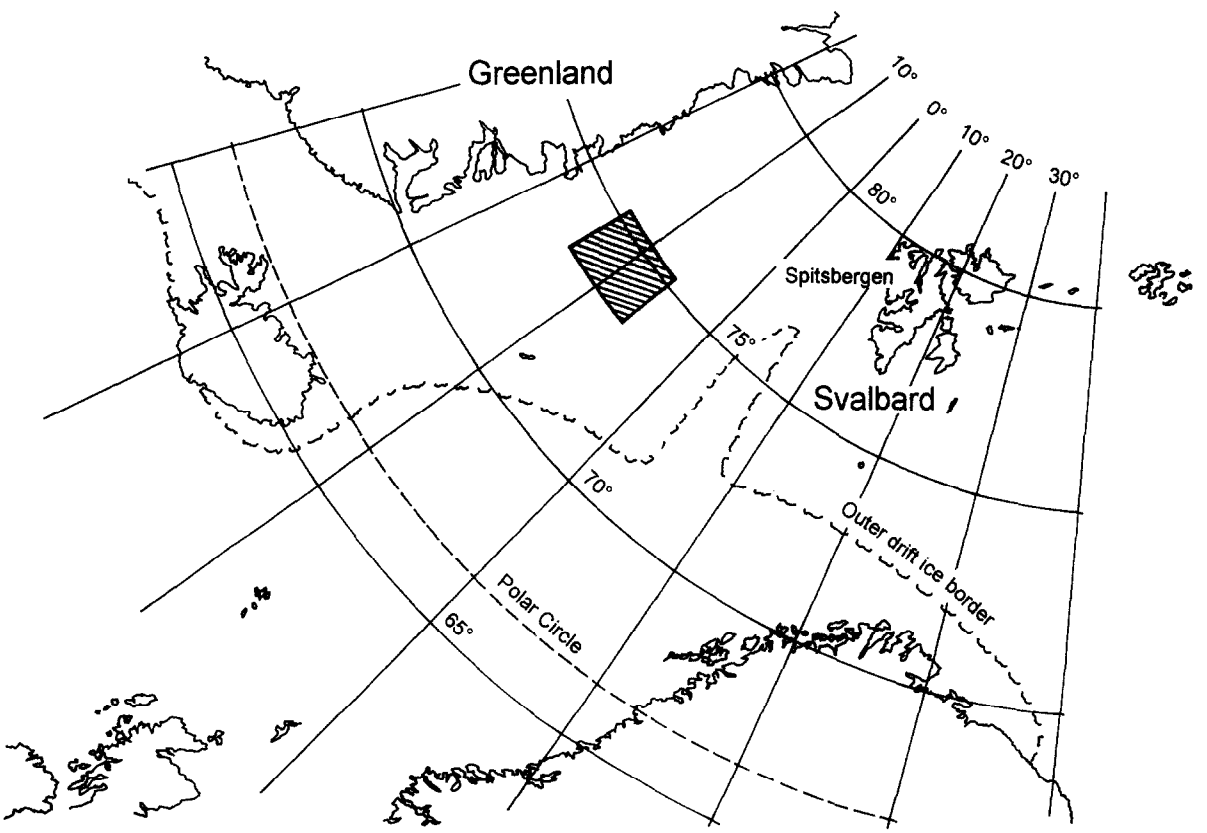

Fig. 1. Catching area of the harp seals.

Table 1
Catching sites and date, age and sex of the harp seals analysed for polychlorinated compounds

Sample no.	Catching area	Date	Age	Sex
1	75°10′ N 10°06′ W	20.04	1	F
2	72°43′ N 09°16′ W	22.04	6	M
3	72°49′ N 09°01′ W	22.04	3	M
4	72°52′ N 09°10′ W	23.04	16	M
5	74°00′ N 13°30′ W	01.05	18	M
6	74°00′ N 13°30′ W	01.05	6	M
7	75°00′ N 09°29′ W	04.05	*	M
8	75°33′ N 04°59′ W	06.05	13	M
9	75°33′ N 04°59′ W	06.05	10	M
10	75°33′ N 04°59′ W	06.05	4	M
11	75°33′ N 04°59′ W	06.05	10	M

*Not determinable (adult)

system. Final clean-up was carried out using a combination of two Pasteur pipettes, one filled with sulphuric acid-coated silica and one with aluminium oxide. The PCDD/PCDF and coplanar PCB congeners were separated on a fused silica capillary of 30 m length, 0.25 mm i.d. coated with 0.1 μm of a polymethyl-polyphenyl siloxane (e.g., DB5, J and W Scientific). Quantification was carried out by high resolution mass spectrometry at a resolution of 10000.

Quality assurance measures were included according to international standards (see, e.g., Ambidge et al., 1990; Yrjänheikki et al., 1991). It was required that the signal-to-noise ratio of a detected PCDD/PCDF or coplanar PCB congener in the mass chromatogram had to be > 3:1. Furthermore, the concentration detected should exceed possible blank levels by at least a factor of 10. Blank values were controlled before and after each series of ten samples. To control possible losses during clean-up and analysis, ^{13}C-isotope labelled 2,3,7,8-chlorine substituted PCDD/PCDF congeners were added to each sample. The recovery rates of all congeners in all samples were within the limits of 50–115% required by international protocols for PCDD/PCDF analysis (Yrjänheikki, 1991).

The same performance criteria were also applied to the quantification of coplanar PCB. The recovery rates for 3,3′,4,4′-TeCB were in some samples lower than 50%. This was possibly caused by losses during the reconcentration step on the active carbon column. However, the quantification by the isotope-dilution method based on the addition of ^{13}C-isotope-labelled congeners before sample clean-up allows to compensate for losses down to 20–30% (unpublished results).

2.3. PCB and other polychlorinated compounds

Ten g of blubber or brain was digested using a mixture of 10 ml perchloric (70%) and 10 ml acetic acid at 80°C for 4 h. Afterwards the compounds listed in Tables 4 and 5 were extracted with 80 ml hexane followed by an additional clean-up of the organic phase with 50 ml of concentrated sulphuric acid. After evaporation of the solvent close to dryness, 2 ml iso-octane was added and the final extract was shaken out once more with 1 ml of sulphuric acid. Quantification was carried out by high resolution gas chromatography (HRGC) combined with electron capture detection on a Hewlett-Packard 5890 gas chromatograph using two capillary columns: 50 m × 0.32 mm i.d. coated with 0.17 μm HP5 or 0.25 μm OV 1701 (Macherey and Nagel). The separation conditions were as follows: injector temperature 220°C, splitless injection of 1 μl; detector temperature 300°C; temperature program, 180°C for 4 min, then 6°C/min to 210°C, 4 min, 4°C/min to 270°C, isothermal 25 min.

Quantification was carried out using response factors from certified individual standards. Recently, the applied method was successfully controlled by an interlaboratory study where 14 laboratories participated (Rimkus et al., 1993). The results deviated 10% or less from the obtained mean values.

3. Results and discussion

To our knowledge this is the first study where levels of PCDD/PCDF, PCB including coplanar PCB and chlorinated pesticides were determined simultaneously in individual seal samples from the Arctic. Very little is known about levels of polychlorinated compounds in Arctic harp seals. Only data for PCB and DDT levels are available from one study carried out by Ronald et al.

(1984). Animals from Northwest and West Greenland as well as the Northern Baffin Bay region were analysed. Furthermore, the only reported PCDD/PCDF levels were from one pooled sample of three juvenile female harp seals from an unspecifically described area (Bignert et al., 1989). Coplanar PCB have not been determined in tissues from harp seals. Since only very few data about concentrations of persistent organochlorines in harp seals were available, the obtained results had also to be compared with levels found in Arctic ringed seals. This is the only seal species from the Arctic for which some data are available for all compounds classes determined in this study.

3.1. PCDD and PCDF

Detection limits for blubber in the order of 0.05–0.1 pg/g at a signal-to-noise ratio of 3:1 allowed to detect all 2,3,7,8-chlorine substituted PCDD/PCDF congeners except 1,2,3,7,8,9-HxCDD and 1,2,3,4,7,8,9,-HpCDF in the blubber samples (see Table 2). Most of the blubber data fulfilled the criterion that blank values should be equal to the detection limits or be at least 10-times

Table 2
PCDD and PCDF concentrations (pg/g wet weight) in seal blubber (*Phoca groenlandica*): recovery rates of added ^{13}C-marked 2,3,7,8-chlorine-substituted isomers were within 83–103%; all values are corrected for recoveries

Sample no.	1	2	3	4	5	6	7	8	9	10
Sampling site	75°10′N 10°06′W	72°43′N 09°16′W	72°49′N 09°01′W	75°52′N 09°10′W	74°00′N 13°30′W	74°00′N 13°30′W	75°00′N 09°29′W	75°33′N 04°59′	75°33′N 04°59′W	75°33′N 04°59′W
Sex	female	male	male	male	male	male	male	male	male	male
Age	1	6	3	16	18	6	(10)	13	10	4
2,3,7,8-TCDD	0.75	0.78	0.71	0.73	0.69	0.55	1.26	1.20	0.52	0.90
1,2,3,7,8-PeCDD	1.76	3.25	2.14	2.00	2.93	1.75	5.20	4.01	1.67	2.38
1,2,3,4,7,8-HxCDD	0.31	0.78	0.52	0.53	0.55	0.57	1.24	0.76	0.40	0.42
1,2,3,6,7,8-HxCDD	1.42	3.73	1.84	2.13	3.11	2.30	9.34	4.29	2.06	2.20
1,2,3,7,8,9-HxCDD	0.35	0.45	0.46	0.38	0.51	0.43	0.81	0.47	0.30	0.34
1,2,3,4,6,7,8-HpCDD	0.44	0.64	0.66	0.96	0.93	0.79	1.02	0.94	0.66	0.91
OCDD	2.25	2.32	2.50	6.59	5.83	1.27	3.38	3.10	3.49	3.57
2,3,7,8-TCDF	5.56	4.48	10.4	3.66	4.82	5.38	4.03	8.72	3.65	8.04
1,2,3,7,8-PeCDF[a]	0.66	0.65	0.85	0.55	0.70	0.59	0.79	1.07	0.41	0.73
2,3,4,7,8-PeCDF	2.86	4.65	5.11	2.91	3.37	2.68	6.73	6.02	2.11	4.46
1,2,3,4,7,8-HxCDF[a]	0.74	1.03	1.38	1.47	0.83	1.38	2.87	0.87	0.56	1.98
1,2,3,6,7,8-HxCDF	0.52	1.07	0.85	0.83	0.68	0.73	1.79	0.77	0.50	0.92
1,2,3,7,8,9-HxCDF	< 0.16	0.10	0.07	< 0.15	0.09	0.10	< 0.12	0.03	< 0.11	< 0.25
2,3,4,6,7,8-HxCDF	0.34	0.73	1.11	1.02	0.46	1.04	2.53	0.57	0.35	1.11
1,2,3,4,6,7,8-HpCDF	0.41	0.35	0.53	0.59	0.47	0.79	0.92	0.41	0.29	0.68
1,2,3,4,7,8,9-HpCDF	< 0.17	< 0.16	0.07	< 0.13	0.13	0.13	< 0.06	< 0.04	< 0.03	< 0.22
OCDF	0.56	0.48	0.50	0.72	0.75	1.27	0.21	0.37	0.39	0.91
2,3,7,8-TE (PCDD/F)[b]	4.0	5.99	6.02	4.23	4.97	3.98	9.53	7.89	3.22	5.88
3,3′4,4′TeCB	158	91	211	141	111	184	152	410	129	199
3,3′,4,4′,5-PeCB	131	205	167	149	213	130	472	326	156	198
3,3′,4,4′,5,5′-HxCB	24.4	128	99	256	30.4	213	942	57	38.3	115
2,3,7,8 TE (PCB)[c]	15.9	27.8	23.8	29.1	23.9	25.5	95.9	39.6	18.8	27.5

<, individual detection limit at a signal-to-noise ratio of 3:1.
[a]Not separated from 1,2,3,4,8-PeCDF or 1,2,3,4,7,9-HxCDF, respectively.
[b]Nordic model (19).
[c]see Safe (1990).

the measured concentrations. Blank levels for OCDD calculated with the same sample amount were in the order of 0.5 pg/g and, therefore, contribute significantly to the measured concentrations in blubber and brain. As can be seen from Table 3, only 2,3,7,8-TCDF was found in measurable quantities in brain. Furthermore, detection limits for the higher chlorinated congeners were somewhat higher due to some matrix residues.

Ringed seals and harp seals feed on both crustaceans and fish from the upper water layers. Compared to the average levels found in Arctic ringed seal blubber (Oehme et al., 1988; Bignert et al., 1989; Oehme et al., 1990, Norstrom et al., 1990) the PCDD/PCDF concentrations in harp seal blubber, expressed as 2,3,7,8-TEQ, are about a factor of 2–3 lower. In the only harp seal blubber sample (three juvenile females, < 1 year) analysed before (Bignert et al., 1989), most PCDD/PCDF congener levels were below the detection limit of 1–2 pg/g. However, the concentrations for the four detected congeners 2,3,7,8-TCDF, 1,2,3,7,8-PeCDD/PeCDF and 2,3,4,7,8-PeCDF were comparable with the levels found in this study. Bignert et al. (1989) found

Table 3
PCDD and PCDF concentrations (pg/g weight weight) in seal brain (*Phoca groenlandica*): recovery rates of added ^{13}C-marked 2,3,7,8-chlorine-substituted isomers were within 57–78%; all values are corrected for recoveries

Sample no.	1	2	3	4	5	6	7	8	9	10
Sampling site	75°10′N 10°06′W	72°43′N 09°16′W	72°49′N 09°01′W	72°52′N 09°10′W	74°00′N 13°30′W	74°00′N 13°30′W	75°00′N 09°29′W	75°33′N 04°59′W	75°33′N 04°59′W	75°33′N 04°59′W
Sex	female	male	male	male	male	male	male	male	male	male
Age	1	6	3	16	18	6	adult	13	10	4
2,3,7,8-TCDD	< 0.17	< 0.25	< 0.25	< 0.4	< 0.26	< 0.28	< 0.20	< 0.14	< 0.15	< 0.18
1,2,3,7,8-PeCDD	< 0.26	< 0.18	< 0.22	< 0.19	< 0.16	< 0.18	< 0.16	< 0.21	< 0.16	< 0.31
1,2,3,4,7,8-HxCDD	< 0.20	< 0.40	< 0.32	< 0.22	< 0.31	< 0.27	< 0.30	< 0.33	< 0.19	< 0.19
1,2,3,6,7,8-HxCDD	< 0.15	< 0.27	< 0.22	< 0.16	< 0.22	< 0.19	< 0.21	< 0.23	< 0.13	< 0.13
1,2,3,7,8,9-HxCDD	< 0.16	< 0.31	< 0.25	< 0.18	< 0.25	< 0.21	< 0.24	< 0.26	< 0.15	< 0.15
1,2,3,4,6,7,8-HpCDD	< 0.21	< 0.20	< 0.22	< 0.22	< 0.21	< 0.25	< 0.24	< 0.20	< 0.15	< 0.15
OCDD	0.96	0.77	0.96	1.0	0.85	0.99	1.7	1.97	1.04	0.66
2,3,7,8-TCDF	0.26	0.28	0.35	0.32	0.29	0.27	0.23	0.40	0.35	< 0.29
1,2,3,7,8-PeCDF[a]	< 0.85	< 0.29	0.19	< 0.51	< 0.14	< 0.32	< 0.30	< 0.56	< 0.31	0.23
2,3,4,7,8-PeCDF	< 0.88	< 0.30	< 0.10	< 0.53	< 0.14	< 0.33	< 0.10	< 0.57	< 0.28	< 0.24
1,2,3,4,7,8-HxCDF[a]	< 0.15	< 0.10	< 0.14	< 0.11	< 0.76	< 0.43	< 0.15	< 0.21	< 0.31	< 0.56
1,2,3,6,7,8-HxCDF	< 0.15	< 0.11	< 0.18	< 0.15	< 0.57	< 0.32	< 0.18	< 0.22	< 0.25	< 0.34
1,2,3,7,8,9-HxCDF	< 0.11	< 0.11	< 0.22	< 0.14	< 0.77	< 0.26	< 0.12	< 0.15	< 0.17	< 0.35
2,3,4,6,7,8-HxCDF	< 0.10	< 0.10	< 0.19	< 0.12	< 0.80	< 0.28	< 0.11	< 0.17	< 0.28	< 0.27
1,2,3,4,6,7,8-HpCDF	< 0.43	< 0.85	< 0.50	< 0.31	< 0.58	< 0.22	< 0.56	< 0.35	< 0.25	< 0.63
1,2,3,4,7,8,9-HpCDF	< 0.83	< 0.16	< 0.90	< 0.62	< 0.60	< 0.35	< 0.46	< 0.44	< 0.21	< 0.42
OCDF	< 0.5	< 0.5	< 0.35	< 0.26	< 0.5	< 0.3	< 0.21	< 0.45	< 0.39	< 0.30
2,3,7,8-TE (PCDD/F)[b]	<	<	<	<	<	<	<	<	<	<
3,3′4,4′TeCB	12.7	11.5	11.5	11.6	11.3	11.3	15.6	13.5	13.3	15.0
3,3′,4,4′,5-PeCB	1.34	3.53	2.23	2.8	1.93	2.07	5.38	3.47	3.4	2.25
3,3′,4,4′,5,5′HxCB	0.29	0.76	< 0.2	1.53	< 0.23	0.97	4.93	0.35	0.33	0.95
2,3,7,8-TE (PCB)[c]	0.28	0.51	0.34	0.47	0.31	0.37	0.94	0.49	0.48	0.42

<, individual detection limit at a signal-to-noise ratio of 3:1.
[a] Not separated from 1,2,3,4,8-PeCDF or 1,2,3,4,7,9-HxCDF, respectively.
[b] Nordic model (19).
[c] see Safe (1990).

higher levels in females than males. In this study all harp seal blubber samples were from males with the exception of one sample. Furthermore, Bignert et al. (1989), Norstrom et al. (1990) and the present study could not find any correlation between age and PCDD/PCDF levels in seals (see below). Together with the missing correlation between age and concentration, geographical or interspecies differences might be the most likely reasons for the lower PCDD/PCDF levels found in the Greenland Sea region.

Very little is known about the mobility of harp seals living in the West Ice region of the Greenland Sea. The significantly higher levels in two of the animals may indicate that they have moved into the Northeast Atlantic from other areas with higher levels of contaminants such as the White Sea. Studies of the genetic variation between the harp seal population in the Greenland Sea and White Sea (Meisfjord et al., 1991) have only revealed very small differences which suggest some exchange of individuals between these two populations.

A comparison of the levels found in brain tissue with those present in seal blubber is difficult due to the following reasons. First, concentrations in brain were equal or close to the detection limits and second the fat composition in both tissues is different. Brain contains up to 30–40% phospholipids, while seal blubber consists mainly of triglycerides. In addition, as shown by a recent intercalibration, the extractable amount of lipids is dependent on the applied method (Yrjänheikki, 1991). The fat content of the blubber samples determined by extraction with dichloromethane was very high and quite uniform (91–95%) while the same method applied for brain tissue gave very inconsistent results. On a wet weight basis 2,3,7,8-TCDF concentrations were about 10–20-times lower in brain than in blubber. This is comparable with the ratio found for 3,3′,4,4′-TeCB, but less than for all other compound except HCH.

Fig. 2 compares the 2,3,7,8-chlorine substituted congener concentrations relative to 1,2,3,7,8-PeCDD in Arctic ringed seal (*Phoca hispida*, Oehme et al., 1988), harp seal and Antarctic fur seal (Oehme et al., 1994a). 1,2,3,7,8-PeCDD was chosen as the basis since this congener was present in every sample at concentrations at least 20 times above the detection limit and since its gas chromatographic signal is never disturbed by interfering isomers. By selecting this congener the influence of the measuring precision on the relative comparison should be minimal. The average and standard deviation of the relative concentrations for individual animals is given.

Beside OCDD (not shown in Fig. 2), 2,3,7,8-TCDF is the most dominant congener in both Arctic species. TCDF and PeCDF were also the most abundant congener groups in sediments from the Barents Sea (Oehme et al., 1994). Despite significant differences in the age and sex distribution between the analysed group of Arctic ringed (about equal numbers of males and females, 3–10 years old, Oehme et al., 1990) and harp seals (mainly male seals, 3–18 years old), the congener profiles were quite similar. As shown in Fig. 2 the standard deviations of the concentrations relative to 1,2,3,7,8-PeCDD are in most cases not more than $\pm 30\%$ for both Arctic seal groups. This is comparable with the precision of the analytical method which is in the order of $\pm 15\%$.

The 2,3,7,8-chlorine-substituted congener profile in Antarctic female fur seals shows a very homogenous concentration distribution of the tetra- to hexachloro-CDD/CDF relative to 1,2,3,7,8-PeCDD. The standard deviation of the average of found absolute concentrations is not larger than about 25–30% for the most abundant tetra- and pentachloro-PCDD/PCDF congeners (Oehme et al., 1994a), although the age of the seals varied between 3 and 11 years. Due to lower signal-to-noise ratios the standard deviation is higher for HxCDF. The most abundant congeners are 1,2,3,4,6,7,8-HpCDD, OCDD and OCDF. 2,3,7,8-TCDF is not a significant congener. Possible explanations for the deviations compared to Arctic seals can be different food habits or differences in the source pattern between the Northern and Southern Hemisphere. Interspecies variations might also be a reason though the relative PCDD/PCDF congener profiles between harp and ringed seals indicate not very large differences.

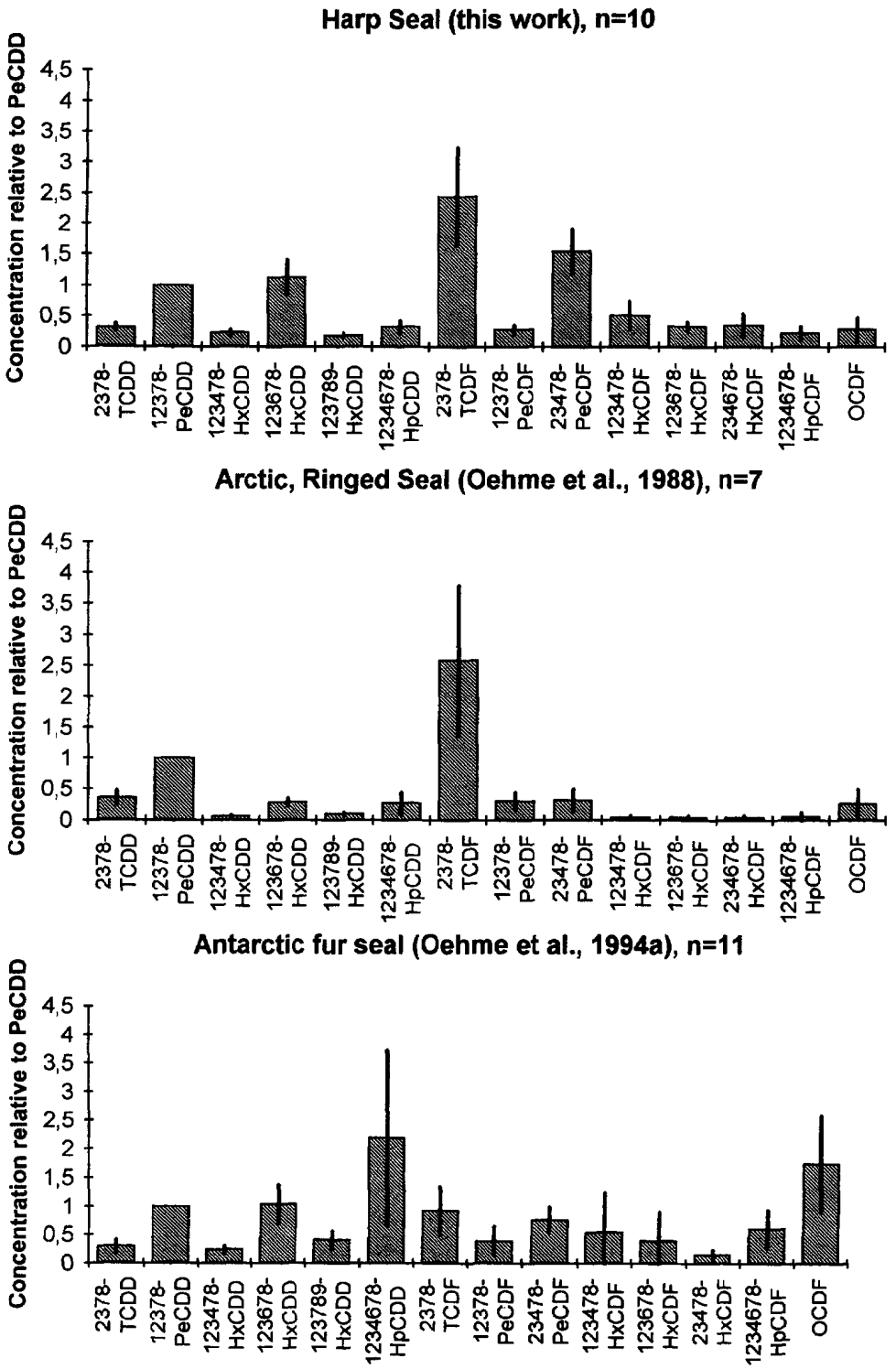

Fig. 2. Average concentration profiles of 2,3,7,8-chlorine-substituted PCDD/ PCDF congeners relative to 1,2,3,7,8-pentachloro dibenzo-*p*-dioxin in seal blubber from Arctic ringed seals, Arctic harp seals and Antarctic fur seals. The standard deviation of the relative concentrations of individual animals is given. The number of samples is indicated. OCDD is not included.

3.2. Non-ortho substituted PCB

Recently the 2,3,7,8-TEQ levels calculated from coplanar PCB and mono-ortho PCB were compared with the 2,3,7,8-TEQ from PCDD/PCDF concentrations in ringed seal blubber (Daelemans et al., 1993). It was concluded that the PCB 2,3,7,8-TEQ levels were about one order of magnitude higher than for PCDD/PCDF and, therefore, pose a greater toxic threat. However, levels of PCDD/PCDF and coplanar PCB in samples from different origin were compared. As can be seen from Table 2, in harp seal blubber the 2,3,7,8-TEQ levels for the three coplanar PCB CB 77,126 and 169 are a factor of about 3–10-times higher than for PCDD/PCDF. This is within the range found by Daelemans et al. (1993) and Muir et al. (1992). Mono-ortho substituted PCB were not included in this calculation since they were only determined in five blubber samples. According to a study from the Canadian Arctic the concentrations of mono-ortho-substituted PCB in ringed seal blubber correspond to about 30% of the 2,3,7,8-TEQ levels of the coplanar PCB (Muir and Ford, 1990).

The concentration ratios of the coplanar PCB in seal blubber from ringed seals from the Canadian Arctic (Muir and Ford, 1990) and from the harp seals varied significantly. The ratio between CB 77 and CB 126 in harp seal blubber varied between 0.3 to 1.4 which is similar to the results reported for Canadian ringed seal blubber (0.3–2.4, Muir and Ford, 1990). The reason for this large difference between individuals is unknown. However, this shows that important information about the variability of concentration ratios is lost when only pooled samples are analysed.

The picture is even more complex for CB 169. Daelemans et al. (1993) reported the level in ringed seal blubber from Spitsbergen as being below an unspecified detection limit. In ringed seal blubber from the Canadian Arctic the level of CB 169 was always much lower than of CB 77 and CB 126 (Muir and Ford, 1990). To our surprise the CB 169 concentrations were the highest of all three congeners in some harp seal blubber samples (see Table 2). They were sometimes one order of magnitude higher than in ringed seal blubber from the Canadian Arctic (Muir and Ford, 1990). During a recent intercalibration, the applied analysis method showed a reasonable agreement for CB 169 (Sergeant et al., 1993). Furthermore, a co-elution with another congener can be excluded on the applied stationary phase (see Experimental). Therefore, at present we cannot see that methodological problems are the reason for the found high levels of CB 169 in harp seal blubber.

3.3. Other polychlorinated biphenyls

Since the first report of Holden (1972) about the presence of PCB in Arctic seals, numerous PCB determinations have been carried which are summarised in the review of Muir et al. (1992). However, until the mid-1980s PCB levels were given as total PCB (ΣPCB) quantified as equivalents of commercial PCB products (e.g., Arochlor 1254, 1260, or mixtures of both). Compared with a complete isomer-specific analysis this leads to an overestimation of the ΣPCB by nearly a factor of 2 (Muir et al., 1988). In newer studies mainly single isomers are quantified and summarised to ΣPCB which makes a direct comparison with older ΣPCB determinations doubtful. In addition, the toxicity, bioaccumulation and metabolisation of different PCB congeners vary very much. Consequently, PCB levels should be reported as concentrations of single PCB congeners. However, the only PCB data available for harp seals were given as ΣPCB based on a quantification as Arochlor 1260 (Ronald et al., 1984). Therefore, in the present study ΣPCB levels were calculated as Arochlor 1260 equivalents by multiplying the sum concentrations of PCB 138 + 163 and 153 with a factor of 5 according to Ballschmiter and Zell (1980). As can be seen from Table 4, the sum of the quantified PCB congeners was almost three-times lower than the calculated ΣPCB levels.

ΣPCB concentrations in harp seal blubber (average 2560 ± 2865 ng/g wet weight) estimated from PCB 138 + 163 and 153 (see Table 4) were comparable to those reported by Ronald et al. (1984) for male harp seals from northern Baffin Bay (3760 ± 3650 ng/g wet weight). Levels were in the same range in male ringed seals from the Canadian Arctic (Muir et al., 1992) and from Spitsbergen (Oehme et al., 1988, 1990; Daelemans et al., 1993). However, one harp seal sample

Table 4
Concentrations of polychlorinated pesticides, hexachlorbenzene and polychlorinated biphenyls in seal blubber (*Phoca groenlandica*): all results are given in ng/g wet weight; the lipid concentration for all samples varied between 83 and 86%

Sample no.	1	3	4	5	6	7	8	9	11		
Sampling site	75°10'N 10°06'W	72°49'N 09°01'W	72°52'N 09°10'W	74°00'N 13°30'W	74°00'N 13°30'W	75°00'N 09°29'W	75°33'N 04°59'W	75°33'N 04°59'W	75°33'N 04°59'W	Average x	Stand. dev.
Sex	female	male	male	male	male	male	male	male	male		
Age	1	3	16	18	6	adult	13	10	10		
HCB	210	87.4	277	61.0	176	179	47.6	33.0	83.3	128.3	84.7
α-HCH	21.1	12.5	33.3	42.3	28.6	24.7	26.4	19.7	22	25.6	8.6
α(+)/α(−) ratio	1.68	1.23	1.70	1.08	1.43	1.29	1.28	2.45	1.27	1.5	0.4
β-HCH	3.9	< 2.4	10.6	7.5	6.3	4.4	8.7	5.3	2.8	6.2	2.6
γ-HCH	2.1	< 1.9	3.4	6.3	5.5	2.8	7.8	4.2	2.0	4.3	2.1
PCB no. 28	2.4	3.4	< 2	< 2	4.9	< 2	< 2	5.8	3.6	3.1	1.4
PCB no. 31	1.1	1.3	n.a.	n.a.	3.6	n.a	n.a.	3.4	2.8	2.4	1.2
PCB no. 52	13.2	18.8	22.7	40.9	22.9	35.7	16.7	26.3	20.0	24.1	9.0
PCB no. 101	25.8	29.1	60.5	83.3	29.6	90.0	21.1	41.7	39.1	46.7	25.5
PCB no. 105	5.8	7.6	n.a.	n.a.	8.2	n.a.	n.a.	11.1	9.8	8.5	2.0
PCB no. 118	16.2	18.4	n.a.	n.a.	32.0	n.a.	n.a.	35.6	29.2	26.3	8.5
PCB no. 153	48.2	58.1	484	1041	80.0	550	200	111	76.9	294.4	337.4
PCB no. 156	1.7	2.2	n.a.	n.a.	3.5	n.a.	n.a.	4.8	3.1	3.1	1.2
PCB no. 138 + 163[a]	42.5	50.0	325	769	75.0	346	183	97.1	76.6	218.2	236.3
PCB no. 180	13.3	20.0	168	228	23.8	157	63.64	30.7	22.2	80.7	81.3
ΣPCB[b]	156.9	188.9	—	—	259.7	—	—	336.8	261.1	—	—
ΣPCB[c]	453	541	4045	9050	775	4480	1915	1041	768	2560	2865
p,p'-DDE	116	176	1047	2757	273	1041	736	479	230	761.7	829.4
p,p'-DDT	26.6	48.4	78.1	263	40.0	52.6	90.9	41.7	31.6	74.8	73.6
p,p'-DDD	8.6	27.4	14.9	54.5	21.9	21.7	17.4	11.1	8.7	20.7	14.2
ΣDDT	151	252	1140	3075	335	1115	844	539	270	857.9	911.7
ΣPCB/ΣDDT	3.0	2.2	3.6	2.9	2.3	4.0	2.3	1.9	2.8	2.8	0.7

[a]Also co-elution with PCB 164.
[b]Calculated on basis of the nine PCB measured.
[c]Calculated from PCB 138 and 153 according to Ballschmiter and Zell (1980).

had an extreme high PCB concentration of 9.0 μg/g.

Single PCB congeners were also determined in brain (see Table 5). PCB levels in brain from Arctic seals have not been reported before. Unfortunately, insufficient storage conditions (too high temperature in one of the freezers) did not allow to determine PCB in brain from all animals. As can be seen from Table 5, the blubber/brain concentration ratios, based on a wet weight basis, differed considerably both between animals and different congeners. The ratio for PCB 52 in males covered a large range [2,5–14] and varied even more for PCB 153 [5–37]. In one sample (no. 9) an extremely low ratio (0.6) was found due to high PCB levels in the brain tissue. Although no obvious reason for a contamination was found, one should interpret this result with caution. We feel that the PCB levels in brain are surprisingly high compared to blubber. However, more data together with a reliable determination of the lipid content are needed for a more thorough evaluation.

The average and standard deviation of the PCB concentrations relative to PCB 153 were calculated on the basis of the results for individual harp seal blubber samples. Fig. 3 compares them with those reported for pooled ringed seal blubber

Table 5
Concentrations of polychlorinated pesticides, hexachlorobenzene and polychlorinated biphenyls in harp seal brain (*Phoca groenlandica*): all results are given in ng/g wet weight

Sample no.	1	3	4	6	7	8	9	10	11
Sampling site	75°10'N 10°06'W	72°49'N 09°01'W	72°52'N 09°10'W	74°00'N 13°30'W	75°00'N 09°29'W	75°33'N 04°59'W	75°33'N 04°59'W	75°33'N 04°59'W	75°33'N 04°59'W
Sex	female	male	male	male	male	male	male	male	male
Age.	1	3	16	6	adult	13	10	4	10
HCB	0.94	1.83	8.2	6.3	3.8	1.89	25.3	1.47	1.06
α(+)-HCH	6.2	2.5	9.9	9.6	6.7	6.5	10.0	3.9	5.5
β-HCH	1.71	< 0.5	1.25	< 0.5	2.0	3.0	3.9	1.53	< 0.5
γ-HCH	1.84	3.4	1.27	< 0.7	6.9	3.6	4.6	1.94	1.92
PCB no. 28	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
PCB no. 52	5.2	3.1	1.67	6.3	3.6	6.7	24.7	3.2	4.0
PCB no. 101	5.2	6.3	7.6	19.3	8.6	17.1	26.6	6.2	6.5
PCB no. 153	5.5	9.1	17.3	15.1	15.0	39.5	187	6.6	8.4
PCB no. 138 + 163	5.5	9.0	16.7	14.4	12.2	30.4	147	6.5	7.4
PCB no. 180	< 0.6	3.3	5.6	5.6	5.9	11.1	33.6	2.3	3.4
p,p-DDE	1.9	3.1	11.9	8.3	10.4	22.9	64.4	3.4	9.0
p,p'-DDT	< 0.6	1.7	1.3	2.9	1.7	12.5	24.4	1.7	1.8
p,p'-DDD	< 0.6	< 0.6	< 0.6	< 0.6	< 0.6	5.6	16.4	< 0.6	< 0.6

[a]Also co-elution with PCB 164.

from the Canadian Arctic (Muir et al., 1988) and from Spitsbergen (Daelemans et al., 1993). Within the given standard deviation, the PCB congener patterns in harp seals and ringed seals are very similar.

3.4. Chlorinated pesticides

Levels of p,p'-DDE in harp seal blubber from males from the Greenland Sea (Table 4) were lower than in ringed seals from Spitsbergen (Oehme et al., 1988; Daelemans et al., 1993) and in male harp seals from the Canadian Arctic (Ronald et al., 1984). However, the concentrations in ringed seal blubber from Spitsbergen were much higher than in animals from any region of the Canadian Arctic (Muir et al., 1992). The higher levels found in male harp seals from the northern Baffin Bay region might also reflect a generally higher p,p'-DDE burden in 1984 compared to 1992 and not a geographical difference. Concentrations of p,p'-DDT and p,p'-DDD were on average 10- and 40-times lower, respectively. No significant correlation ($P < 0.05$) could be found between levels of p,p'-DDE, p,p'-DDT and p,p'-DDD in harp seal blubber. In Arctic ringed seal blubber, much lower p,p-DDE/p,p'-DDT ratios were found (Oehme et al., 1988; Luckas et al., 1990). In most publications, DDT levels are still reported as total concentrations of p,p'-DDE, p,p'-DDT and p,p'-DDD (Muir et al., 1992). Single values for all three compounds in Arctic seal blubber are only occasionally given.

α-HCH and total HCH concentrations in harp seal blubber were considerably lower than in ringed seal blubber from the Arctic while β- and γ-HCH levels were comparable (Carlberg et al., 1981; Oehme et al., 1988; Muir et al., 1992). As can be seen from Fig. 4, much higher α-/γ-HCH ratios were found in ringed seal blubber from the Arctic (40–60:1, Carlberg et al., 1981; Oehme et al., 1988) in contrast to the North Sea (2–3:1, Luckas et al., 1990; Oehme et al., 1990). However, in the analysed harp seal blubber, the ratio was much lower and varied between 3 and 11. In addition, the hexachlorobenzene levels were about one order of magnitude higher than in ringed seals (Fig. 4). This resulted in a HCB/ΣHCH ratio of 3–4 in harp seals compared to 0.1–0.6 in

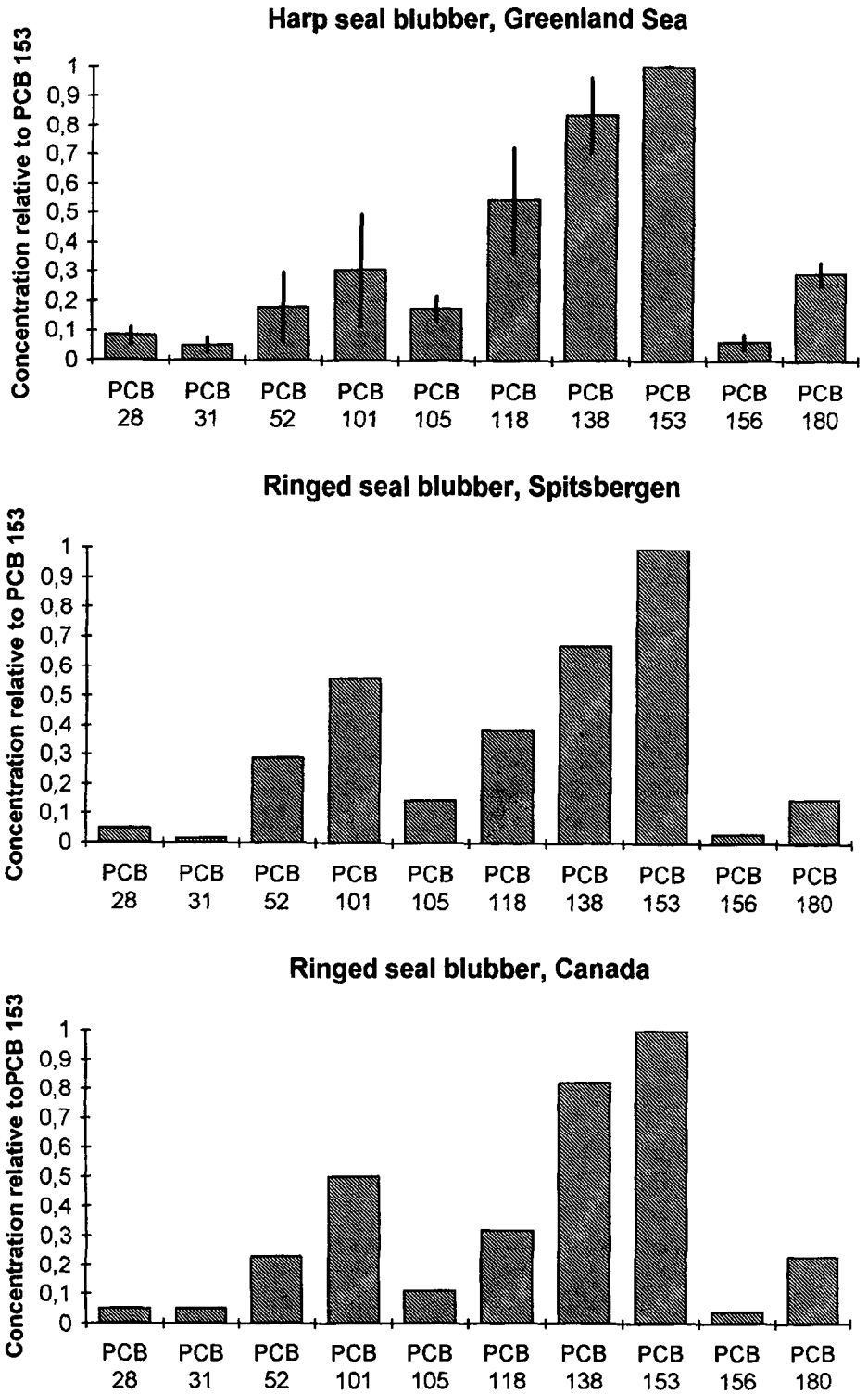

Fig. 3. Average concentration profiles of selected PCB congeners relative to PCB 153 in seal blubber from Arctic ringed seals (Spitzbergen, Daelemans et al. (1993), and Canada, Muir et al. (1988)) and harp seals. The standard deviation of the relative concentrations of individual animals is given for the harp seal samples.

ringed seals (Muir et al., 1988, Luckas et al., 1990).

α-HCH is the only HCH isomer which is chiral and exists as two enantiomers. While the enantiomer ratio in the technical product is exactly 1.0 (also called a racemate), the different enantiomer metabolisation rate in marine mammals changes the ratio. In harbour seal blubber (*Phoca vitulina*)

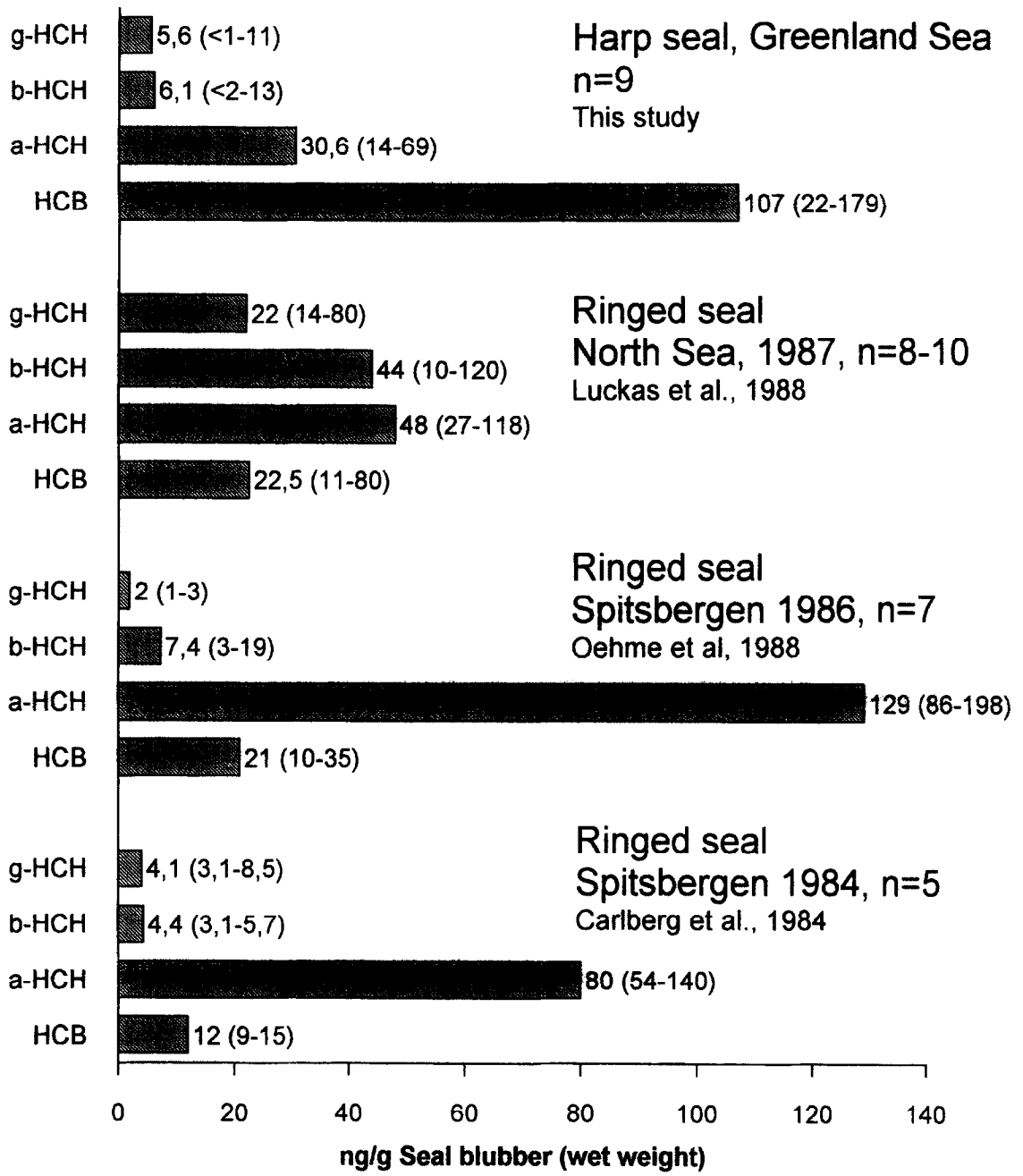

Fig. 4. Comparison of the concentration profiles for hexachlorocyclohexanes and hexachlorobenzene in ringed seal (Spitsbergen) and harp seal blubber.

$\alpha(+)/\alpha(-)$ enantiomer ratios of about 1.3–5 have been reported (Müller et al., 1992; Hühnerfuss et al., 1992). Ratios in seal blubber close to one might indicate either a recent uptake or a weakened ability to metabolise α-HCH. As can be seen from Table 3, the $\alpha(+)/\alpha(-)$ enantiomer ratio in harp seal blubber varied from 1.1 to 2.5. No relationship between the α-HCH enantiomer ratio and age was found. Only $\alpha(+)$-HCH was found in harp seal brain indicating a highly selective accumulation process via the blood–brain barrier. The same observation was reported for harbour seals by Hühnerfuss et al. (1992) who found about 5–10-fold higher $\alpha(+)$-HCH levels per lipid weight in brain than in blubber. However, no information was given about the lipid determination method and what type of compounds the term lipids included. Our results show a brain/blubber $\alpha(+)$-HCH ratio of 0.3–1.3 on a wet weight basis. A recalculation, assuming a phospholipid content of 30–40% in brain, results in ratios which are still considerably lower than the values given by Hühnerfuss et al. (1992).

3.5. Correlations between different compound groups as well as age

The different compound groups quantified in the seal blubber extracts are quite different in terms of sources, transport mechanisms, toxicological interactions, bioaccumulation and metabolisation. The data obtained were analysed for possible correlations between single compounds and substance classes as well as between age and concentration levels. Although the number of individual was very limited, the following correlations, at a significance level of $P < 0.05$, were found (see Table 6).

The levels of PCB congeners with five or more Cl atoms correlated very well with those of p,p'-DDE (e.g., for PCB 153, $r = 0.977$, see Fig. 5). However, no relationship was found between di-ortho-substituted PCB and any of the PCDD/PCDF congeners or non-ortho PCB (see Fig. 5 and Table 6). Both non-ortho and other PCB are part of technical PCB mixtures, and one can expect that congeners with the same number of chlorines behave in a similar way with regard to long-range transport to the Arctic. However, di-ortho- and non-ortho-substituted PCB behave completely differently regarding distribution in the body, half life and enzyme-inducing properties (see Ahlborg et al. (1992), for a detailed survey). Di-ortho-substituted PCB have much higher half lives and cannot be biotransformed to hydroxylated metabolites if the 4 and 5 positions are occupied (e.g., PCB 153). Non-ortho-substituted PCB have dioxin-like properties and accumulate especially in the liver. Norstrom et al. (1990) found that ΣPCB concentrations were highly correlated with 2,3,7,8-TCDF ($P < 0.01$) and 2,3,7,8-TCDD levels ($P < 0.05$) in ringed seals from the Canadian Arctic. They also found a significant correlation for PCB and 1,2,3,7,8-PCDD but not for 2,3,7,8-TCDF in ringed seals from Spitsbergen (Oehme et al.,1988). However, the rather small number of samples ($n = 7$) in the study of Oehme et al. (1988) contained two animals with 1,2,3,7,8-PCDD levels which were nearly

Table 6
Selected correlation coefficients between levels of polychlorinated compounds and age in seal blubber from male harp seals: the number of samples is given in parantheses

Compound	PCB 126	1,2,3,7,8-PeCDD	p,p-DDE	α-HCH	Age
PCB 153	0.342 (8)	0.416 (8)	**0.977*** (8)	**0.843*** (8)	**0.875**** (7)
γ-HCH	—	—	0.478 (8)	0.633 (8)	0.545 (7)
PCB 126	—	**0.967*** (9)	—	—	0.298 (8)
HCB	0.022 (7)	0.123 (7)	0.018 (8)	0.162 (8)	0.347 (7)
2,3,7,8-TCDD	**0.891*** (9)	**0.884*** (9)	—	—	0.112 (8)
2,3,7,8-TCDF	0.022 (9)	0.020 (9)	—	—	0.336 (8)
Age	0.305 (8)	0.250 (8)	**0.804**** (7)	**0.834**** (7)	

*r significant at $P < 0.01$; **r significant at $P < 0.05$.

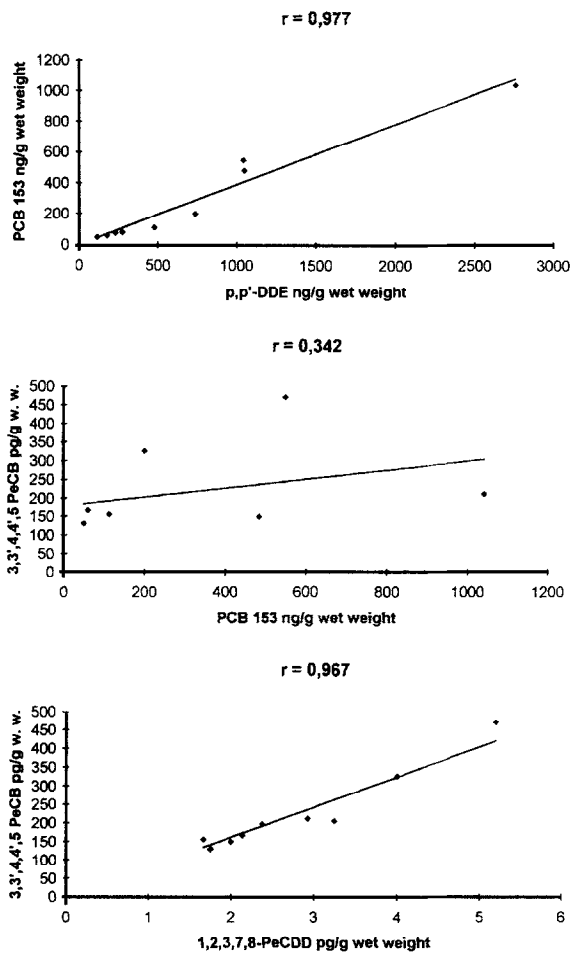

Fig. 5. Correlation diagrams for selected polychlorinated compounds.

one order of magnitude higher. They were exclusively responsible for the found trend.

A surprisingly good correlation between levels of 1,2,3,7,8-PeCDD and 3,3',4,4'5-PeCB was found (see Fig. 5). They obviously behave in a very similar way with regard to bioaccumulation in harp seals. These congeners were selected since they were present at levels far above the detection limits, and their quantification is normally not affected by interferences or influenced by other analytical problems. Non-ortho PCB and PCDD/PCDF levels, expressed as 2,3,7,8-TEQ, showed also a significant but weaker trend ($r = 0.84$). The main reason is that most of the PCDF congeners levels did not have the same strong relationship to non-ortho PCB as PCDD.

As can be seen from Table 6, α-HCH levels correlated well with those of PCB 153. Comparing α-HCH with γ-HCH, a weaker relationship just above the significance level for $P < 0.1$ was found. This indicates that PCB 153 and α-HCH have more similar behaviour in terms of bioaccumulation and persistence than α- and γ-HCH.

Finally, despite the limited number of analysed samples, levels of penta- and hexachloro-PCB, such as PCB 153 as well as p,p'-DDE, were positively correlated with age for the male seals (see Table 6). Muir et al. (1988) reported a similar correlation. However, no significant trends with age was found for any PCDD/PCDF as reported by Bignert et al. (1989) and Norstrom et al. (1990).

4. Conclusions

The determination of selected polychlorinated compounds in seal blubber from male harp seals permitted the following conclusions.

- Levels of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in harp seals are lower than in ringed seals from the Arctic. Interspecies or geographical differences may be the reason.
- No correlation with age was found for any of the PCDD/PCDF congeners.
- Levels of the non-ortho-substituted PCB CB 77 and 126 were comparable with those found in ringed seal blubber from the Arctic. An exception was CB 169, which showed variable concentrations. In some samples it was the most dominant non-ortho-substituted congener.
- ΣPCB concentrations were comparable to those reported in 1984 for harp seals from the northern Baffin Bay region and to those found in ringed seals from the Arctic.
- The concentrations of 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD and other PCDD correlated strongly with CB 126 but not with CB 77, CB 169, 2,3,7,8-TCDF, other PCDF, di-ortho PCB or any other measured compound.

- A significant trend between age and major di-ortho-substituted PCB compounds (e.g., PCB 153) and p,p'-DDE was found, but this was not the case for non-ortho PCB and PCDD/PCDF.
- PCDD/PCDF congener patterns relative to 1,2,3,7,8-PeCDD were similar for ringed seal and harp seal blubber. The same was found for the PCB patterns relative to PCB 153 in ringed seals from Spitsbergen, the Canadian Arctic and the harp seals analysed in this study.
- There might be a difference in the PCDD/PCDF patterns of Arctic and Antarctic seals.

Acknowledgements

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