Temporal trends and factors affecting PCB levels in baleen whales and environmental samples from the western North Pacific

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ABSTRACT

To examine accumulation features and yearly changes of PCBs in the western North Pacific, PCB levels in samples from surface seawaters, atmospheres and three baleen whales (common minke whales *Balaenoptera acutorostrata*; Bryde's whales *B. edeni*; sei whales *B. borealis*) were measured. The ranges of PCB level in surface water samples were 3.5-11ng/L and 1.5-4.3 ng/L in 2003 and 2005, respectively. In these years PCB levels in seawater decreased with distance from the Japanese coast. The fact that PCB levels in seawaters decreased from coastal to offshore regions was probably due to that PCB has been released from Japan and Eurasia continent. This result is consistent with those of previous studies, such as PCB monitoring in pelagic fish and squid. On the other hand, the trend in air samples was unclear since PCB stability in the atmosphere is easily affected to a significant extent by weather conditions. PCB levels in blubber samples of common minke, Bryde's and sei whales exhibited ranges from 0.13-4.0, 0.04-0.21 and 0.03-0.47 ppm wet wt., respectively. To examine the temporal trend of PCB levels in whales from sub-areas 7, 8 and 9, a multiple regression analysis was conducted having sampling years, sampling longitude, latitude, sampling date, body length and blubber thickness as individual factors. By considering the results of this and previous studies, PCB levels in baleen whales decreased until the end of the decade of the 90's and then it stabilized.

KEYWORDS: COMMON MINKE WHALE; BRYDE'S WHALE; SEI WHALE; NORTH PACIFIC; PCB; SEAWATER; ATMOSPHERE

INTRODUCTION

Pollutants accumulate through the food chain in a marine ecosystem. Because cetaceans are located at the top of the food chain they are frequently used for monitoring the pollutants in the marine environment. Further cetaceans are mobile and long-lived animals, and these characteristics mean that pollutants can be monitored in wide areas and integrated in some way over time. Polychlorinated biphenyls (PCBs) are man-made chemicals and had been widely used in the world in electrical equipments such as transformers and capacitors until 1970s. In spite of the ban of production and usage in 1970s, PCBs are still being released into the environment due to its lipophilic and highly persistent character. Especially, top marine predators such as whales tend to accumulate PCBs in fatty tissues with growth (Tanabe *et al.*, 1981; Connell, 1988). The Japanese Whale Research Programme under Special Permit for North Pacific Whales Phase II (JARPN II) was designed so that samples can be obtained for investigating the quantitative behaviour and fate of the PCBs in the marine environment.

In the western North Pacific, yearly change of PCBs levels in marine mammals have been reported previously. Tanabe *et al.*, (1994) reported a study on temporal variation of PCBs in female northern fur seal. The level showed a maximum in around 1976 and then decreased until 1988. Aono *et al.*, (1997) reported a study of PCB in blubber of common minke whales from the western North Pacific between 1987 and 1994. No trend in the level was observed in this period. Kajiwara *et al.* (2004) reported that the PCB level and compositions were steady-stable in fat tissues of female northern fur seals collected in the period 1972-1998 from the Japanese Pacific coast, suggesting a continuous input of PCBs into the marine environment in significant quantities.

To examine accumulation features and yearly changes of PCBs in the western North Pacific, PCB levels in samples from surface seawaters, atmospheres and three baleen whales (common minke whales *Balaenoptera acutorostrata*; Bryde's whale *B. edeni*; sei whales *B. borealis*), were measured.

MATERIALS AND METHODS

Samples and sampling method

During the 2003 and 2005 JARPN II surveys, air samples were collected by researchers on board the research base (*Nishin-Maru*). Researchers also collected seawater samples on board the dedicatedf sighting vessel *Kyoshin-Maru No.2*. The sampling date and location together with surface temperature are shown in Table 1. A modified version of the method of Iwata *et al*. (1993) was used for sampling air and seawater.

Sampling equipment for air samples was a modification of Iwata *et al.* (1993). Air pump and flow meter were made in IWAKI Co. Ltd. and pre-filter, flow line and absorbent column for collection of OCs were made from polyethylene nonwoven fabric, polyethylene tube and PILEX glass (Asahi Co. Ltd.), respectively. The absorbent column were packed with 5 polyurethane form plugs that were cleaned with acetone in a Soxhlet extractor for more than 7 days until OCs had not been detected in the eluate. The system covered with stainless case was set on the upper deck at about 50 meters above sea level. Sampling equipment for surface seawater was a modification of Iwata *et al*. (1993). The pre-filter and integral flow meters were made in ADVANTEC® and Nitto Seiko Co. Ltd., respectively. The seawater plumbing lines from ship bottom to sampler in *Kyoshin-Maru* were used with stainless tube. Both samplers were made in Miura Institute of Environmental Science (Ehime prefecture, Japan), and also all column were cleaned in it.

Blubber tissues from the common minke whales (Okhotsk Sea-West Pacific stock) from sub-areas 7, 8 and 9, Bryde's whales from sub-area 8 and sei whales from sub-area 9 were collected by JARPN II researchers in the surveys conducted in the period from 2002 to 2007. Table 2 shows the sample sizes and body length of the whales sampled, by species, sub-area and survey. The blubber tissue samples were frozen and shipped to the laboratory and stored -20˚C until chemical analysis.

In order to understand the pattern of accumulation of pollutants in cetaceans, it is essential to consider some biological information from whales. In this study we considered information on the body length and maturity status of male whales. Males of minke, Bryde's and sei whales were defined as sexually mature by testis weight (larger side) of more than 290g, 560g and 1,090g, respectively (Bando *et al.*, unpublished data). Only minke whales from the Okhotsk Sea-West Pacific stock as identified by the microsatellite analysis (Kanda *et al.*, 2009), were used.

Laboratory analysis

The columns with environmental samples (air and sea water) were sent to the Miura Institute of Environmental Science (Ehime, Japan) for PCB analyses. Analyses were performed according to the Analytical Manual for Dioxin in Soils (Japan Environmental Agency, 1998). The blubber samples were sent to the Japan Food Research Laboratories (Tokyo, Japan) for PCB analyses. Analyses were performed according to the public analytical method of Japan (Japan Ministry of Welfare, 1972).

Statistical analysis

The extent of differences in PCB levels among baleen whales and sub-areas was assessed by Mann-Whitney U test (Zar, 1999). In order to examine the yearly trend of PCB levels in blubber of whales, multiple linear regression analyses were conducted (Zar, 1999). The following independent variables: "sampling year" , "sampling date (1 May = day 1)", "sampling latitude", "sampling longitude", "body length" and "blubber thickness", were allowed, and all parameters were normalized. These statistical analyses were executed by SPSS ver.11 for Windows (SPSS Co. Ltd.).

RESULTS AND DISCUSSION

Air and surface seawater

The ranges of PCB level of air samples were ND-22 pg/m^3 and 8-13 pg/m^3 in 2003 and 2005, respectively (Table 1, Fig. 1). Iwata *et al.* (1993) reported a range of 12-390 pg/m^3 for air samples collected in the North Pacific in 1990. They used a similar methodology as that used in the present study. Thus, residue levels of PCBs in atmosphere in 2003 and 2005 might be lower than those in 1990. However, it should be noted that PCB stability in atmosphere is easily affected to a significant extent by weather condition (Montone *et al.*, 2003).

The ranges of PCB level in surface water samples were 3.5-11ng/L and 1.5-4.3 ng/L in 2003 and 2005, respectively (Table 1, Fig. 2). In both years the PCB levels in seawater decreased with distance from the Japanese coast. The fact that PCB levels in seawaters decreased from coastal to offshore region was probably due to that PCB has been released from Japan and Eurasia continent. This result is consistent with those of previous studies, such as PCB monitoring in pelagic fish (Ueno *et al.*, 2003) and squid (Yamada *et al.*, 1997).

Baleen whales

Table 2 shows the PCB level in blubbers of common minke, Bryde's and sei whales. The ranges were 0.13-4.0, 0.04-0.21 and 0.03-0.47 ppm wet wt. in these species, respectively. In sub-area 8 the PCB levels in common minke whales were significantly higher than those in Bryde's whales ($p<0.05$); in sub-area 9 the levels in common minke whales were significantly higher than those in sei whales ($p<0.05$). Differences in feeding habitat and trophic levels of preys might be one of the reasons for the differences. It is known that common minke whales selectively feed on relatively higher trophic organisms, mainly pelagic fish such as Pacific saury and Japanese anchovy, and krills (Tamura and Fujise, 2002), which are different from the prey items of Bryde's and sei whales, which feed mainly on krill and copepod (Tamura *et al.*, 2009).

Plots of PCB levels against sampling year in sub-areas 7, 8 and 9 are shown for common minke whales (Fig. 3a-c), Bryde's (Fig. 3d) and sei whales (Fig. 3e). For common minke whales in sub-area 7, sampling year, body length and sampling date were significantly selected (Table 3) with a rank order of effects of sampling date (B:-0.633) > year (-0.372) > body length (+0.226). For those from sub-area 9, only body length (+) were significantly selected (Table 5). For common minke whales from sub-area 8 (Table 4), Bryde's whales from sub-area 8 (Table 6), sei whales from sub-area 9 (Table 7) no factor was significantly selected in the regression equation. Primary factor to PCB levels in common minke whales from sub-area 7 was sampling date, namely seasons. This phenomenon would reflect the dilution by increasing of fat volume in the whale body. The year would not be important factor for PCB levels in sub-area 7, because the sampling year was selected as secondary factor and the value in the 2003 only was apparently higher than the other years (Table 2 and Fig. 3a). Therefore, PCB levels in baleen whales from the western North Pacific seems to be in a steady state since 2002.

There are some reports of temporal trend of PCB levels in the western North Pacific using marine mammals as biological indicator. For example Tanabe *et al.*, (1994) reported temporal variation of PCBs with a maximum in 1976 and a decreasing trend until 1988 in female northern fur seal. Aono *et al.*, (1997) reported that the PCB levels in blubber of common minke whales from the western North Pacific (offshore) between 1987 (1.5-3.0 ppm wet wt.) and 1994 (0.62-3.1 ppm wet wt.) were stable. Kajiwara *et al.* (2004) reported that PCB concentrations in fat tissues of female northern fur seals collected from off Sanriku coast in the period 1972 - 1998 continually decreased since early 1980s, and their PCB compositions have not changed in all period. This suggest a continuous input of PCBs into the marine environment near the Japanese coast. The PCB levels presented in this study for the period 2002-2007 is lower than those in 1987 and 1994. The PCB levels in marine mammals in coastal Japan had continually decreased until the end of 1990's, and it has stabilized since at least from 2002.

There are a number of advantages of using marine organism, especially higher torophic animals, as biomonitors for residual pollutants rather than environmental samples, such as air, seawater (Vetter *et al.*, 1996). There were few reports of PCB levels in environmental samples from offshore ocean, because the direct measurement of environmental pollutants suffer from the disadvantages of the stability in the preservation until the measuring and risk of treatment contamination due to the extremely lower levels. Predators accumulate pollutants through food chain, then they are useful for the monitoring of marine pollutants. Predator, such as cetacean, are mobile, so pollutants will be picked up from several food items, and they are long-lived, so pollutant burdens may be integrated in some way over time.

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Table 1. Location of atmosphere and sea water samples collected from western North Pacific and levels of PCB.

Location	Date	Site		PCBs
		Started	Finished	
Atmosphere				(pg/m3)
03NP-A1	2003/6/16	39-35N, 157-13E	41-04N, 159-09E	22
03NP-A2	2003/7/28	44-48N, 162-29E	42-39N, 168-13E	ND
03NP-A3	2003/8/10	40-59N, 155-35E	38-23N, 143-06E	ND
05NP-A4	2005/7/7	39-59N 143-05E	37-24N 145-58E	13
05NP-A5	2005/7/16	39-10N 146-06E	41-02N 152-35E	11
05NP-A6	2005/8/15	43-28N 159-55E	39-43N 146-44E	8
Seawater				(ng/L)
03NP-S1	2003/6/17	38-39N, 156-51E	39-30N, 157-29E	7.7
03NP-S2	2003/7/1	44-37N, 157-46E	44-15N, 156-45E	7.2
03NP-S3	2003/7/11	40-11N, 146-54E	40-52N, 150-21E	11
03NP-S4	2003/9/6	46-49N, 158-58E	44-43N, 156-09E	3.5
05NP-S5	2005/7/6	37-40N 146-20E	40-07N 143-06E	2.5
05NP-S6	2005/7/10	39-10N 146-06E	40-22N 150-50E	4.3
05NP-S7	2005/8/25	43-28N 159-55E	43-30N 154-50E	1.5

Table 2 PCB levels (ppm wet) in blubber of common minke , Bryde's and sei whales in the western North Pacific during 2002 and 2007.

species	sub area		body length	2002	2003	2004	2005	2006	2007	total
common minke whal	7	ave±sd	7.48 ± 0.28	0.75 ± 0.31	1.34 ± 0.77	0.42 ± 0.11	0.83 ± 0.44	0.91 ± 0.40	0.76 ± 0.33	0.82 ± 0.44
		ragne	(6.73 - 8.15)	(0.35 - 1.50)	(0.28 - 2.70)	(0.32 - 0.60)	(0.44 - 2.30)	(0.22 - 1.50)	(0.21 - 1.60)	(0.21 - 2.70)
		n	126	35	12	6	17	14	42	126
	8	ave±sd	7.54 ± 0.25	0.91 ± 0.35	1.17 ± 0.85	±	±	1.03 ± 0.76	0.83 ± 0.38	1.04 ± 0.72
		ragne	(6.91 - 8.02)	(0.31 - 1.40)	(0.21 - 4.00)	(-)	(-)	(0.21 - 2.90)	(0.29 - 1.40)	(0.21 - 4.00)
		n	69	8	27			24	10	69
	9	ave±sd	7.51 ± 0.25	0.70 ± 0.24	0.60 ± 0.24	0.45 ± 0.22	0.64 ± 0.44	0.62 ± 0.43	0.76 ± 0.44	0.58 ± 0.31
		ragne	(6.77 - 8.18)	(0.29 - 1.10)	(0.24 - 1.20)	(0.13 - 1.20)	(0.14 - 2.10)	(0.21 - 1.60)	(0.22 - 1.30)	(0.13 - 2.10)
		n	152	24	33	51	24	16	4	152
	total	ave±sd	7.51 ± 0.26	0.75 ± 0.30	0.94 ± 0.70	0.45 ± 0.22	0.71 ± 0.45	0.88 ± 0.61	0.78 ± 0.34	0.76 ± 0.50
		ragne	(6.73 - 8.18)	(0.29 - 1.50)	(0.21 - 4.00)	(0.13 - 1.20)	(0.14 - 2.30)	(0.21 - 2.90)	(0.21 - 1.60)	(0.13 - 4.00)
		n	347	67	72	57	41	54	56	347
Bryde's whale	8	ave±sd	12.54 ± 0.41	0.11 ± 0.06	±	0.13 ± 0.04	±	0.07 ± 0.03	±	0.1 ± 0.05
		ragne	(11.90 - 13.11)	(0.05 - 0.21)	(-)	(0.08 - 0.19)	(-)	(0.04 - 0.11)	(-)	(0.04 - 0.21)
		n	15	5		5		5		15
Sei whale	9	ave±sd	13.68 ± 0.31	0.14 ± 0.18	±	0.12 ± 0.07	±	0.09 ± 0.06	±	0.12 ± 0.11
		ragne	(13.23 - 14.48)	(0.03 - 0.47)	(-)	(0.04 - 0.22)	(-)	(0.04 - 0.19)	(-)	(0.03 - 0.47)
		n	15	5		5		5		15

Table 3 Results of multiple linear regression analyses with "PCB levels in blubbers of common minke whales from sub-areas 7" as the dependent variable.

a) Model of regression

Model	R	R2	R2'
1	0.581	0.337	0.304

b) Analysis of Variance Table

Model	Sum of Squares	DF	Mean Square	F value	P value
Regression	33.132	6	5.522	10.088	0.000
Residual	65.140	119	0.547		
Total	98.273	125			

c) Variables

Model	B	SE	B'	T	P value
Constant	1.190	0.509		2.336	0.021
year	-0.372	0.126	-0.497	-2.955	0.004
date	-0.633	0.154	-0.884	-4.105	0.000
latitude	0.225	0.287	0.131	0.785	0.434
longitude	0.733	0.485	0.163	1.510	0.134
body length	0.226	0.064	0.274	3.521	0.001
blubber thickness	0.082	0.053	0.105	1.882	0.061

Table 4 Results of multiple linear regression analyses with "PCB levels in blubbers of common minke whales from sub-areas 8" as the dependent variable.

a) Model of regression

Model	R	R2	R2'
1	0.371	0.138	0.054

b) Analysis of Variance Table

Model	Sum of Squares	DF	Mean Square	F value	P value
Regression	19.701	6	3.283	1.650	0.149
Residual	123.388	62	1.990		
Total	143.088	68			

c) Variables

Model	B	SE	B'	T	P value
Constant	0.272	0.401		0.679	0.500
year	-0.154	0.177	-0.110	-0.868	0.389
date	-0.713	0.889	-0.317	-0.801	0.426
latitude	0.208	1.082	0.079	0.192	0.848
longitude	-0.379	0.831	-0.058	-0.456	0.650
body length	0.431	0.192	0.277	2.249	0.028
blubber thickness	0.181	0.293	0.084	0.617	0.539

Table 5 Results of multiple linear regression analyses with "PCB levels in blubbers of common minke whales from sub-areas 9" as the dependent variable.

a) Model of regression

Model	R	R2	R2'
1	0.314	0.099	0.061

b) Analysis of Variance Table

Model	Sum of Squares	DF	Mean Square	F value	P value
Regression	5.973	6	0.995	2.644	0.018
Residual	54.583	145	0.376		
Total	60.556	151			

c) Variables

Model	B	SE	B'	T	P value
Constant	-0.268	0.130		-2.060	0.041
year	-0.048	0.077	-0.055	-0.616	0.539
date	0.017	0.141	0.017	0.122	0.903
latitude	-0.159	0.103	-0.217	-1.555	0.122
longitude	0.034	0.122	0.023	0.282	0.779
body length	0.114	0.053	0.173	2.163	0.032
blubber thickness	-0.058	0.066	-0.087	-0.873	0.384

Table 6 Results of multiple linear regression analyses with "PCB levels in blubbers of Bryde's whales from sub-areas 8" as the dependent variable.

a) Model of regression

Model	R	R2	R2'
1	0.706	0.498	0.121

b) Analysis of Variance Table

Model	Sum of Squares	DF	Mean Square	F value	P value
Regression	6.971	6	1.162	1.323	0.348
Residual	7.029	8	0.879		
Total	14.000	14			

c) Variables

Model	B	SE	B'	T	P value
Constant	-1.40E-10	0.242		0.000	1.000
year	-0.527	0.325	-0.527	-1.621	0.144
date	0.929	1.343	0.929	0.691	0.509
latitude	-1.139	1.001	-1.139	-1.138	0.288
longitude	1.315	1.221	1.315	1.077	0.313
body length	0.351	0.270	0.351	1.298	0.230
blubber thickness	0.110	0.297	0.110	0.372	0.719

Table 7 Results of multiple linear regression analyses with "PCB levels in blubbers of sei whales from sub-areas 9" as the dependent variable.

a) Model of regression

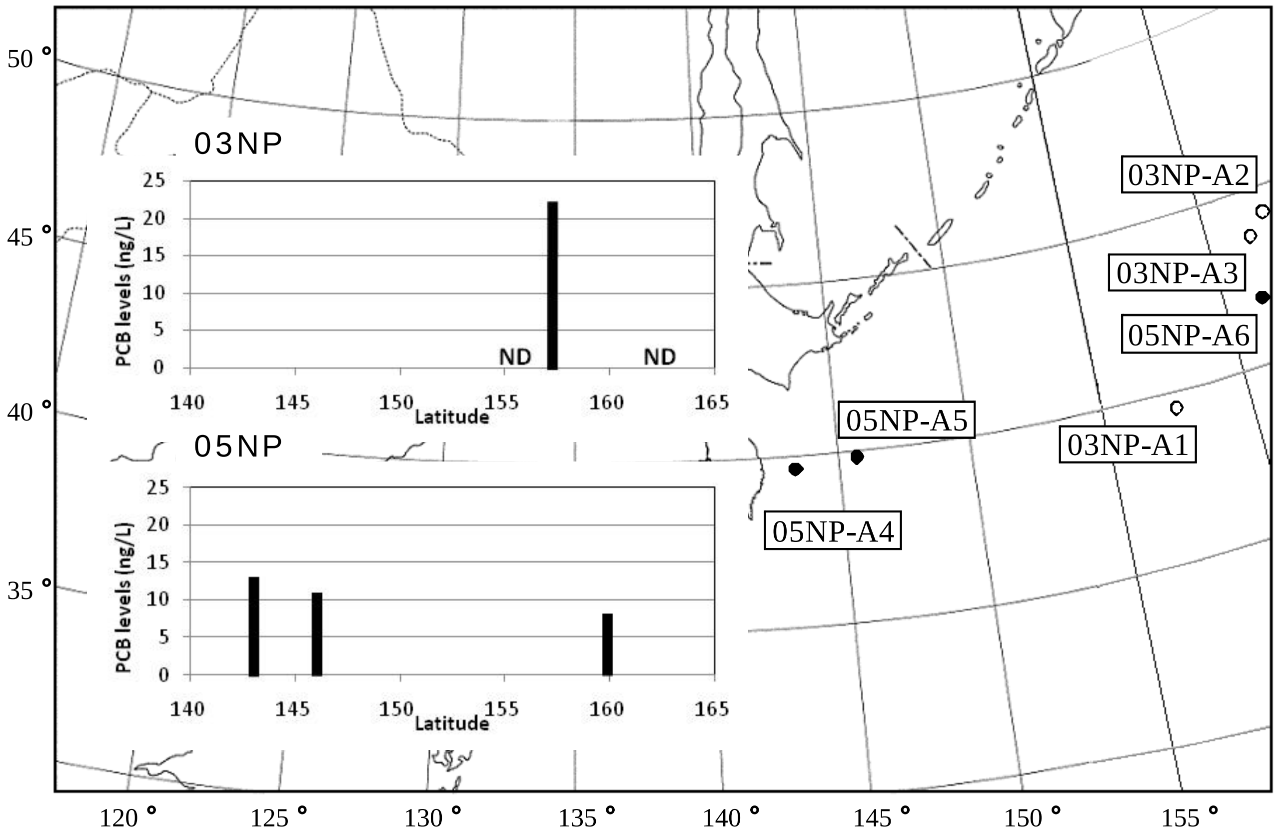
Model	R	R2	R2'
1	0.624	0.389	-0.069

b) Analysis of Variance Table

Model	Sum of Squares	DF	Mean Square	F value	P value
Regression	5.450	6	0.908	0.850	0.567
Residual	8.550	8	1.069		
Total	14.000	14			

c) Variables

Model	B	SE	B'	T	P value
Constant	-2.55E-10	0.267		0.000	1.000
year	-0.769	0.531	-0.769	-1.447	0.186
date	1.008	0.747	1.008	1.350	0.214
latitude	-1.044	0.933	-1.044	-1.119	0.296
longitude	0.170	0.404	0.170	0.420	0.685
body length	-0.181	0.393	-0.181	-0.461	0.657
blubber thickness	-0.121	0.525	-0.121	-0.231	0.823



ND: Not detected

Fig. 1 Concentrations of PCBs (pg/m^3) in air samples from western North Pacific Ocean in the 2003 and 2005 JARPN II.

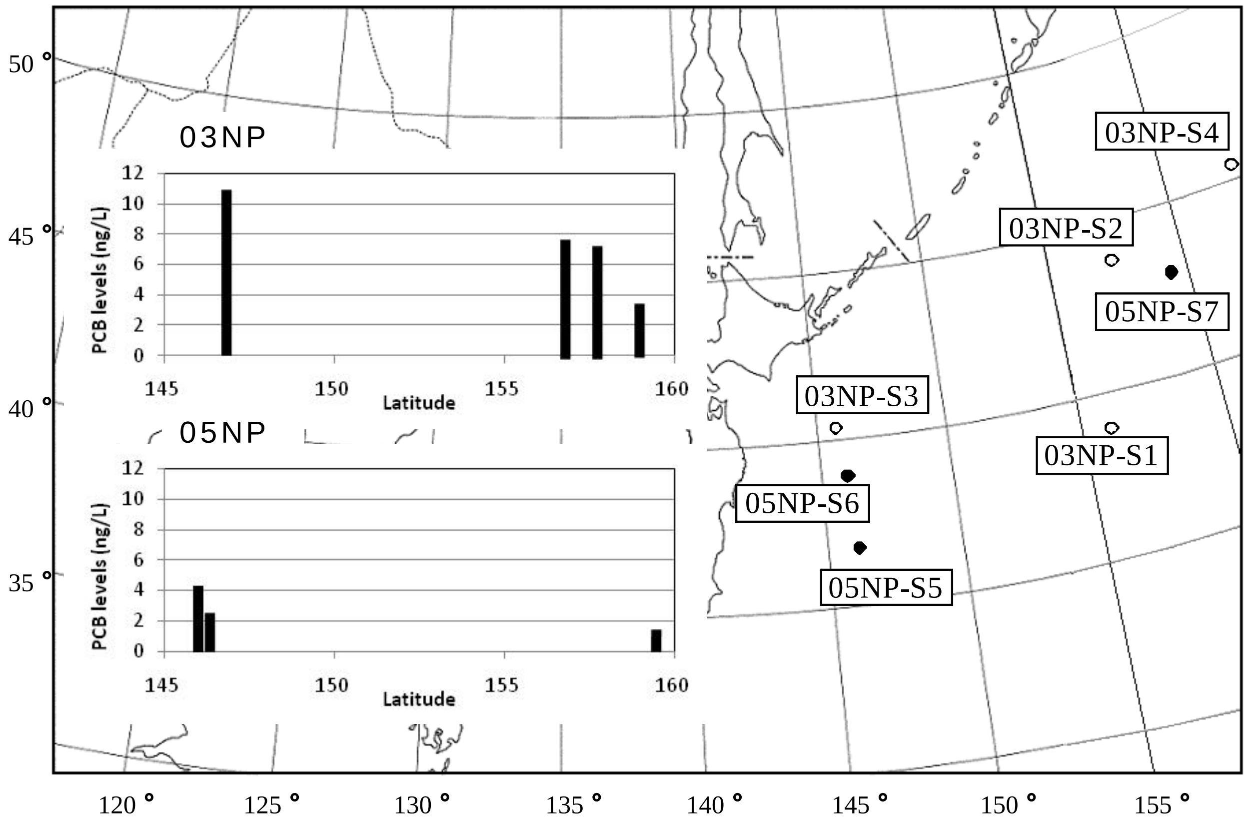


Fig. 2 Concentrations of PCBs (ng/L^3) in surface seawater samples from western North Pacific Ocean in the 2003 and 2005 JARPN II.

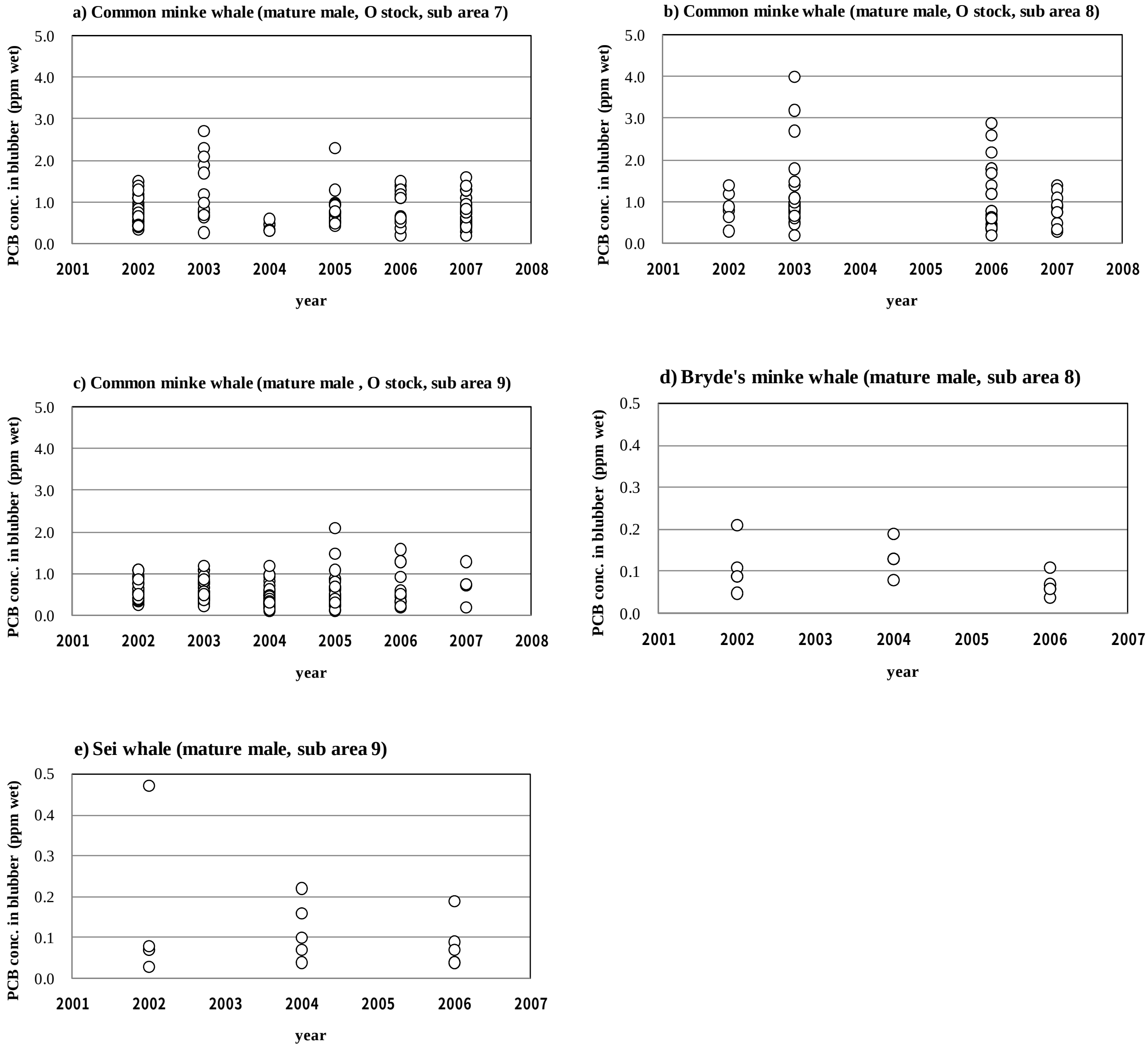


Fig. 3a-e. PCB levels in blubber of common minke, Bryde’s and sei whales in the western North Pacific during a period from 2002 to 2007.