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Spatial and temporal variations in organochlorine contaminants in the Antarctic minke whale, *Balaenoptera bonaerensis*

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SUMMARY

Concentrations of polychlorinated biphenyls (PCBs), dichlorodiphenyl trichloroethanes and metabolites (DDTs), hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB) and chlordane compounds (CHLs) were determined in the blubber of mature males of Antarctic minke whales, *Balaenoptera bonaerensis*, (21 to 25 years) from the Antarctic Areas IV and V. The ranges of concentrations for each compound were, in ng/g fat wt: PCBs, 7.7-89; DDTs, 29-340; HCHs, 0.20-4.3; HCB, 75-430; CHLs, 10-120. Geographical differences of organochlorines in whales were observed in PCB and HCH levels between 1993/94-1997/98 in Area IV and 1994/95-1998/99 in Area V, and PCB and CHL levels between 1999/00-2003/04 in Area IV and 2000/01-2004/05 in Area V. All organochlorine levels in Area V and HCHs and HCB levels in Area IV significantly decreased temporally. HCHs/PCBs ratio decreased by a factor of about ten in a span of 16 years in both Areas IV and V, while temporal trends of DDTs/PCBs, HCB/PCBs and CHLs/PCBs ratios were stable. These results indicate that PCBs, DDTs, HCB and CHLs levels did not vary or slightly decreased in Areas IV and V during the study period. However HCHs levels clearly decreased. Consequently, increase of organochlorines in the Antarctic environment and Antarctic minke whales observed in the 1980s came to an end in the 1990s. The levels of PCBs and DDTs in blubber of Antarctic minke whales were one order of magnitude lower than those of other whales in the mid and low latitude areas of the Northern Hemisphere, and these levels may not have any adverse effect on whale health.

KEY WORDS: ANTARCTIC MINKE WHALE; ORGANOCHLORINE; MONITORING; TEMPORAL TREND

INTRODUCTION

In the 1995/96 research plan of Japan's whale research program under special permit in the Antarctic (JARPA) added the following objective, 'Elucidation of the effect of environmental change on cetaceans' (Government of Japan, 1995). This object was added in response to resolutions adopted by the Commission on research on the environmental and whale stocks and on the promotion of research on the conservation of large whales (IWC, 1995). Given this, the Science Committee held two specialised Workshops, one relating to chemical pollutants and cetaceans, and the other on the potential ecological effects on cetaceans of climate change and ozone depletion (IWC, 1999). Thus, we have given higher priority to studies on chemical pollutant upon environmental change.

Organochlorines (OCs), such as polychlorinated biphenyls (PCBs), dichlorodiphenyl trichloroethanes and metabolites (DDTs), hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB) and chlordane compounds (CHLs), are man-made chemicals and persistent in the environment. Particularly PCBs, DDTs and its metabolites concentrate in animal bodies through food web. Whales are in a high position in the food web of the ocean. Thus they may be suitable as monitors of pollutant levels and their adverse effects (Tanabe *et al.*, 1986; O'Shea and Brownell, 1994). Thus, we have been continuing to monitor organochlorine levels in blubber of Antarctic minke whale, *Balaenoptera bonaerensis* since the beginning of JARPA research.

Aono *et al.* (1996) reported that PCBs residue levels in Antarctic minke whales increased between 1984/85 and 1992/93 seasons, implying continuous discharge of PCBs in the southern hemisphere. Furthermore, Niimi *et al.* (2000) analysed those samples from 1996/97 season, and reported that DDTs levels of Antarctic minke whale had decreased in the early 1990's and PCBs levels decreased in the late 1990's, while they had increased until the mid 1990's.

We analysed organochlorine levels in blubber of mature males of Antarctic minke whale (21-25 years) from Areas IV and V caught during 1987/88 and 2004/05 seasons. In this paper, the yearly changes and spatial differences of organochlorine accumulations in Antarctic minke whales in the JARPA surveys are presented.

MATERIALS AND METHODS

In this study, concentrations of PCBs, DDTs, HCHs, HCB and CHLs in the blubber samples of 90 Antarctic minke whales (mature males: 21-25 years) taken from the Antarctic Areas IV and V during 1987/88 and 2004/05 seasons were determined (Table 1). In the field, blubber samples were collected, and stored at –20°C until analysis.

Organochlorine pesticides and PCBs were determined by a GC-ECD (Hewlett Packard 5890 Series) and by GC-MS (JEOL Ltd., JMS-700; JMS-SX102A). Chemical analysis of the organochlorines was carried out using the standard method described by Environmental Agency of Japan, with some modifications (Japan Environmental Agency, 1998). Concentrations of organochlorines were expressed on a fat weight basis. Accuracy and precision of the methods were confirmed using 'Organics in cod liver oil' (NIST 1588a). Chemical analyses were performed by the Miura Institute of Environmental Science.

The differences of organochlorine concentrations between areas were assessed by Mann-Whitney U test and the temporal trends of organochlorine concentrations were assessed by linear regression analysis (Zar, 1999). A single sample was found to contain high concentrations of HCB (910 ng/g) which was 2.1 times higher than the second highest concentration in the sample population of 90. This value was found to be outlier using the Smirnoff test, and omitted in the statistical analysis. These statistical analyses were executed by SPSS ver.11 for Windows (SPSS Co. Ltd.).

RESULTS AND DISCUSSION

Table 2 shows the concentrations of organochlorines in blubber in Antarctic minke whales from Areas IV and V. The ranges of concentrations for each compound were, in ng/g fat wt: PCBs, 7.7-89; DDTs, 29-340; HCHs, 0.20-4.3; HCB, 75-430; CHLs, 10-120. The patterns were in the order of DDTs> HCB> PCBs> HCHs > CHLs from both the Areas IV and V, while Aono *et al.* (1997) reported this pattern as HCB> DDTs> CHLs> PCBs> HCHs in male Antarctic minke whales from Area V in 1990/91 and 1992/93 seasons.

Geographical difference

Table 3 shows the comparison of organochlorine levels in the blubber of Antarctic minke whales from Areas IV and V. Figure 1 represents organochlorine levels versus the sampling longitude. PCB and HCH levels between 1993/94-1997/98 in Area IV were significantly higher than those of 1994/95-1998/99 in Area V, and PCB and CHL levels between 1999/00-2003/04 in Area IV were significantly higher than those of 2000/01-2004/05 in Area V.

These results are not consistent with those of trace element accumulation. Hepatic mercury and cadmium concentrations in mature males of Antarctic minke whales taken during 1995/96 and 2002/03 significantly increased from Area III to Area VI (Yasunaga *et al.*, 2006). These results raise three possibilities: 1) background levels of PCBs, DDTs and CHLs between Areas IV and V changed; 2) food intake these animals in Areas IV and V changed, and 3) the organochlorines in the blubber concentrated due to decreasing body fat in Area IV. Their trace element accumulation suggested food availability of Antarctic minke whales from Area IV was lower than those form Area V. And also, Konishi *et al.* (2006) reported that blubber thickness, as obesity index, of Antarctic minke whales from Areas IV and V became thick toward the East. On the basis of these results, if it can be change in body fat of Antarctic minke whales may contribute to geographical differences of pollutants in their bodies.

Temporal trend

Figures 2 and 3 show plots of organochlorine concentrations in blubber of Antarctic minke whales from Areas IV and V versus the sampling years. Table 4 shows simple linear correlation coefficients and probability between year and log OC concentrations in blubber from each area ($p<0.05$). All organochlorine levels in Area V

significantly decreased with year, while HCHs and HCB levels in Area IV significantly decreased with year. Such a trend may be considered as due to the changes in background levels, food intake and changes in body fat however a relationship between food intake index and pollutant burden in each compound should be examined in future studies in order to distinguish these factors.

Figure 4 shows organochlorine pesticides standardized with respect to PCBs, which are stable in the environment as well as animal body. In addition, PCBs have declined slowly in the open ocean and the Antarctic (Tanabe *et al.*, 2003) and the influence of changes in the body fat is nullified by standardization with PCBs. HCHs/PCBs ratio decreased by a factor of about ten in a span of 16 years in both Areas IV and V, while temporal trends of DDTs/PCBs, HCB/PCBs and CHLs/PCB were stable. These results indicated that PCBs, DDTs, HCB and CHLs levels were unchanged or slightly decreased in Areas IV and V during the research period. However, only HCHs levels clearly decreased. Consequently, increase of OCs in the Antarctic environment and Antarctic minke whales observed in the 1980s ended in the 1990s. Organochlorine levels in ocean are affected by historical usage and their physicochemical properties. About 10 Mt of HCHs have been released in the environment, especially in the northern hemisphere (Li, 1999). Emission of HCHs increased steadily after their introduction in the 1940s until they reached a peak in the early 1970s in northern hemisphere (Li & Macdonald, 2005). However, the usage history is similar to other OCs, because organochlorine levels generally declined in the 1970s and leveled off in the 1980s as several bans were enforced in developed countries (Weber & Goerke, 2003). Concentrations of PCBs, DDTs and HCHs in northern fur seals from the North Pacific decreased in between the years 1971 and 1990 (Tanabe *et al.*, 1994). While this trend is likely to continue for DDTs in the future, it can be expected that until at least the 2000s, PCB levels will stabilise may be degradation is compensated by new inputs in the northern hemisphere (Borrell and Rejinders, 1999). HCHs levels in the Antarctic decreased during JARPA research period, while slight temporal changes of the other OCs studied in Antarctic minke whales might be attributable to the decrease in food intake.

Comparison of PCBs and DDTs levels in whales around world

Figure 5 shows the preliminary reported levels of PCBs and DDTs in blubber of baleen whales in the world in comparison with our data. PCBs and DDTs levels in blubber of Antarctic minke whales from Areas IV and V were one order of magnitude lower than those of the other whales in the mid and low latitude areas of the northern hemisphere. PCBs levels in blubber of Antarctic minke whales from the Antarctic were lower than those of bowhead whales, while DDTs were higher (O'Hara *et al.*, 1999; Krone *et al.*, 1999).

Kannan *et al.* (2000) reported a threshold concentration, 17,000 ng/g fat wt, for adverse effects from PCB exposure in the blubber of marine mammals. PCB levels in Antarctic minke whales are extremely lower than this level. There were few reports of adverse effects by DDTs in cetaceans around polluted areas.

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Table 1 Sample number and mean body length (m) of mature male (21-25 years) Antarctic minke whales from Areas IV and V

Area	Year	*n*	Average body length (m)
IV	1987/88	5	8.49
	1989/90	5	8.39
	1991/92	5	8.62
	1993/94	5	8.55
	1995/96	5	8.51
	1997/98	5	8.30
	1999/00	5	8.88
	2001/02	5	8.53
	2003/04	5	8.51
	Total	45	8.53
V	1988/89	5	8.50
	1990/91	5	8.52
	1992/93	5	8.61
	1994/95	5	8.37
	1996/97	5	8.37
	1998/99	5	8.41
	2000/01	5	8.62
	2002/03	5	8.66
	2004/05	5	8.44
	Total	45	8.50

Table 2 Organochlorine concentrations (ng/g lipid wt.) in the blubber of mature male (21-25 years) Antarctic minke whales in the Areas IV and V during 1987/88 and 2004/05 seasons

Area	Year	*n*	Fat (%)	PCBs	DDTs	HCHs	HCB	CHLs
IV	1987/88	5	84.9	45	166	2.6	318	55
			(79.3 - 88.7)	(7.7 - 72)	(29 - 290)	(1.1 - 4.3)	(170 - 410)	(15 - 80)
	1989/90	5	71.4	48	157	2.3	276	59
			(65.0 - 78.6)	(29 - 67)	(85 - 240)	(0.80 - 3.8)	(200 - 380)	(30 - 88)
	1991/92	5	67.3	32	103	1.2	202	34
			(50.3 - 82.5)	(16 - 58)	(42 - 160)	(1.0 - 1.7)	(160 - 330)	(21 - 51)
	1993/94	5	66.6	45	155	1.7	290	57
			(50.3 - 83.6)	(20 - 89)	(45 - 340)	(0.90 - 3.8)	(170 - 430)	(21 - 120)
	1995/96	5	68.7	48	152	1.4	276	59
			(52.1 - 82.1)	(25 - 79)	(69 - 280)	(0.70 - 3.1)	(190 - 400)	(30 - 98)
	1997/98	5	64.9	43	144	1.5	250	66
			(52.5 - 75.5)	(32 - 56)	(110 - 190)	(0.50 - 2.6)	(180 - 390)	(41 - 97)
	1999/00	5	68.3	37	124	0.86	208	57
			(53.7 - 74.0)	(30 - 43)	(91 - 170)	(0.50 - 1.3)	(170 - 250)	(45 - 70)
	2001/02	5	66.3	30	99	0.72	187	43
			(58.9 - 78.5)	(17 - 43)	(42 - 140)	(0.20 - 1.5)	(94 - 310)	(18 - 62)
	2003/04	5	75.7	30	88	0.46	287	35
			(68.9 - 80.3)	(21 - 44)	(65 - 145)	(0.01 - 1.2)	(84 - 908)	(23 - 60)
Total		40	70.5	40	132	1.4	255	52
			(50.3 - 88.7)	(7.7 - 89)	(29 - 340)	(0.01 - 4.3)	(84 - 908)	(15 - 120)
V	1988/89	5	82.0	30	114	1.8	226	37
			(77.7 - 87.9)	(23 - 40)	(71 - 150)	(0.6 - 3.2)	(130 - 340)	(27 - 51)
	1990/91	5	70.5	33	118	1.8	224	39
			(61.7 - 80.3)	(20 - 70)	(57 - 260)	(0.6 - 4.1)	(160 - 380)	(19 - 84)
	1992/93	5	62.4	50	181	1.6	264	56
			(44.8 - 78.2)	(22 - 78)	(74 - 300)	(0.4 - 3.8)	(130 - 400)	(25 - 91)
	1994/95	5	69.7	38	136	1.4	278	58
			(55.4 - 77.2)	(26 - 49)	(80 - 180)	(0.4 - 3.0)	(180 - 380)	(34 - 73)
	1996/97	5	69.4	31	130	0.5	208	45
			(45.2 - 83.1)	(18 - 60)	(44 - 300)	(<0.2 - 1.0)	(120 - 370)	(17 - 89)
	1998/99	5	72.0	27	107	0.4	164	44
			(62.6 - 86.2)	(18 - 35)	(50 - 150)	(0.2 - 0.6)	(140 - 190)	(27 - 57)
	2000/01	5	73.3	17	69	0.4	109	28
			(54.6 - 83.8)	(10 - 27)	(30 - 130)	(<0.1 - 0.4)	(75 - 210)	(10 - 60)
	2002/03	5	75.9	24	66	0.8	152	28
			(63.4 - 84.3)	(21 - 26)	(51 - 74)	(0.7 - 0.9)	(110 - 190)	(23 - 32)
	2004/05	5	67.9	28	103	0.8	136	25
			(61.9 - 75.0)	(21 - 43)	(81 - 159)	(<0.2 - 0.8)	(110 - 191)	(19 - 40)
Total		40	71.5	31	114	1.2	196	40
			(44.8 - 87.9)	(10 - 78)	(30 - 300)	(<0.2 - 4.1)	(75 - 400)	(10 - 91)

Table 3 Geographical differences of organochlorine concentrations in the blubber of Antarctic minke whales (males, 21-25 years) in Areas IV and V obtained using Mann-Whitney U test ($p<0.05$)

			PCBs	DDTs	HCHs	HCB	CHLs
1987/88-1991/92	Area IV	Average	42	140	2.0	270	49
		range	(7.7-72)	(29-290)	(0.80-4.3)	(160-410)	(15-88)
		n	15	15	15	15	15
1988/89-1992/93	Area V	Average	38	140	1.7	240	44
		range	(20-78)	(57-300)	(0.40-4.1)	(130-400)	(19-91)
		n	15	15	15	15	15
		p					
1993/94-1997/98	Area IV	Average	45	150	1.5	270	61
		range	(20-89)	(45-340)	(0.050-3.8)	(170-430)	(21-120)
		n	15	15	15	15	15
1994/95-1998/99	Area V	Average	32	120	0.76	220	49
		range	(18-60)	(44-300)	(<0.2-0.90)	(120-380)	(17-89)
		n	15	15	15	15	15
		p	0.023*		0.004**		
1999/2000-2003/04	Area IV	Average	32	100	0.68	180	45
		range	(17-44)	(42-170)	(0.006-1.5)	(84-310)	(18-702)
		n	15	15	15	14	15
2000/01-2004/05	Area V	Average	23	79	0.40	130	27
		range	(9.6-43)	(30-160)	(<0.2-0.90)	(75-210)	(10-60)
		n	15	15	15	15	15
		p	0.002**				0.004**

*: $p<0.05$, **: $p<0.01$, ***: $p<0.001$

Table 4 Simple linear correlation coefficients and probability between year and organochlorine contaminant concentrations (ppm fat wt) in the blubber of Antarctic minke whales

			LN (PCBs conc.)	LN (DDTs conc.)	LN (HCHs conc.)	LN (HCB conc.)	LN (CHLs conc.)
Area IV	Constant	B±SE			233.751±48.808	75.606±19.673	
	Year (1987-2004)	B±SE			-0.121±0.024	-0.039±0.010	
		P value			<0.001	<0.001	
Area V	Constant	B±SE	51.964±23.347	63.539±29.098	307.438±58.501	85.807±21.486	64.195±27.849
	Year (1987-2004)	B±SE	-0.028±0.012	-0.033±0.015	-0.158±0.029	-0.044±0.011	-0.034±0.014
		P value	0.022*	0.029*	<0.001***	<0.001***	0.020*

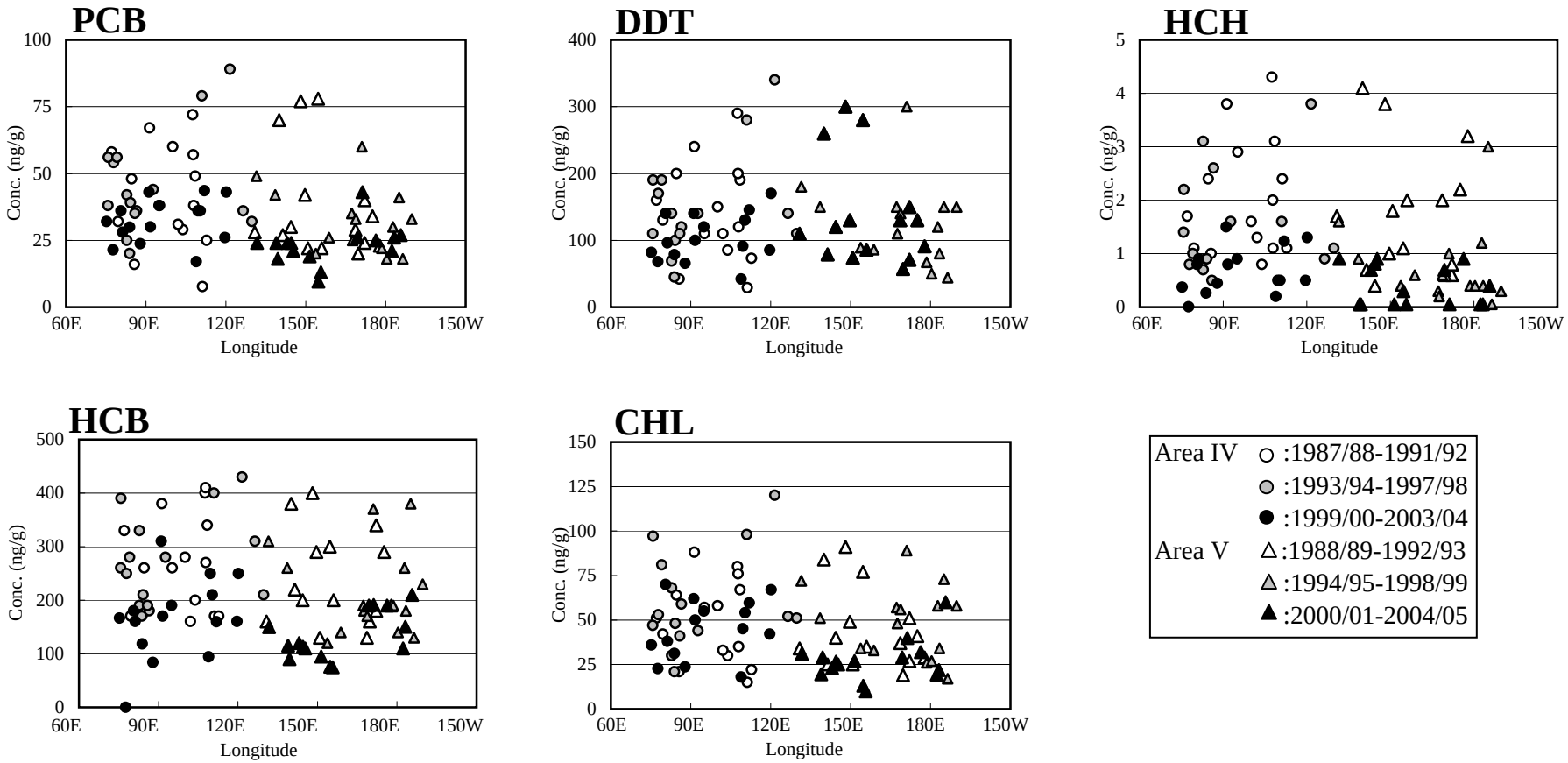

Fig. 1 Relationships PCBs , DDTs, HCHs, HCB and CHLs concentrations (ng/g fat wt) in blubbers of Antarctic minke whales and the sampling longitude

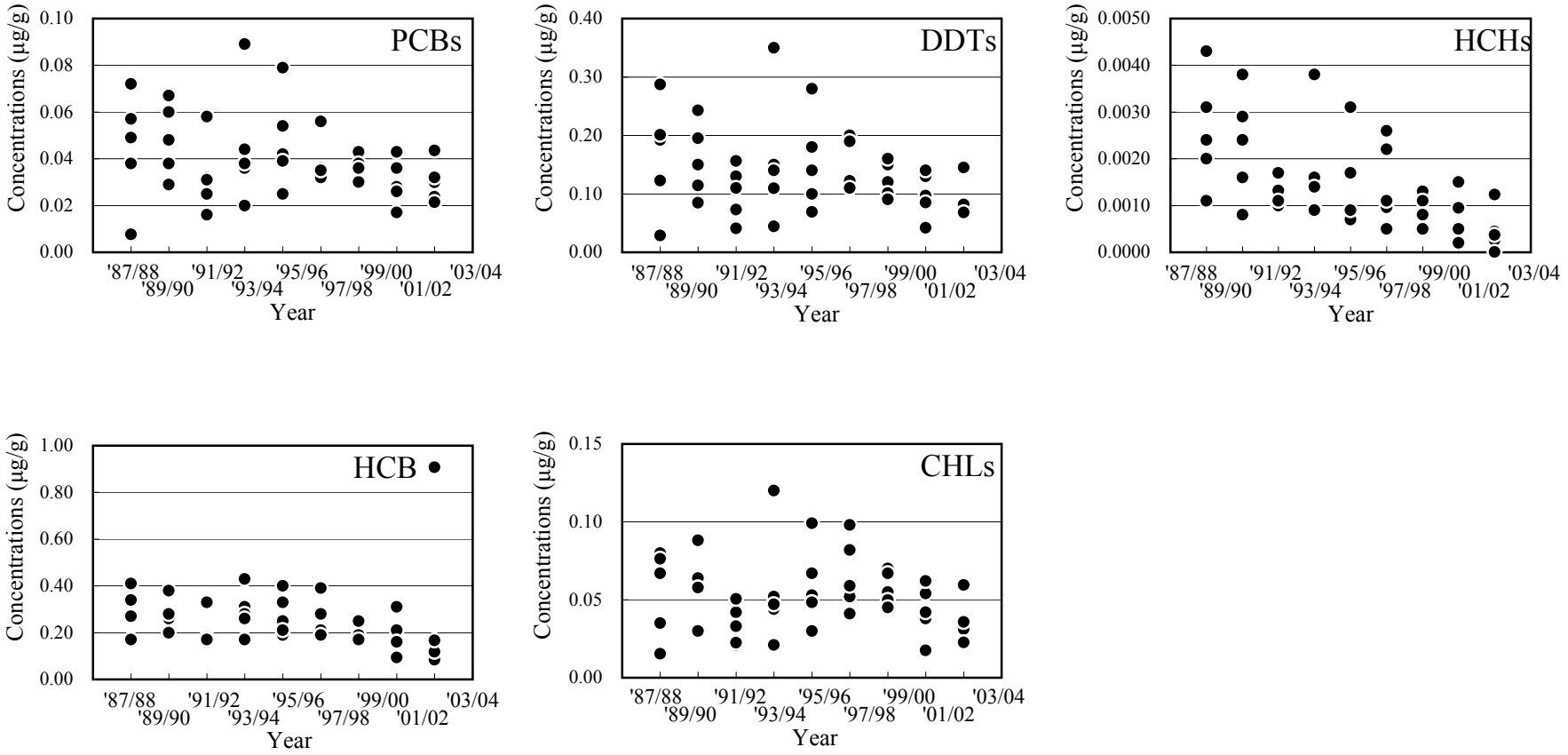

Fig. 2 Temporal trend of organochlorine concentrations (μg/g fat wt.) in the blubber of Antarctic minke whales from Area IV during 1987/88 and 2003/04 seasons

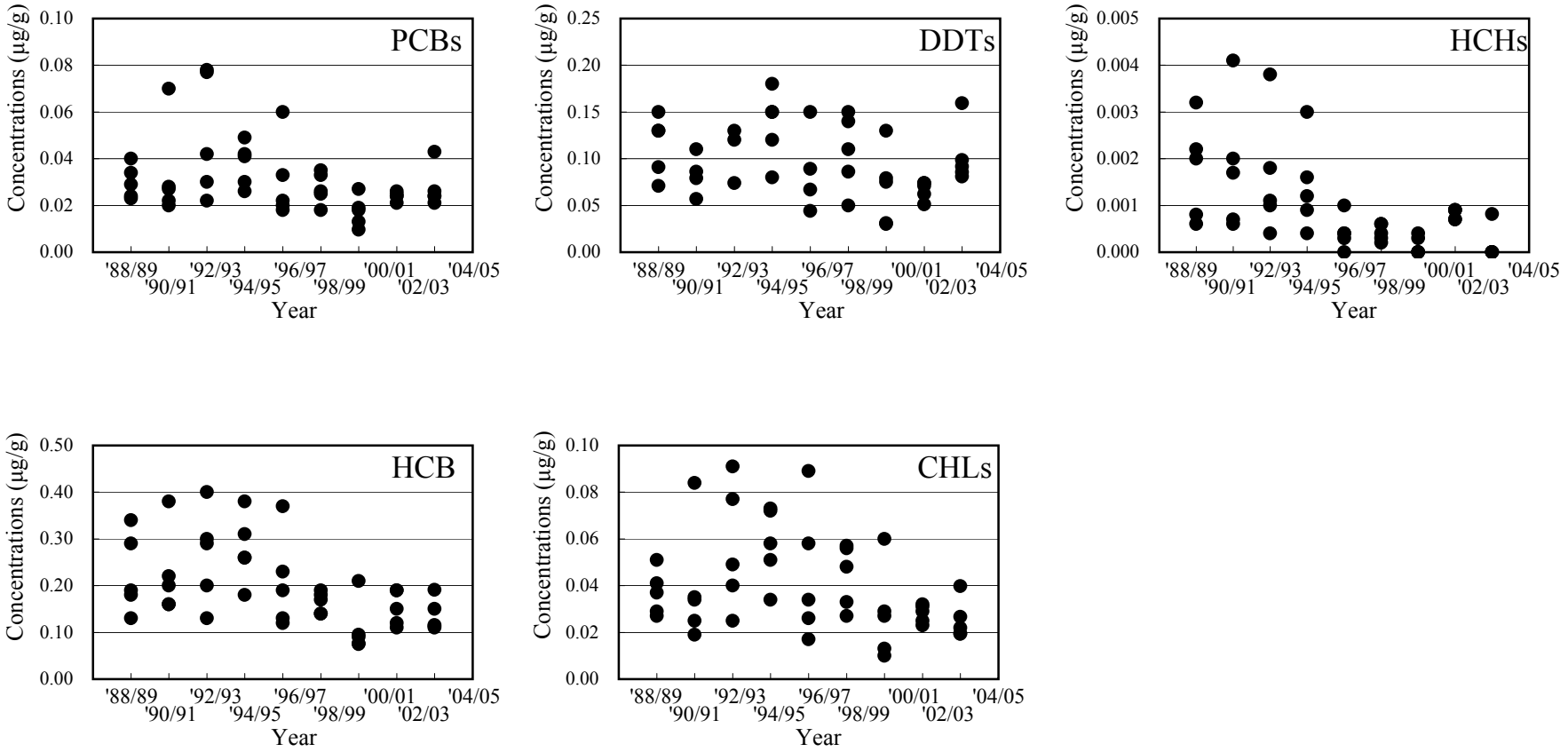

Fig. 3 Temporal trend of organochlorine concentrations (μg/g fat wt.) in the blubber of Antarctic minke whales from Area V during 1988/89 and 2004/05 seasons

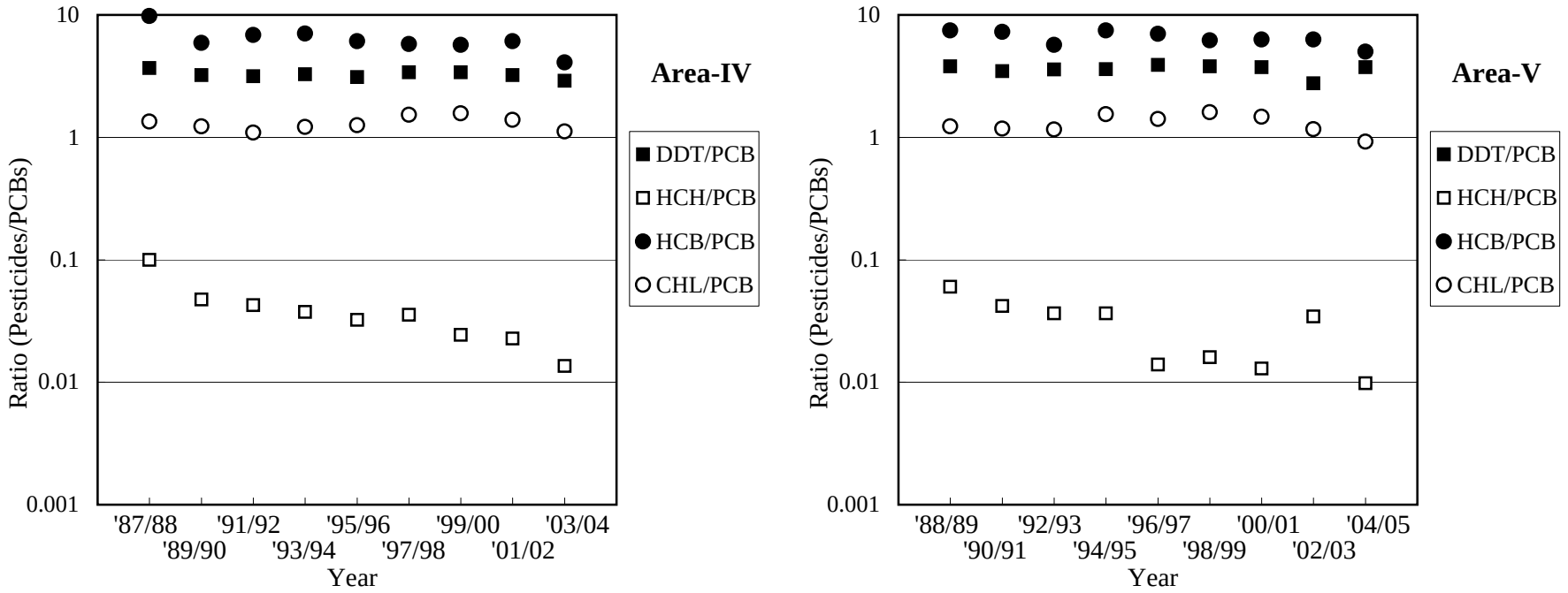

Fig. 4 Ratios of organochlorine pesticides and PCBs in the blubber of Antarctic minke whales from Areas IV and V during 1987/88 and 2004/05 seasons

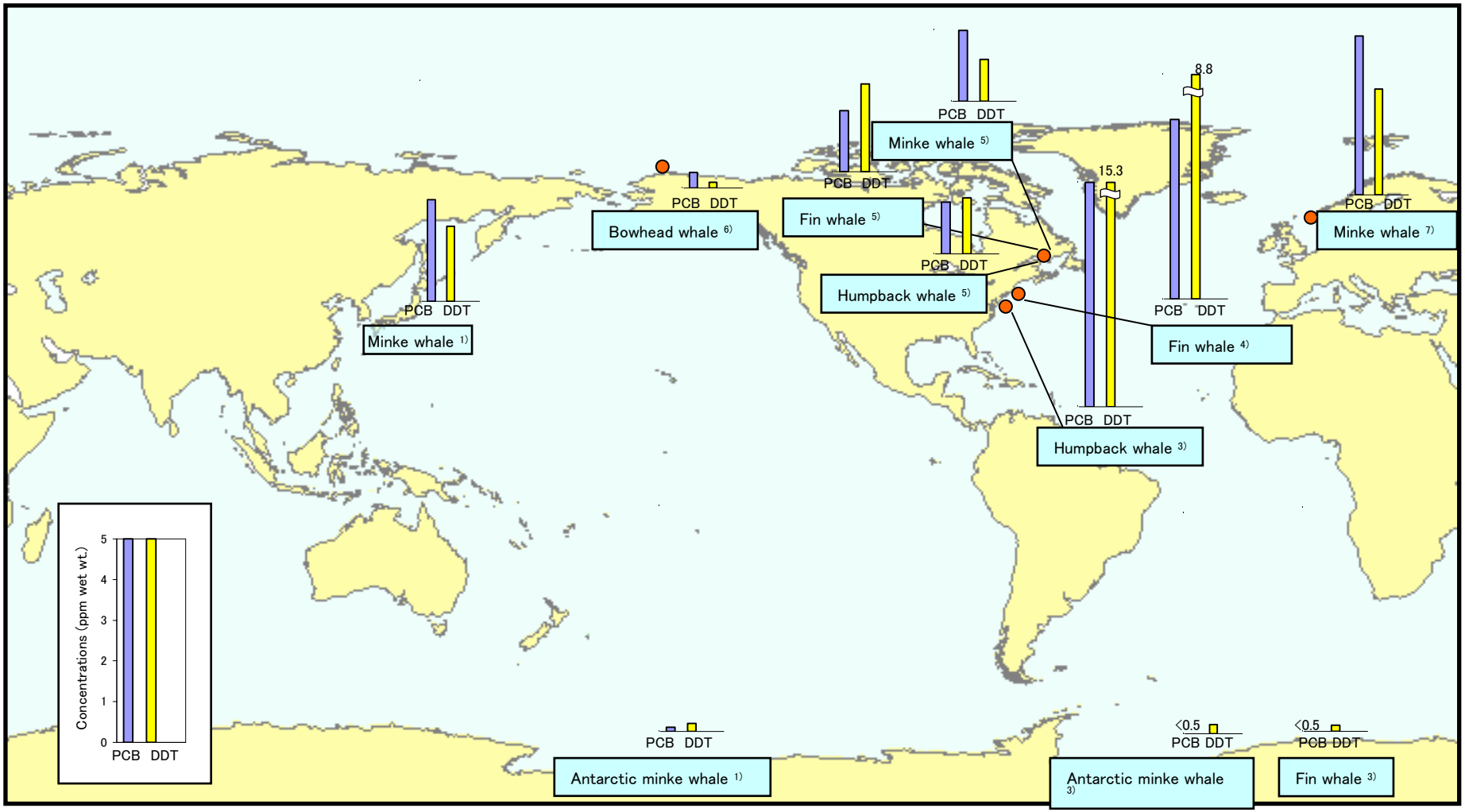

Fig. 5. PCB, DDT Levels in the Blubber of Baleen Whales

1)This study; 2) Henry and Best (1983); 3) Taruski *et al.* (1975) ; 4) Hobbs *et al.* (2001); 5) Gauthier *et al.* (1997); 6) O'Hara *et al.* (1999); 7) Kleivane and Skaare (1998)