

# PCB's and other organochlorine contaminants in white whales (*Delphinapterus leucas*) from West Greenland: variations with age and sex

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Blubber samples from 138 white whales from two locations in West Greenland (the Nuussuaq area, ca. 74°N, 58°W, and the Disko Bugt area, ca. 69°N, 53°W) and skin and kidney samples from 20 white whales from Nuussuaq were analysed for PCB congeners and related organochlorines (hexachlorohexanes ( $\Sigma$ HCH), chlorinated bornanes ( $\Sigma$ CHB), chlordanes ( $\Sigma$ CHLORD), DDT-related compounds ( $\Sigma$ DDT), dieldrin and mirex). The large sample size permitted a detailed assessment of the variation of contaminant levels with age and sex. Mean concentrations of all major organochlorine (OC) groups ( $\Sigma$ PCB,  $\Sigma$ DDT,  $\Sigma$ CHLORD,  $\Sigma$ CHB) were not significantly different between the Nuussuaq and Disko Bugt groups. After an age of 5.5 years, a consistent decline in concentration levels of all four major OC's was observed in females. In contrast, only marginal differences in  $\Sigma$ PCB,  $\Sigma$ CHB and  $\Sigma$ CHLORD concentrations and a substantial increase in  $\Sigma$ DDT were observed in males. The observed decline in females occurred near the age of attainment of sexual maturity and preceding commencement of lactation. Residue levels in sexually immature animals (male and female) and male-female differences in mean concentrations of  $\Sigma$ PCB and p,p'-DDE in adult animals are consistent with a lactation period of less than one year. Young animals could be distinguished from adults, using principal component analysis, by higher proportions of lower chlorinated PCB's and more watersoluble OC's such as p,p'-DDE which are preferentially transferred during lactation.

## Key words:

White whale, beluga, *Delphinapterus leucas*, Greenland, age, sex, lactation, parturition, organochlorine, PCB, DDT.

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## Introduction

Previous studies of white whales (*Delphinapterus leucas*) from Canadian and Alaskan waters indicated that organochlorine chemicals such as polychlorinated biphenyls (PCB's) and other organochlorines (OC's) were present at low parts per million ( $\mu$ g/g) concentrations in blubber samples of all stocks (Muir *et al.* 1990, Becker *et al.* 1992). The presence of these contaminants, which are associated with reproductive failure in other marine

mammals and birds (e.g. Peakall 1975, Reijnders 1986), is of concern from the point of view of the long-term health of the white whale populations (Martineau *et al.* 1987, Muir *et al.* 1990). The white whale is important to the Inuit population in arctic coastal communities of Greenland, Canada and Alaska, since marine mammals constitute an important part of their traditional diets. Knowledge of levels of the contaminants in the whale tissues is useful for assessing the extent of dietary exposure (Kinloch *et al.* 1992).

In the studies by Muir *et al.* (1990) and Becker *et al.*

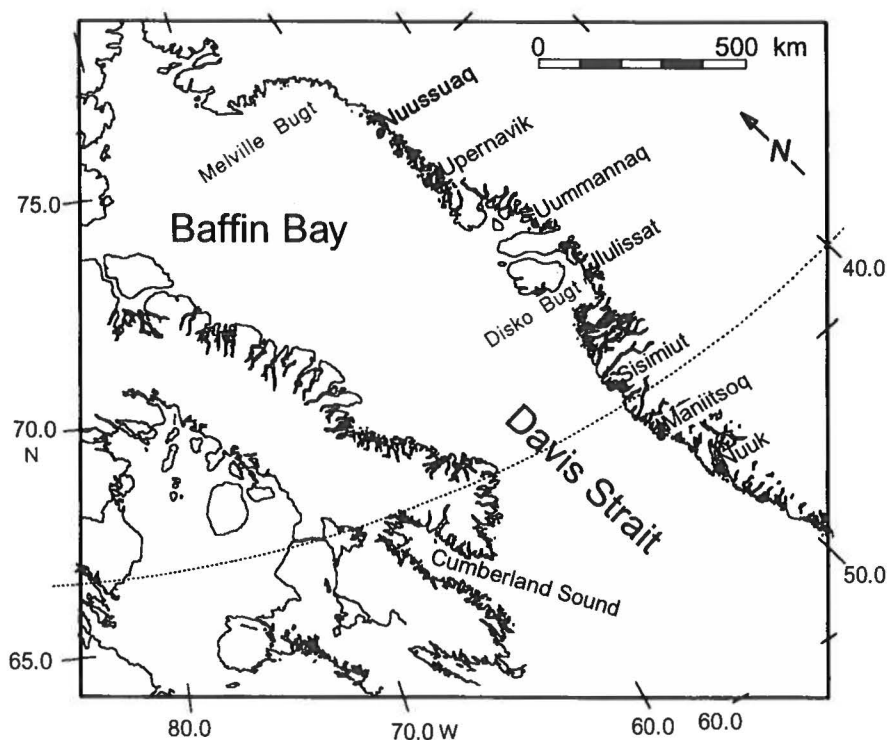


Fig. 1. Map showing Nuussuaq and Disko Bugt regions of West Greenland where white whale samples were collected.

(1992) comparisons of contamination levels between stocks were carried out on a limited number of sex and age classes. The main objective of the present study, was to examine the level and the influence of sex and age on concentrations of major contaminants in white whales sampled in western Greenland. This knowledge should enable better design of future monitoring studies of white whales and other small toothed cetaceans. A second objective was to provide information on levels of contaminants in skin (mattak) and kidney for which figures have not previously been reported.

## Materials and methods

### Samples

A total of 174 white whale blubber, 113 skin and 110 kidney samples from the Nuussuaq (ca. 74°N, 58°W) and Disko Bugt (ca. 69°N, 53°W) areas of West Greenland (Fig. 1) were received from the Greenland Environmental and Fisheries Research Institutes during 1989 and 1990. Details of the collection procedure were given by Hansen *et al.* (1990) and Heide-Jørgensen & Teilmann (1994). Samples were shipped frozen in styrofoam insulated containers and were stored upon receipt at -40°C. Animals were classified by sampling location, standard length and sex. Age was estimated as described by Heide-Jørgensen *et al.* (1994). From the 174 blubber samples, 138 were

selected for analysis to cover a range of ages with approximately equal numbers of males and females. Analysis was performed on 20 skin and kidney samples selected from animals containing the highest contaminant levels in blubber. All were age 5 years and younger.

### Organochlorine Analysis

Determinations of PCB's and organochlorine pesticides in blubber tissues followed the procedures described by Muir *et al.* (1990). Briefly, samples of blubber (2.2 g) were mixed with anhydrous sodium sulfate (pretreated by heating at 600°C for 6 hours) and ball-milled (30 minutes) with hexane. The extract was centrifuged and a portion (1/11) removed for lipid determination. Internal standards of aldrin and octachloronaphthalene (OCN) were added to a 0.5 ml aliquot of the extract which was then fractionated on Florisil® (1.2% (v/w) deactivated with water) into three eluates; hexane (F1), hexane: DCM (85:15) (F2) and hexane DCM (1:1) (F3) (Norstrom & Won 1985). The chromatography on Florisil separated PCB's, chlorobenzenes, p,p'-DDE and mirex in F1 from most toxaphene components, chlordane-related compounds and p,p'-DDT in F2. F3 contained heptachlor epoxide and dieldrin. Florisil eluates were evaporated to about 2 ml, transferred to vials and made up to 2.5 ml in isoctane for gas chromatographic (GC) analysis.

Skin and kidney samples, because of their lower lipid

Table 1. Age groups and numbers for male and female white whales used in statistical comparisons.

Group	Age (years)	Sex	N
1	NB - 1.0	M	8
		F	6
2	1.5 - 2.5	M	19
		F	11
3	3.0 - 4.0	M	11
		F	13
4	4.5 - 7.5	M	20
		F	19
5	8.0 - 15.0	M	9
		F	14
6	> 15.0*	M	4
		F	4

\* Oldest male and female animals were 22.0 and 18.5 years, respectively.

content, were analysed as above with some minor modifications: 5.0 g samples were homogenized by blending with dry ice. The dry ice was allowed to sublime and the homogenate was mixed with anhydrous sodium sulfate (pretreated by heating at 600°C for 6 hours) and ball-milled (30 minutes) with hexane. Internal standards of aldrin and OCN were added and an aliquot of the extract equivalent to about 0.1 g of lipid was then fractionated on Florisil as above.

Florisil eluates were analysed by capillary GC with (<sup>63</sup>Ni) electron capture detection. Samples were injected (splitless mode) on a 60 m × 0.25 mm i.d. DB-5 column (film thickness 0.25 μm) with H<sub>2</sub> carrier gas (Muir *et al.* 1988). PCB's and organochlorine pesticides were identified and quantified as described previously (Muir *et al.* 1988). Total PCB (ΣPCB) was the sum of all congeners. Total chlordanes (ΣCHLORD) was the sum of all chlordanes-related compounds including heptachlor epoxide while total DDT (ΣDDT) was the sum of p,p'-DDE, -DDD, -DDT and o,p'-DDT. Chlorinated bornanes (CHBs) were quantified after first obtaining response factors for individual CHB peaks from the weight percent of each peak in the total ion chromatogram (electron ionization GC-mass spectrometry) of a toxaphene standard. Total CHBs (ΣCHB) was the sum of the areas of 19 peaks (8 major peaks were generally observed) (Muir *et al.* 1990).

Internal standard recoveries were determined in all samples. Recoveries of aldrin and OCN were uniformly greater than 90%. A cod liver standard reference material (SRM-1588) from the National Institute of Standards and Technology (Gaithersburg, Va) was employed for major organochlorine pesticides and PCB congeners.

## Statistical Analysis

Correlation analysis was used to measure the relationship between concentrations of major contaminants and ratios

of selected compounds with age and length. The general linear models procedure of SAS (SAS Institute 1992) with an analysis of covariance was then used to test the hypothesis that mean concentrations of major contaminants and ratios of selected compounds did not vary with age group. To compare whales at varying stages of growth, blubber samples from both males and females were assigned to one of six different age groups (Table 1). Where there were significant effects ( $P < 0.05$ ), the mean square error from the ANCOVA was used with Tukey's test to identify the age groups which differed in mean concentrations (wet weight). Principal component analysis (PCA) was conducted using the statistical software program SIRIUS (Karstang & Kralheim 1990) to examine pattern similarities and differences between mean concentrations of individual PCB and organochlorine contaminants in blubber samples with age group and sex. Mean concentrations for each age group were normalized (expressed as a fraction of the combined sum of the concentrations of all residues) to avoid the influence of absolute concentrations. Autoscaling to unit variance was used to minimize any statistical bias associated with the order-of-magnitude differences in chemical concentration. The number of statistically significant principal components was determined using cross validation (Sharaf *et al.* 1985). Contaminants with the highest contributions to the differences observed between the individual age groups and clusters will be located furthest from the origin of the axis. The greater the distance along an individual principal component axis, the more an individual contaminant contributes to the variation accounted for along that axis. A negative or positive position along the axis indicates whether a certain contaminant is negatively or positively correlated with the principal component. An age group or cluster positioned along a positive principal component axis will contain low concentrations of contaminants with negative loadings and higher concentrations of contaminants with positive loadings. An age group or cluster positioned along a negative principal component axis contains the opposite relationship. As a result, the position of an age group or cluster on a principal component axis relative to other age groups or clusters indicates differences in contaminant profiles.

## Results

### Age and sex differences

Mean concentrations (arithmetic) of major organochlorine groups (ΣPCB, ΣDDT, ΣCHB, ΣCHLORD) and p,p'-DDE in male and female blubber samples were calculated for animals of the same age (Tables 2 and 3, respectively). The relationships between age and the resulting mean concentrations of ΣPCB, ΣDDT, ΣCHB and ΣCHLORD and the ratios p,p'-DDE/ΣDDT and ΣDDT/ΣPCB of males and females are described in Figs.

Table 2. Arithmetic mean concentrations of major contaminants with age in male white whale blubber from West Greenland ( $\mu\text{g/g}$  wet weight).

AGE	N	$\Sigma\text{PCB}$	$\Sigma\text{DDT}$	$\Sigma\text{CHB}$	$\Sigma\text{CHLORD}$	p,p'-DDE
0.0	2	6.75	5.28	4.82	2.94	3.20
0.25	1	10.87	8.10	6.22	4.86	5.80
0.75	1	1.84	1.01	2.36	1.04	0.52
1.0	4	6.48	3.92	4.49	2.51	2.33
1.5	6	5.59	3.61	4.06	2.65	2.06
2.0	6	4.83	3.59	4.43	2.64	1.78
2.5	7	6.51	5.22	5.08	3.47	3.28
3.0	8	5.44	4.00	3.50	2.48	2.42
3.5	1	3.56	1.90	2.47	1.63	0.91
4.0	2	4.84	2.31	2.38	1.82	1.25
4.5	2	4.54	2.79	3.57	2.12	1.49
5.0	8	5.17	3.98	3.31	2.21	2.26
5.5	2	3.58	2.88	2.73	2.06	1.60
6.0	6	5.52	4.18	2.74	2.05	2.53
7.0	2	5.32	3.13	3.15	1.84	1.84
8.0	1	7.37	5.44	4.46	2.96	2.83
10.0	1	3.36	2.14	2.68	1.28	1.12
11.0	1	5.53	4.94	4.46	2.69	2.95
12.0	3	3.68	2.54	2.54	1.48	1.48
13.0	3	4.90	5.03	3.39	2.05	2.87
16.0	1	6.05	6.85	4.13	2.35	3.35
17.0	1	5.62	7.02	3.65	2.33	3.52
21.0	1	5.79	5.61	2.72	2.07	3.25
22.0	1	4.42	5.38	2.74	1.77	2.89

2, 3 and 4 (preliminary statistical analysis indicated that geometric means calculated from  $\log_{10}$  transformed data were generally only slightly lower than the corresponding

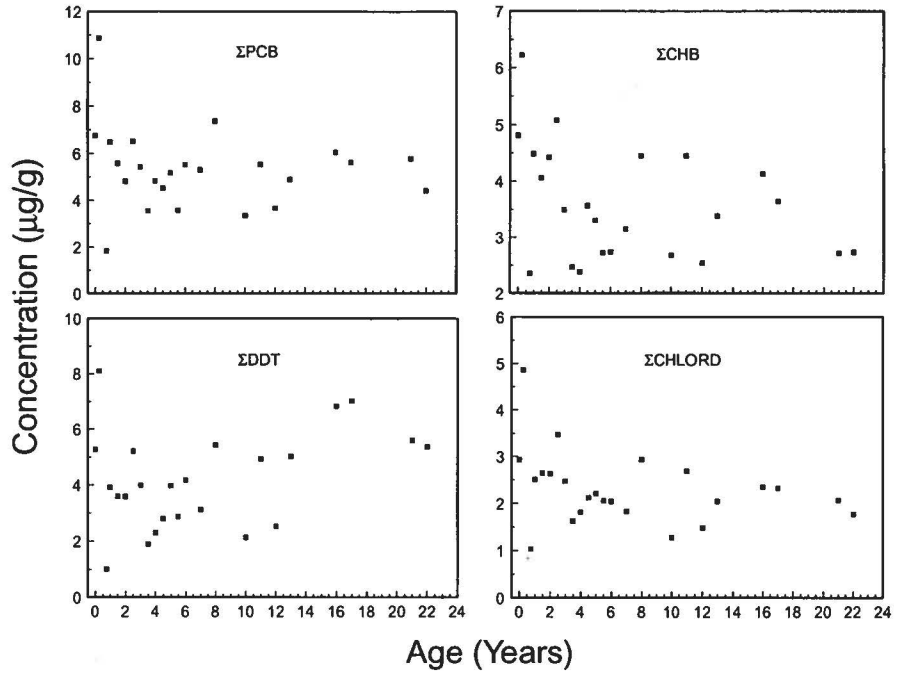
arithmetic means. Arithmetic means were therefore used in Tables 2 and 3 and Figs. 2, 3 and 4).

With the exception of mirex and  $\Sigma\text{OCTA}$  (octachlor-

Table 3. Arithmetic mean concentrations of major contaminants with age in female white whale blubber from West Greenland ( $\mu\text{g/g}$  wet weight).

AGE	N	$\Sigma\text{PCB}$	$\Sigma\text{DDT}$	$\Sigma\text{CHB}$	$\Sigma\text{CHLORD}$	p,p'-DDE
0.25	1	7.16	5.36	5.38	3.52	3.07
0.50	1	3.86	2.41	5.43	2.12	1.24
0.75	1	2.84	1.73	3.65	1.65	0.86
1.0	3	5.10	4.53	5.66	3.35	2.16
1.5	1	2.43	1.35	3.33	1.46	0.62
2.0	6	5.34	3.40	4.55	2.67	1.78
2.5	4	5.48	4.17	4.23	2.66	2.51
3.0	4	5.62	3.49	4.73	2.71	1.80
3.5	6	5.10	3.61	3.31	2.15	2.17
4.0	3	4.91	2.93	2.94	1.93	1.76
4.5	1	6.97	4.80	2.65	2.52	2.81
5.0	3	4.50	3.80	3.35	2.22	2.18
5.5	2	5.29	4.38	3.65	2.68	2.54
6.0	2	2.89	2.06	2.73	1.38	1.04
7.0	8	3.68	2.76	2.31	1.64	1.63
7.5	3	2.53	1.57	2.04	1.20	0.86
8.0	1	0.84	0.35	0.55	0.33	0.18
9.0	3	2.63	2.79	1.75	1.13	1.42
10.0	3	0.91	0.44	1.31	0.41	0.20
11.0	1	1.00	0.45	1.23	0.49	0.23
12.0	1	1.10	0.56	1.55	0.50	0.23
13.0	2	1.45	1.40	1.44	0.66	0.66
14.0	2	1.17	0.67	1.65	0.56	0.32
15.0	1	0.61	0.30	1.00	0.30	0.14
17.0	3	0.93	0.77	1.24	0.50	0.32
18.5	1	0.94	0.50	1.40	0.43	0.26

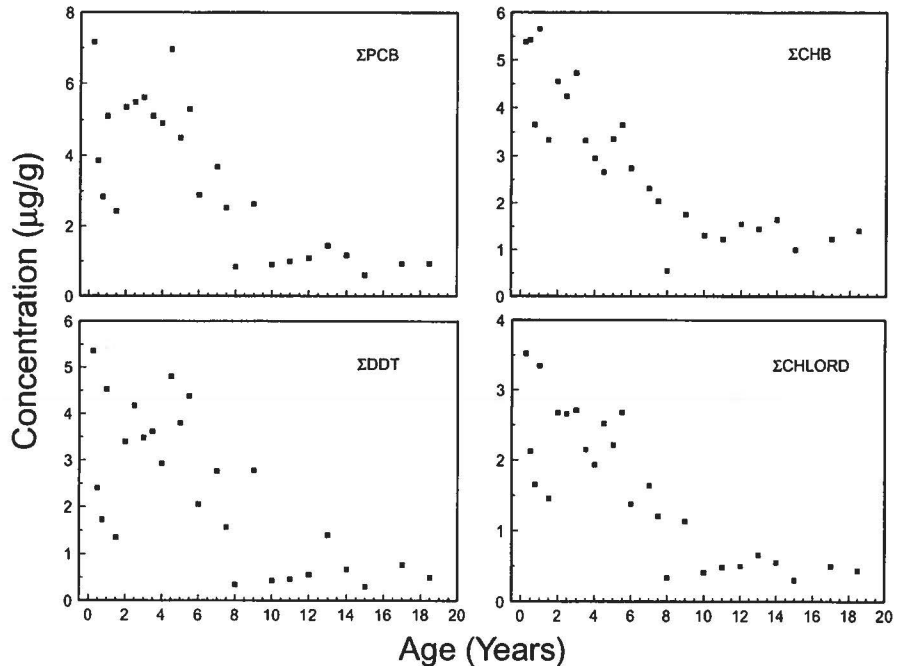
Fig. 2. Relationships between age and the arithmetic mean concentrations (for animals of the same age) of  $\Sigma$ PCB,  $\Sigma$ DDT,  $\Sigma$ CHB and  $\Sigma$ CHLORD in blubber samples of male white whales from West Greenland.



inated biphenyls), significant ( $P < 0.01$ ) negative correlations were observed for both age and length in female blubber samples (Table 4). In male blubber samples, dieldrin,  $\Sigma$ HCH and  $\Sigma$ TRI (trichlorinated biphenyls) were significantly negatively correlated and mirex and  $\Sigma$ OCTA significantly positively correlated with age and length. The p,p'-DDE/ $\Sigma$ DDT concentration ratio was sig-

nificantly negatively correlated with age in females and the  $\Sigma$ DDT/ $\Sigma$ PCB concentration ratio significantly positively correlated with both age and length in males. In kidney samples, age was significantly ( $P < 0.05$ ) negatively correlated with  $\Sigma$ CBz and  $\Sigma$ TRI concentrations in females and dieldrin,  $\Sigma$ HEXA and  $\Sigma$ HEPTA concentrations in males (Table 5). In contrast to what was observed

Fig. 3. Relationships between age and the arithmetic mean concentrations (for animals of the same age) of  $\Sigma$ PCB,  $\Sigma$ DDT,  $\Sigma$ CHB and  $\Sigma$ CHLORD in blubber samples of female white whales from West Greenland.



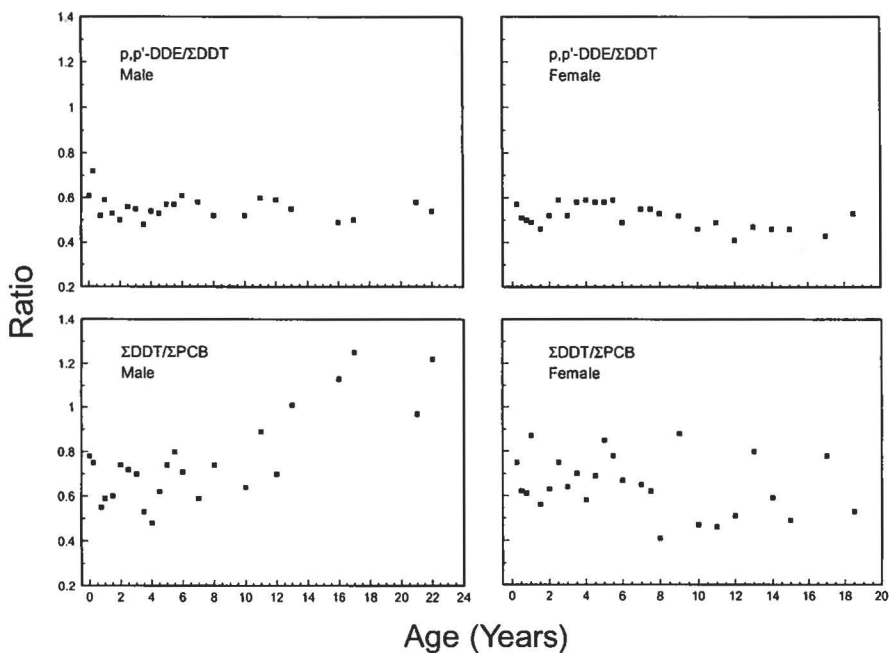


Fig. 4. Relationships between age and p,p'-DDE/ $\Sigma$ DDT and  $\Sigma$ DDT/ $\Sigma$ PCB in blubber samples of male and female white whales from West Greenland.

in blubber, the p,p'-DDE/ $\Sigma$ DDT and  $\Sigma$ DDT/ $\Sigma$ PCB concentration ratios were strongly positively and moderately negatively correlated, respectively, with age in females. No significant correlations were observed with skin samples.

Results from ANCOVA and Tukey's test, used to compare mean concentrations of selected organochlorine contaminants in female and male blubber samples with age group, are shown in Tables 6 and 7, respectively. Generally, mean concentrations in females 8 years and older (groups 5 and 6) were significantly ( $P < 0.05$ ) lower than those in most younger animals. In addition, mean concentrations in females aged 4.5 to 7.5 years (group 4) were

significantly lower ( $P < 0.05$ ) than those observed in animals 2.5 years and younger (groups 1 and 2). For  $\gamma$ -HCH,  $\Sigma$ HCH,  $\Sigma$ CBz and  $\Sigma$ CHB, females aged 3.0 to 4.0 years (group 3) had significantly lower mean concentrations than did those which were 1 year of age and younger (group 1). In males, few contaminants showed any significant differences in mean concentration with age group. When observed, however, differences between the youngest and oldest animals predominated.

Pattern similarities and differences between the mean concentrations of 91 individual PCB and organochlorine contaminants in blubber samples with age group (Table 1) and sex were examined using PCA. Using cross vali-

Table 4. Correlations of organochlorine concentrations (wet weight) with age and length in white whale blubber samples from West Greenland.

Sex	N	Factor	DIELD	MIREX	$\Sigma$ CHB	$\Sigma$ DDT	$\Sigma$ CHLORD	$\Sigma$ HCH	$\Sigma$ CBz	$\Sigma$ PCB
M	71	Age	-0.325*	0.461*	-0.272	0.108	-0.254	-0.473*	-0.213	-0.136
		Length	-0.358*	0.485*	-0.221	0.105	-0.217	-0.544*	-0.252	-0.122
F	67	Age	-0.699*	-0.118	-0.720*	-0.545*	-0.703*	-0.716*	-0.666*	-0.658*
		Length	-0.613*	-0.208	-0.606*	-0.490*	-0.586*	-0.596*	-0.589*	-0.525*

Sex	N	Factor	$\Sigma$ TRI	$\Sigma$ TETRA	$\Sigma$ PENTA	$\Sigma$ HEXA	$\Sigma$ HEPTA	$\Sigma$ OCTA	DDE/ $\Sigma$ DDT	$\Sigma$ DDT/ $\Sigma$ PCB
M	71	Age	-0.419*	-0.093	0.259	0.008	0.198	0.602*	-0.008	0.528*
		Length	-0.439*	-0.100	0.281	0.030	0.269	0.579*	-0.036	0.454*
F	67	Age	-0.625*	-0.663*	-0.651*	-0.601*	-0.517*	-0.183	-0.417*	-0.099
		Length	-0.582*	-0.545*	-0.484*	-0.485*	-0.414*	-0.160	-0.295	-0.204

\* Correlation coefficient significant at  $P < 0.01$

Table 5. Correlations of organochlorine concentrations (wet weight) with age (newborn to 5 years) in white whale kidney and skin samples from West Greenland.

Sample	Sex	N	DIELD	ΣCHB	ΣDDT	ΣCHLORD	ΣHCH	ΣCBz	ΣPCB	ΣTRI
Skin	M	10	-0.156	-0.179	-0.117	-0.161	-0.214	-0.222	-0.387	-0.110
	F	9	-0.079	0.075	-0.015	-0.083	-0.473	-0.426	-0.095	-0.653
Kidney	M	10	-0.743*	-0.379	-0.686	-0.595	-0.575	-0.573	-0.651	-0.151
	F	10	-0.485	-0.604	-0.591	-0.523	-0.621	-0.760*	-0.524	-0.678*

Sample	Sex	N	ΣTETRA	ΣPENTA	ΣHEXA	ΣHEPTA	ΣOCTA	DDE/ΣDDT	ΣDDT/ΣPCB
Skin	M	10	-0.439	-0.442	-0.335	-0.029	0.143	-0.481	0.361
	F	9	-0.223	-0.112	0.073	0.052	0.441	0.274	0.047
Kidney	M	10	-0.601	-0.627	-0.662*	-0.719*	-0.596	-0.039	-0.009
	F	10	-0.567	-0.567	-0.424	-0.424	-0.417	0.717*	-0.560

\* Correlation coefficient significant at P < 0.05

dation, it was determined that the first and second principal components accounted for 38.7 and 27.4%, respectively, of the total variability in the data set. The most notable feature in the resulting score plot (Fig. 5A) is the alignment of the female and male age groups along the first and second principal component axis, respectively. Two distinct clusters are apparent. The first consists of males aged 4 years and younger (groups 1–3) and females aged 1.5 to 4 years (groups 2 and 3). The second cluster consists of males and females aged 4.5 to 7.5 years (group 4). For animals 8 years of age and older (groups 5

and 6) different contaminant profiles are evident in each age group as well as with sex. These results correspond well with those presented earlier (Tukey's test). The importance of individual contaminants in determining the positions of age groups and clusters of age groups along the principal component axes is partially explained by the principal component loading plots (Fig. 5B-D). The loading plot depicting individual OC's only (Fig. 5B) shows that males 8 years and older (groups 5 and 6) are characterized by higher proportions of DDT related compounds, some chlordane-related compounds and mirex. In fe-

Table 6. Comparison between mean concentrations of selected organochlorine contaminants in female blubber samples with age group<sup>†</sup> (comparison significant at P < 0.05).

Residue	F Value	R <sup>2</sup>	Significantly different age groups
α-HCH	6.20	0.44	GR5<2; GR1>6,5,4,3
β-HCH	9.39	0.54	GR6<4,3,2,1; GR5<4,3,2,1; GR4<2
γ-HCH	10.42	0.57	GR6<3,1; GR5<3,2,1; GR4<2,1; GR3<1
OXYCHLORD	7.90	0.50	GR6<3,2,1; GR5<4,3,2,1; GR4<1
<i>l</i> -CHLORD	3.39	0.30	GR5<2,1; GR4<1
<i>c</i> -CHLORD	4.47	0.37	GR1>6,5,4,3,2
<i>l</i> -NONA	9.12	0.53	GR6<4,3,2,1; GR5<3,2,1
<i>c</i> -NONA	15.62	0.66	GR6<3,2,1; GR5<4,3,2,1; GR4<3,2,1
DIELD	10.84	0.58	GR6<3,2,1; GR5<4,3,2,1; GR4<2,1
<i>p,p'</i> -DDE	4.86	0.38	GR6<3,2; GR5<4,3,2,1
ΣCHLORD	10.49	0.57	GR6<3,2,1; GR5<4,3,2,1; GR4<1
ΣHCH	11.62	0.59	GR6<3,2,1; GR5<4,3,2,1; GR4<2,1; GR3<1
ΣCBz	12.60	0.61	GR6<3,2,1; GR5<3,2,1; GR4<2,1; GR3<1
ΣCHB	13.16	0.62	GR6<3,2,1; GR5<4,3,2,1; GR4<2,1; GR3<1
ΣDDT	5.43	0.40	GR6<3,2,1; GR5<4,3,2,1
ΣPCB	9.16	0.53	GR6<4,3,2,1; GR5<4,3,2,1
ΣTRI	8.16	0.51	GR6<2,1; GR5<3,2,1; GR4<2,1
ΣTETRA	7.53	0.49	GR6<3,2,1; GR5<4,3,2,1
ΣPENTA	9.78	0.55	GR6<3,2,1; GR5<4,3,2,1; GR4<3
ΣHEXA	7.60	0.49	GR6<4,3,2,1; GR5<4,3,2,1
ΣHEPTA	5.55	0.41	GR6<3,2; GR5<4,3,2
ΣOCTA	2.46	0.23	GR5<3
DDE/ΣDDT	4.34	0.35	GR6<4,3; GR5<4,3

<sup>†</sup>For group definition see Table 1.



Table 7. Comparison between mean concentrations of selected organochlorine contaminants in male blubber samples with age group\* (comparison significant at P < 0.05).

Residue	F Value	R <sup>2</sup>	Significantly different age groups
β-HCH	4.95	0.40	GR6<2,1; GR5<2,1; GR4<1
γ-HCH	4.87	0.40	GR5<2,1; GR4<2,1
c-CHLORD	2.63	0.26	GR6>4,3,2
c-NONA	5.70	0.43	GR5<1; GR2>6,5,4
p,p'-DDT	3.42	0.32	GR6>5,4,3,2,1
MIREX	3.78	0.34	GR6>4,3,2,1
ΣHCH	4.15	0.36	GR5<2,1; GR4<1
ΣTRI	3.40	0.31	GR1>6,5,4
ΣOCTA	5.93	0.44	GR6>4,3,2,1; GR5>1
ΣDDT/ΣPCB	4.34	0.37	GR6>4,3,2,1

\*For group definition see Table 1.

males o,p'-DDT, p,p'-DDT, p,p'-DDD and mirex are predominant in the older animals while higher proportions of p,p'-DDE are found in animals aged 7.5 years and younger (group 1-4). Females 8 years and older (groups 5 and 6) are distinguished from younger females and older males (> 4 years) by greater loadings of higher chlorinated OC's (ΣCHB, *cis*- and *trans*-chlordane and

OCS) as well as 1,2,3,4- and 1,2,4,5-T<sub>4</sub>CBz, P<sub>5</sub>CBz and α-HCH. In males and females OC's such as *cis*- and *trans*-nonachlor, β- and γ-HCH, HCBz and dieldrin are most predominant in animals younger than 7.5 years (groups 14).

Five distinct clusters of PCB's are observed in the loading plots in Figs. 5C and 5D. Moving clockwise,

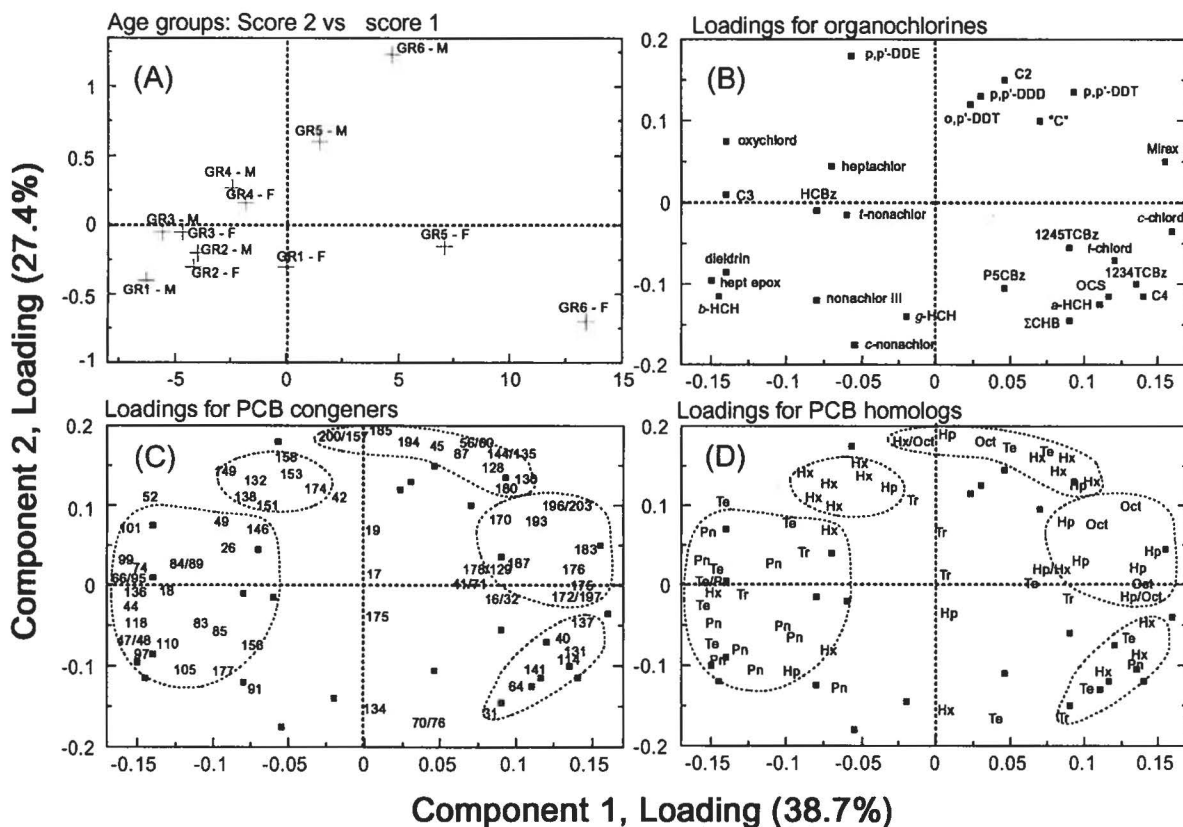


Fig. 5. (A) Score plot of principal components 1 and 2 for 91 individual OC's and PCB contaminants in the blubber of white whales from West Greenland showing similarities and differences between age group and sex. Loading plots for data in (A) showing (B) OC's (C) PCB congeners and (D) PCB homologs. Generally, the higher chlorinated OC's and PCB's are more predominant in male and female animals above the age of 15 years (group 6).



Table 8. Concentrations (mean  $\pm$  standard deviations) of major organochlorines in blubber from Greenland and Canadian waters ( $\mu\text{g/g}$  wet weight).

Location	Sex	Age (years)	N	$\Sigma\text{PCB}$	$\Sigma\text{DDT}$	$\Sigma\text{CHB}$	$\Sigma\text{CHLORD}$
Nuussuaq/ Disko Bugt*	M	5.2 $\pm$ 4.7	71	5.38 $\pm$ 2.27	4.06 $\pm$ 2.50	3.69 $\pm$ 1.46	2.41 $\pm$ 1.08
	F	6.3 $\pm$ 4.6	67	3.74 $\pm$ 2.31	2.69 $\pm$ 1.94	3.01 $\pm$ 1.62	1.79 $\pm$ 1.11
Jones Sound	M	4.4 $\pm$ 2.2	8	2.53 $\pm$ 0.57	1.96 $\pm$ 3.20	4.25 $\pm$ 1.02	1.87 $\pm$ 0.44
	F	4.6 $\pm$ 2.9	7	2.46 $\pm$ 1.98	2.19 $\pm$ 1.69	3.74 $\pm$ 2.12	1.84 $\pm$ 1.13
Cumberland Sound	M	7.3 $\pm$ 6.5	6	4.91 $\pm$ 0.25	6.83 $\pm$ 1.89	5.78 $\pm$ 5.39	2.38 $\pm$ 0.40
	F	8.1 $\pm$ 7.3	6	1.15 $\pm$ 0.41	0.93 $\pm$ 0.55	1.77 $\pm$ 1.76	0.62 $\pm$ 0.15
West Hudson Bay	M	13.0 $\pm$ 4.8	4	3.12 $\pm$ 0.34	3.13 $\pm$ 0.20	5.10 $\pm$ 0.42	2.33 $\pm$ 0.26
	F	10.3 $\pm$ 4.1	4	0.96 $\pm$ 1.00	0.85 $\pm$ 0.96	1.77 $\pm$ 0.41	0.85 $\pm$ 0.80
East Hudson Bay	M	12.0 $\pm$ 9.5	8	2.77 $\pm$ 0.51	2.27 $\pm$ 0.68	4.13 $\pm$ 0.82	1.86 $\pm$ 0.35
	F	11.9 $\pm$ 6.4	8	1.23 $\pm$ 0.84	0.98 $\pm$ 0.73	1.99 $\pm$ 1.10	0.87 $\pm$ 0.58

\* Preliminary statistical results indicated that the mean concentrations of all major organochlorine groups were not significantly different between Nuussuaq and Disko groups for males or females (using the Student t-test at  $P < 0.05$ ). Results from both regions were therefore combined.

cluster 1 consists primarily of tetra and pentachlorobiphenyl congeners which are predominant in males less than 7.5 years of age (groups 1–4) and in females 1.5 to 7.5 years old (groups 2–4). Cluster 2 consists almost exclusively of hexachlorobiphenyl congeners which are most significant in males 4.5 to 15 years old (groups 4 and 5) and in females 7.5 years old and younger (groups 1–4). Cluster 3 is comprised of a mixture of tetra- through to octachlorinated PCB congeners which are predominant in males older than 15 years (group 6) and in females 1 year and younger (group 1) and 8 years and older (groups 5 and 6). Cluster 4 consists of hepta- and octachlorinated PCB congeners which are predominant in older females (groups 5 and 6) and in males aged 1.5 to 15 years (groups 2–5). Finally, cluster 5 consists of a mixture of tri- through to hexachlorinated PCB's. This group of congeners is most significant in females 8 years of age and older (groups 5 and 6) and in males 2.5 years old and younger (groups 1 and 2).

## Discussion

### Geographical variation in element concentrations

The major organochlorine contaminants in all white whale blubber samples were  $\Sigma\text{PCB}$ ,  $\Sigma\text{CHB}$ ,  $\Sigma\text{DDT}$  (mainly p,p'-DDE) and  $\Sigma\text{CHLORD}$  (mainly transnonachlor and oxychlorane).  $\Sigma\text{HCH}$ , chlorobenzenes ( $\Sigma\text{CBz}$ ; tetra-, penta- and hexachlorobenzene) and dieldrin were present at lower concentrations than the four major groups. The results (mean  $\pm$  standard deviation) for  $\Sigma\text{CHLORD}$ ,  $\Sigma\text{DDT}$ ,  $\Sigma\text{PCB}$  and  $\Sigma\text{CHB}$  are presented in Table 8 along with published results from other locations (Muir *et al.* 1990). Mean concentrations of  $\Sigma\text{PCB}$  and

$\Sigma\text{DDT}$  in males from Nuussuaq and Disko Bugt were significantly higher than those in males from Jones Sound and East and West Hudson Bay (Student's t-test at  $P < 0.01$ ) but were not statistically different from levels in Cumberland Sound animals. This may be partly attributed to differences in dietary exposure to organochlorines between whales inhabiting the Davis Strait/Baffin Bay area and those inhabiting the generally shallower waters of Hudson Bay and Hudson Strait. Greenland halibut (*Reinhardtius hippoglossoides*), for example, which are important prey for white whales in West Greenland (Degerbøl & Nielsen 1930, Kleinenberg *et al.* 1969), are widely distributed from the Gulf of St. Lawrence to Baffin Bay but are not found in the shallower waters of Hudson Strait. In females significantly higher levels ( $P < 0.01$ ) of  $\Sigma\text{PCB}$ ,  $\Sigma\text{DDT}$  and  $\Sigma\text{CHLORD}$  were observed in animals from Nuussuaq and Disko Bugt compared to those from Cumberland Sound and Hudson Bay. Although dietary exposure may account for some of the variation among stocks, decreasing levels of contamination with age, due to lactational transfer to offspring, is most likely the major contributing factor. Animals from Cumberland Sound and Hudson Bay were older, on average, than those from Nuussuaq and Disko Bugt.

### Transference of contaminants to offspring

With the exception of animals from Jones Sound (which were all juveniles), mean concentrations of  $\Sigma\text{DDT}$  and  $\Sigma\text{PCB}$  were significantly lower in females than in males (t-test at  $P < 0.01$ ). This difference in tissue burden is the result primarily of lactational transfer of persistent organochlorines to offspring and, secondarily of placental transport from female lipid tissue to foetus (Addison & Brodie 1977, Duinker & Hildebrand 1979, Reijnders

Table 9. Mean, ranges and male-female differences in the concentrations ( $\mu\text{g/g}$  wet weight) of  $\Sigma\text{PCB}$  and p,p'-DDE in marine mammals.

Species	Sex	N	Concentration <sup>†</sup> ( $\mu\text{g/g}$ wet weight)		Male-Female differences <sup>‡</sup>		Lactation period (months)
			$\Sigma\text{PCB}$	p,p'-DDE	$\Sigma\text{PCB}$	p,p'-DDE	
Baird's beaked whale* <i>Berardius bairdii</i>	M	13	3.6 [2.0–5.1]	14 [9.5–19]	55	52	< 6
	F	7	2.0 [1.5–2.8]	7.8 [4.1–17]			
Minke whale* <i>Balaenoptera acutorostrata</i>	M	10	0.02 [0.01–0.03]	0.09 [0.08–0.14]	40	43	3–6
	F	11	0.008 [0.003–0.02]	0.04 [0.01–0.08]			
White whale <i>Delphinapterus leucas</i>	M	30	5.0 [1.5–8.7]	2.4 [0.72–6.0]	46	40	< (12) <sup>§</sup>
	F	36	2.3 [0.47–8.5]	0.96 [0.01–4.2]			
Dall's porpoise* <i>Phocoenoides dalli</i>	M	14	12 [7.1–16]	12 [6.6–15]	23	34	8–18
	F	13	2.8 [1.0–4.0]	4.1 [1.3–7.2]			
Striped dolphin* <i>Stenella coeruleoalba</i>	M	21	32 [15–56]	39 [21–60]	15	11	18
	F	3	4.7 [1.5–6.7]	4.0 [0.79–5.8]			

\* Subramanian *et al.* (1987).

<sup>†</sup> Mean/[range]. Values include those of adult specimens only.

<sup>‡</sup> Percentage of  $\Sigma\text{PCB}$  and p,p'-DDE concentrations in females to those of males.

<sup>§</sup> Calculated lactation period from the relationship between male-female difference and lactation period.

1980, Wagemann & Muir 1984, Subramanian *et al.* 1988, Boon *et al.* 1992). Previous studies have shown that the differences in  $\Sigma\text{PCB}$  and p,p'-DDE levels between adult males and adult females can vary widely among species of cetacean and that the male-female differences are negatively correlated with the length of lactation periods (Subramanian *et al.* 1987). For example the lactation period for every parturition is relatively short (3–6 months) in the minke whale (*Balaenoptera acutorostrata*) (Stewart & Leatherwood 1985). The  $\Sigma\text{PCB}$  and p,p'-DDE levels in females of this species were 40 and 43%, respectively, of those in males of corresponding ages (Subramanian *et al.* 1987). In comparison the lactation period of female striped dolphins (*Stenella coeruleoalba*) is 18 months (Miyazaki 1981) and the male-female differences in  $\Sigma\text{PCB}$  and p,p'-DDE levels were only 15 and 11%, respectively (Subramanian *et al.* 1987). Comparing the male-female percentages of  $\Sigma\text{PCB}$  and p,p'-DDE in white whales (46 and 40%, respectively) with four other cetaceans, a lactation period of less than 1 year could be inferred (Table 9). This estimate is in agreement with that of Vladykov (1944) and Kleinenberg *et al.* (1969), but is much shorter than the more recently suggested 2 year period (Brodie 1971, Sergeant 1973, Burns & Seaman 1985, Heide-Jørgensen & Teilmann 1994).

A lactation period, with the main transference within a year or less is also implied by the relationships between

age and the arithmetic mean concentrations (for animals of the same age) of  $\Sigma\text{PCB}$ ,  $\Sigma\text{DDT}$ ,  $\Sigma\text{CHB}$  and  $\Sigma\text{CHLORD}$  (Figs. 2 and 3). Highest residue levels were observed in 3-month old males and females. Dramatically lower levels were observed in animals 6 and 9 months of age. These results suggest that the diet of calves consists solely of milk during the first 3 months and that milk is possibly supplemented by easily captured prey during the next few months. The young whales would become nutritionally independent from their mothers at about 9 months of age. The observed drop in residue levels might also be attributed partially to the rapid growth (and subsequent dilution of contaminant levels) that occurs during the first year of age (Brodie 1971, Heide-Jørgensen & Teilmann 1994). Residue levels had returned to pre-nursing levels in animals aged 1 to 2 years. After age 5.5 a sharp decrease in the levels of all four major contaminant groups was observed in females compared to only marginal differences in  $\Sigma\text{PCB}$ ,  $\Sigma\text{CHB}$  and  $\Sigma\text{CHLORD}$  concentrations and a substantial increase in -DDT in males. This result is consistent with the age of attainment of sexual maturity in females and commencement of lactation (Brodie 1971, Heide-Jørgensen & Teilmann 1994). If one looks closely at the  $\Sigma\text{PCB}$ ,  $\Sigma\text{DDT}$  and  $\Sigma\text{CHLORD}$  contaminant levels in females 6 years and older, mean concentrations seem to peak at 7, 9, 13 and 17 years, suggesting somehow synchronously parturition at two-year intervals in the early years of an animal's reproduc-

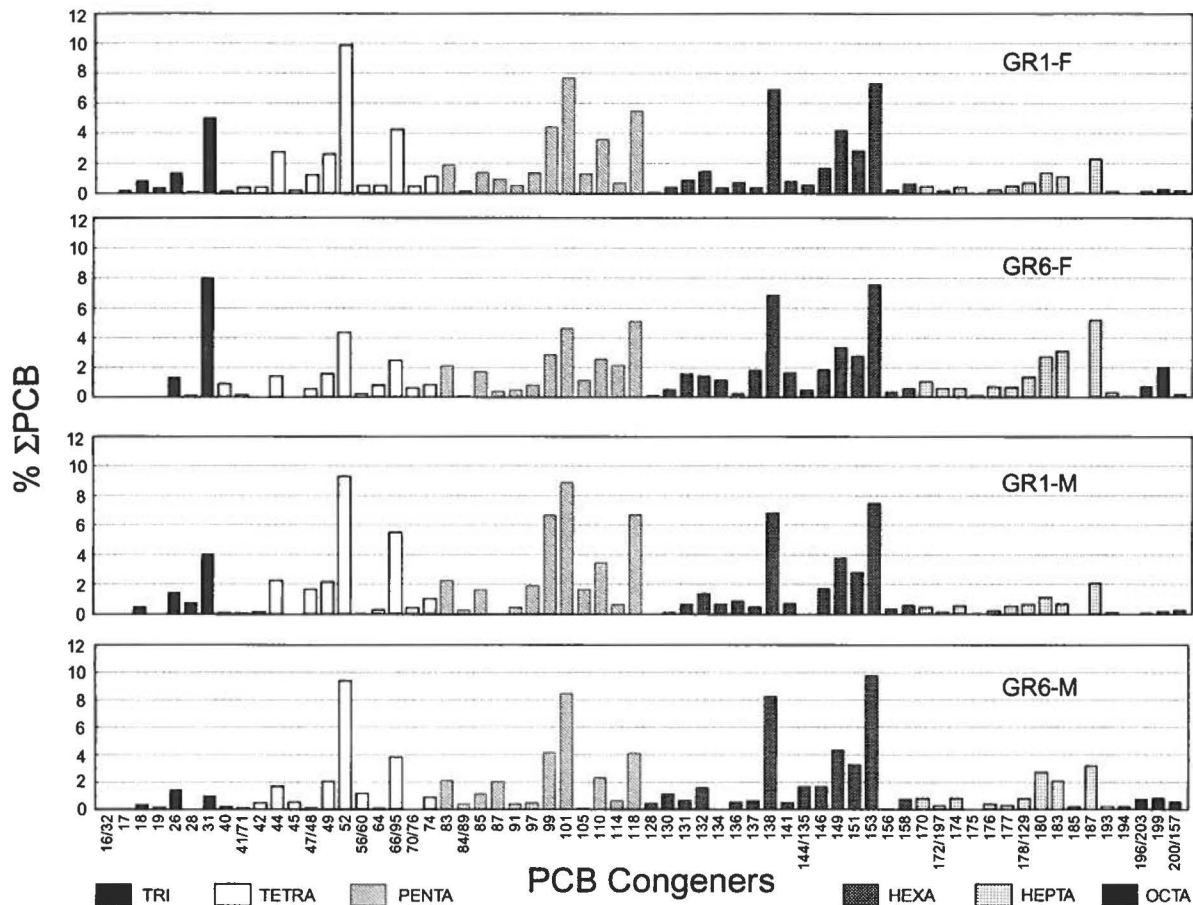


Fig. 6. PCB congeners expressed as a percent of  $\Sigma$ PCB in blubber samples of male and female white whales 1 year old and younger (Group 1) and 15 years and older (Group 6).

tive life and a lengthening resting period between parturitions to 4 years as the animal gets older (for an average parturition frequency overall of approximately 3 years).

Evidence supporting this observation is very weak, however, since no significant differences were observed (t-test at  $P < 0.05$ ) between mean concentration levels at

Table 10. Comparison of ratios of blubber to skin and of blubber to kidney organochlorine concentrations in male white whales from West Greenland, as measured on a wet and lipid weight basis.

Age	Wet weight basis				Lipid weight basis			
	Blubber/Skin		Blubber/Kidney		Blubber/Skin		Blubber/Kidney	
	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ PCB	$\Sigma$ DDT
0.0	10.68	8.29	53.53	58.19	1.24	0.96	1.43	1.56
1.0	15.18	8.38	50.21	51.60	1.11	0.61	2.13	2.19
1.5	30.67	24.07	120.39	66.45	1.45	1.14	3.58	1.98
2.0	12.80	8.19	38.46	39.79	1.06	0.67	0.81	0.83
2.0	6.36	4.46	82.00	88.08	0.86	0.61	1.55	1.67
2.0	9.68	4.24	137.97	106.54	2.42	1.06	4.55	3.51
2.5	14.67	9.71	64.45	54.78	2.07	1.37	2.01	1.71
3.0	12.17	7.31	64.90	58.87	1.55	0.93	2.02	1.83
3.0	12.21	8.70	53.74	43.68	0.94	0.67	2.34	1.90
4.5	11.47	5.14	101.16	93.98	1.27	0.57	1.70	1.58
Mean	13.59	8.85	76.68	66.20	1.40	0.86	2.21	1.88

Table 11. Comparison of ratios of blubber to skin and of blubber to kidney organochlorine concentrations in female white whales from West Greenland, as measured on a wet and lipid weight basis.

Age	Wet weight basis				Lipid weight basis			
	Blubber/Skin		Blubber/Kidney		Blubber/Skin		Blubber/Kidney	
	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ PCB	$\Sigma$ DDT	$\Sigma$ PCB	$\Sigma$ DDT
0.25	8.11	5.82	28.79	21.29	0.98	0.70	0.90	0.67
0.50	16.34	8.59	15.18	11.95	1.26	0.66	1.13	0.89
0.75	10.08	4.76	12.29	8.28	1.18	0.56	0.78	0.53
1.0	10.63	8.51	32.92	29.64	1.11	0.77	1.16	1.04
1.5	8.10	3.92	19.14	12.74	1.33	0.65	1.26	0.84
2.0	25.25	24.04	45.80	47.09	1.51	1.44	1.32	1.35
2.5	6.85	4.39	21.78	19.05	0.96	0.61	0.96	0.84
3.0	—	—	26.95	26.57	—	—	0.92	0.91
3.0	13.50	8.42	50.00	33.27	1.83	1.15	1.96	1.31
3.5	8.15	3.87	19.40	16.21	1.25	0.59	1.14	0.95
Mean	11.89	8.04	27.23	22.61	1.27	0.79	1.27	0.93

yearly intervals (*i.e.* between 6 and 7, 7 and 8 year olds etc.) due to limited numbers of identical year classes.

### Contaminant ratios

The mean *p,p'*-DDE/ $\Sigma$ DDT ratios determined from the blubber of males and females were 0.56–0.05 and 0.52–0.05, respectively. These ratios were comparable to those from Hudson Bay and Jones Sound, but were significantly lower than those from the St. Lawrence River and higher than those from the Beaufort Sea (Muir *et al.* 1990). Ratios for the kidney of males and females are,

respectively, 0.60–0.03 and 0.56–0.05. *p,p'*-DDE/ $\Sigma$ DDT ratios approaching 0.6 imply that no recent significant input of DDT has occurred in Baffin Bay/Davis Strait waters (Aguilar 1984). In young white whales of both sexes (Fig. 4) the *p,p'*-DDE/ $\Sigma$ DDT ratio is highest when they begin to be suckled as newborn calves, drops off at the beginning of the weaning period and increases once again when they are nutritionally independent from their mothers. The higher ratio in suckling calves suggests that proportionally more *p,p'*-DDE is transferred to offspring during lactation. Similar results were observed in a study of fur seals (Kurtz & Kim 1976) where it was found that blubber of nursed pups had a higher *p,p'*-DDE/ $\Sigma$ DDT ratio (90%) than blubber of mothers with yearlings (60%)

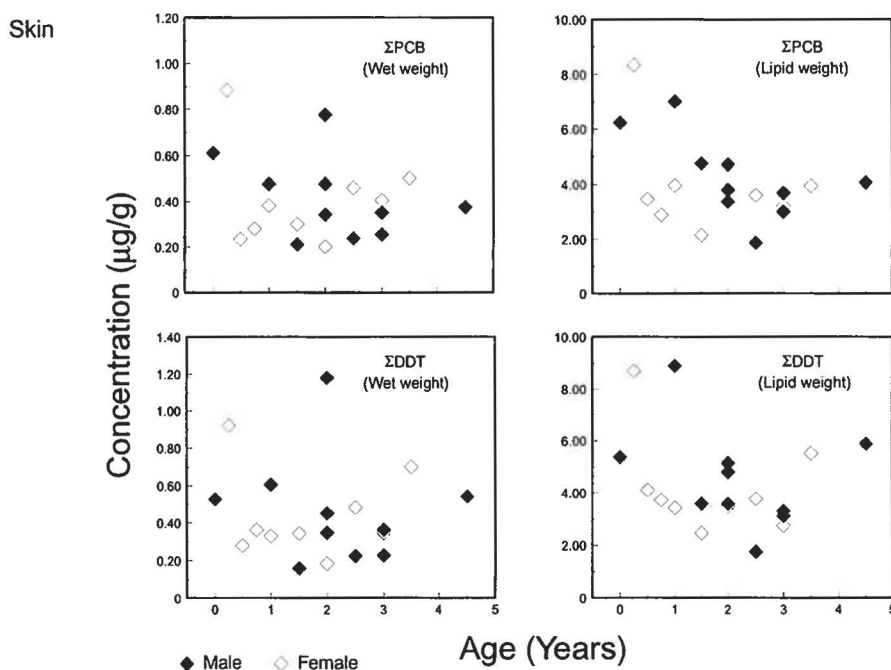
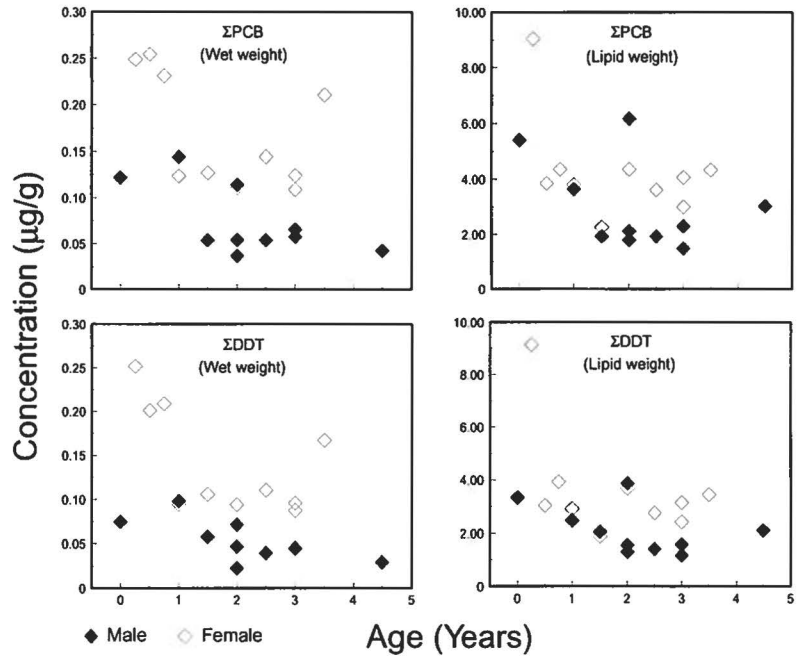


Fig. 7. Relationships between concentrations (wet and lipid weight) of  $\Sigma$ PCB and  $\Sigma$ DDT with age (5 years and younger) in skin samples from male and female white whales from Nuussuaq.

Fig. 8. Relationships between concentrations (wet and lipid weight) of  $\Sigma$ PCB and  $\Sigma$ DDT with age (5 years and younger) in kidney samples from male and female white whales from Nuussuaq.

Kidney



of the same population. Lower water solubility and higher lipophilicity (octanol-water partition coefficient) of a compound should reduce the reproductive-transfer rate to offspring. The above results, therefore, are not surprising since the water solubility of p,p'-DDE is approximately 10 times that of p,p'-DDT (Suntio *et al.* 1988). As young white whales become nutritionally independent and continue to mature, their diet progresses from easily captured prey such as molluscs, annelids and crustacea to larger items higher in the food chain such as squid and various fish species (Kleinenberg *et al.* 1969, Brodie 1971, Heide-Jørgensen & Teilmann 1994). Relative proportions of p,p'-DDE to  $\Sigma$ DDT have been shown to increase from lower to higher trophic levels (Jensen *et al.* 1969, Tanabe *et al.* 1984), suggesting, that the observed rise in levels of p,p'-DDE relative to  $\Sigma$ DDT beginning at the end of the weaning period might in part be due to a change in diet. Consistent with the previous findings of p,p'-DDE/ $\Sigma$ DDT variation in the blubber of fin (*Balaenoptera physalus*) and sei whales (*Balaenoptera borealis*) (Borrell & Aguilar 1987, Aguilar & Borrell 1988) and of St. Lawrence white whales (Martineau *et al.* 1987), an increasing p,p'-DDE/ $\Sigma$ DDT ratio might also be attributed to the corresponding rise in organochlorine levels. Higher tissue pollutant levels induce enzymatic activity and result in proportionally higher abundances of metabolized forms. Once females reach sexual maturity, a gradual decrease in the p,p'-DDE/ $\Sigma$ DDT ratio with age is observed due to preferential excretion of p,p'-DDE during lactation. In males the proportions of p,p'-DDE and  $\Sigma$ DDT remain fairly constant, suggesting a

trend toward equilibrium between the rate of pollutant intake and subsequent degradation of DDT to p,p'-DDE.

The  $\Sigma$ DDT/ $\Sigma$ PCB ratio shows a substantial increasing trend with age (and with pollutant burdens) in males and only a marginal change in females, resulting in higher overall ratios in adult males than in adult females (Fig. 4). As with p,p'-DDE/ $\Sigma$ DDT, this ratio is similar in immature whales of both sexes but differs in adults. As shown earlier in Fig. 3, both  $\Sigma$ DDT and  $\Sigma$ PCB levels decrease substantially with age in sexually mature females. The drop in  $\Sigma$ DDT levels is due primarily to preferential unloading of p,p'-DDE while the decrease in  $\Sigma$ PCB is attributed to preferential elimination of the tri, tetra and pentachlorinated congeners (Tanabe *et al.* 1982, Subramanian *et al.* 1988). While the  $\Sigma$ PCB levels seem to change only marginally with increasing age in adult males, a substantial increase in  $\Sigma$ DDT is observed and is thus accountable for the observed increasing trend in the  $\Sigma$ DDT/ $\Sigma$ PCB ratio.

Among the 64 PCB congeners routinely detected in white whale blubber (Fig. 6) (coeluting congeners are considered as a single component here), the relative proportions of the normally recalcitrant hexa, hepta and octochlorobiphenyls (*i.e.* those having either no adjacent unsubstituted positions or those with unsubstituted *ortho-meta* (*o,m*) positions with more than one *ortho*-chlorine substituent), as well as those which are unsubstituted at the *meta-para* (*m,p*) positions (the above two categories account for all the hexa, hepta and octachlorobiphenyls detected with the exception of PCB 156) were generally either higher or remained unchanged in animals (male

and female) 15 years and older (group 6) compared to that of the corresponding congeners in animals 1 year old and younger (group 1). The reduced capability for biotransformation in the latter category of PCB's is thought to be due to the relatively low activity of cytochrome P450 monooxygenase enzymes which are responsible for CYP2B-type metabolism (Boon *et al.* 1989, Boon *et al.* 1992, Norstrom *et al.* 1992). A different trend is observed for the lower chlorinated congeners (*i.e.* tri, tetra and pentachlorobiphenyls). In this case, especially in females, greater relative proportions are generally observed in the younger animals. In females this is attributed to the preferential elimination of these PCB's during lactation and to the fact that a larger proportion of lesser chlorinated PCB's possess vicinal hydrogens in the *metapara* position in combination with only one *ortho*-chlorine substituent (*i.e.* they are more easily metabolized). In males only the latter explanation is relevant thus accounting for the reduction in observed relative differences compared to the females.

In agreement with our results,  $\Sigma$ PCB,  $\Sigma$ DDT and  $\Sigma$ CHLORD concentrations in the blubber of white whales from the Hudson Bay region were found to be negatively correlated with age and length in females (Muir *et al.* 1990). In contrast to our results however, the levels of these contaminants in the blubber of female white whales from the St. Lawrence estuary were found to be strongly positively correlated with age (Martineau *et al.* 1987, Muir *et al.* 1990). A positive correlation with age in sexually mature females suggests that contaminant intake exceeds contaminant loss during reduced or non-occurring lactation. This result is consistent with the much higher level of exposure of St. Lawrence white whales to PCB's and other pollutants compared to that of arctic white whales (Muir *et al.* 1990) and to possible reproductive dysfunction (Martineau *et al.* 1987, Addison 1989).

## Tissue comparisons

On a wet-weight basis  $\Sigma$ PCB and  $\Sigma$ DDT levels were 4 to 140 times higher in blubber than in skin and kidney. However when expressed on a lipid weight basis the distribution of these contaminants approached proportionality with tissue fat content (Tables 10 and 11). As was also observed in St. Lawrence white whales (Martineau *et al.* 1987)  $\Sigma$ PCB and  $\Sigma$ DDT concentration in males were approximately two times lower in the kidney than in the blubber. Figs. 7 and 8 demonstrate the relationship between age and concentration of  $\Sigma$ PCB and  $\Sigma$ DDT in skin and kidney, respectively, on a wet- and lipid-weight basis for both males and females. The highest contaminant levels observed in kidney and skin of females occurred at approximately 3 months of age. Unfortunately kidney and skin samples from the male of the same age were not analysed. However since  $\Sigma$ PCB and  $\Sigma$ DDT levels were highest in blubber of the same animal

and a certain proportionality has been shown to exist between contaminant levels in blubber and skin and kidney, it is not unreasonable to expect that correspondingly high levels would have been observed in this individual.

## Acknowledgements

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