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Horizontal and vertical profiles of PCBs in sediments from the southeastern coastal areas of Korea

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The concentrations of 20 congeners of PCBs were determined in surficial and dated sediments from Jinhae, Busan, Ulsan and Yeongil Bay of Korea to assess current distributions as well as historical trends. The concentration ranges of total CBs in sediments were nd–138.60 ng/g dry wt. The concentrations of total CBs were significantly different among bays in the southeastern coastal areas (Kruskal-Wallis Test, p=0.036), showing the order of Busan>Ulsan>Yeongil>Jinhae Bay. Predominant PCB homologues were penta-, hexa- and hepta-CBs. The down-core distributions of PCBs showed that the highest concentrations occurred in the late 1960s, coinciding with the onset of industrialization of Korea. The compositions of PCB congeners in sediment among bays were uniform, suggesting that major sources were not changed over time.

INTRODUCTION

Polychlorinated biphenyls (PCBs) are ubiquitous pollutants, and have been widely used by various industries over the past 50 years. PCB mixtures are thermally stable, resistant to chemical degradation and are of low flammability (Alford-Stevens, 1986). However, since the discovery of PCBs in the environment in 1966, it has been recognized that the usage of PCB mixtures involves a major risk for organisms in the aquatic environment, due to their persistence and known toxicity (Bavel *et al.*, 1995). Environmental contamination by PCBs has arisen exclusively from human activities and their heavily contaminated areas tend to be located around the industrial areas (Edgar *et al.*, 1999). Wastewater discharge to the sea is an important route for PCBs into the marine environment. Jinhae, Busan, Ulsan, and Yeongil Bay on the southeastern part of Korea, have been subjected to pollution by effluent and accidental discharges from a large number of engineering works and dumping of sewage sludge by major cities such as Masan, Busan, Ulsan, and Pohang. They are accumulated in the sediments due to their low solubility and adsorption on the particles (Parts *et al.*, 1990). The contaminated sediments serve as pollutant reservoirs and therefore pose ecological and human health risks for the prolonged periods of time. The objective of this study was to examine the concentrations and patterns of individual CBs in the sediments and to explore the input history of organic contaminants to the southeastern coastal areas of Korea by the interpretation of the sediment record.

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MATERIAL AND METHODS

Surface sediments and sediment cores were collected from Jinhae, Busan, Ulsan and Yeongil Bay on the southeastern part of Korea, which are surrounded by heavily industrialized cities such as Masan, Busan, Ulsan and Pohang (Fig. 1). The surface sediment samples from the top 0–5 cm were obtained from 21 sites by box core in November 2000. Sediment cores were also collected by diver using an acrylic tube (8.5 cm×200 cm) in Jinhae Bay (station 1') and Ulsan Bay (station 3) in September 1999 and Busan Bay (station 1) and Yeongil Bay (stations 2) in September 2000. Care was taken to obtain core sample without disturbing the floccules existing above the surface sediment. After collection, the samples were freeze-dried and then sieved at 2 mm. Samples were stored at –20°C until extraction. Separate samples were also taken for the organic carbon determination. Sediments were extracted (10 g) using ASE (Accelerated Solvent Extraction) system under the conditions of the extraction of 10 min at 30 W. The

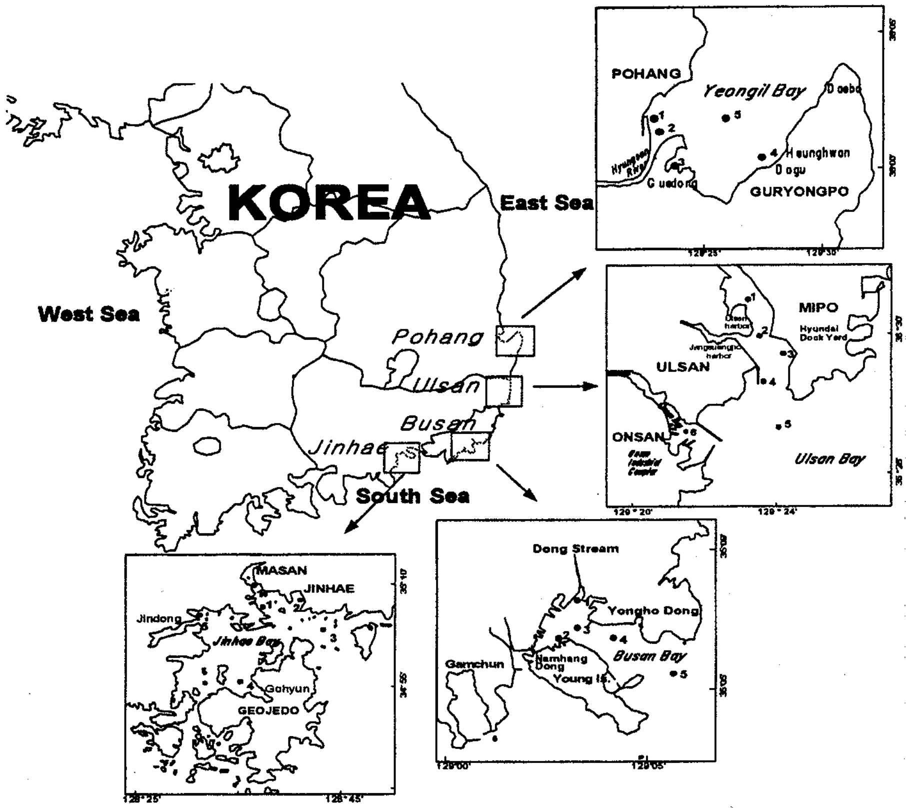

Fig. 1. Sampling sites of the southeastern coastal areas of Korea.

extraction solvent was hexane and acetone (1:1). Activated copper was added to the extraction vessel to desulfurize the extract. The sample was filtered and the total organic extract reduced to a small volume using a rotary evaporator and then purified on a multi-layer silica column (1 g silicagel/4 g NaOH-silicagel/1 g silicagel/8 g H_2SO_4-silicagel/2 g silicagel, Aldrich). In this study, 20 PCB congeners reported and were identified (CB 8, 18, 29, 28, 52, 44, 101, 87, 110, 118, 153, 138, 187, 128, 180, 170, 195, 205, 206, 209). PCB quantifications were conducted on a Hewlett-Packard 6890 gas chromatograph (GC) with an electron capture detector equipped with HP autosampler 7683. A capillary column DB-XLB was used (60 m×0.32 mm internal diameter×0.25 μm film thickness, J&W). The oven temperature was programmed from 100°C (1 min) to 265°C at 1.2°C/min. The injection port and detector temperatures were set at 250°C and 300°C, respectively. Nitrogen was used as carrier gas and make-up gas. Quality assurance and quality control procedure included initial and ongoing calibration of the standards, procedural blanks and the analysis of a reference material. The recovery yield of certified PCBs in a standard reference marine sediment (CRM, NRC) extracted using the same methodology was 70–80%. Detection limits were about 0.1 ng/g for the individual PCB congeners. Total organic carbon (TOC) was measured by CHN analyzer. Sedimentation rates using ^{210}Pb and ^{137}Cs profiles were determined. Multivariate analysis was conducted using SPSS 10.0 package.

RESULTS AND DISCUSSION

Horizontal distribution

The concentration of selected PCB congeners and their sum in the marine sediments from Jinhae, Busan, Ulsan and Yeongil Bay on the southeastern part of Korea are shown in Table 1. The total PCB concentrations were in the range of nd–138.60 ng/g dry wt. The highest concentration was found at station 2 of Ulsan Bay, followed stations 3, 1, and 2 of Busan Bay. As expected, sites near industrial activities contained high levels. However, this tendency was not registered at station 6 of Ulsan Bay adjacent to Onsan complex and station 3 of Yeongil Bay close to POSCO. The high amount of sand (62%) might contribute to low levels of PCBs at station 6 of Ulsan Bay. Camcho-Ibar and McEvoy (1996) reported that PCB concentrations in sediments are strongly dependent on the amount of fine particles in the samples. The anal-

Table 1. Results on the individual PCB congeners concentrations (ng/g dry wt), organic carbon (%), grain size (%) and normarlized PCBs concentrations in sediments from the southeastern coastal areas of Korea

	Jinhae Bay					Busan Bay					Ulsan Bay						Yeongil Bay				
	1	2	3	4	5	1	2	3	4	5	1	2	3	4	5	6	1	2	3	4	5
PCB 8	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PCB 18	nd	nd	nd	nd	nd	0.15	0.74	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.24	0.41	nd	nd	nd
PCB 28	nd	nd	nd	nd	nd	nd	nd	1.23	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PCB 44	0.94	nd	nd	nd	0.13	13.45	0.52	0.66	0.22	0.11	0.13	2.19	0.27	nd	nd	nd	0.22	0.40	nd	nd	nd
PCB 52	0.80	nd	nd	nd	nd	17.12	0.60	0.87	0.24	0.13	nd	4.32	0.44	0.10	nd	nd	0.20	0.48	nd	nd	nd
PCB 87	0.39	nd	nd	nd	nd	nd	nd	nd	nd	0.16	0.37	nd	nd	0.32	nd	nd	nd	0.45	nd	nd	nd
PCB 101	1.72	nd	nd	nd	0.52	nd	4.29	6.72	2.59	nd	1.19	14.68	3.35	nd	nd	nd	2.18	1.65	nd	nd	nd
PCB 110	0.76	nd	nd	nd	nd	2.58	1.49	2.38	0.78	0.27	0.55	7.86	1.69	0.44	0.20	0.17	0.43	0.68	nd	0.13	0.12
PCB 118	0.70	nd	nd	nd	nd	2.80	1.48	2.45	0.75	0.26	0.48	7.39	1.50	0.38	0.19	nd	0.40	0.66	nd	0.14	0.12
PCB 128	0.19	nd	nd	nd	nd	nd	0.83	1.66	0.43	0.11	0.47	4.09	0.83	0.16	nd	nd	0.18	0.22	nd	nd	nd
PCB 138	1.02	nd	nd	nd	0.22	nd	5.99	11.10	3.09	0.64	1.16	27.39	5.72	0.92	0.36	0.39	4.85	1.38	0.11	0.24	1.72
PCB 153	1.23	0.23	nd	0.35	0.44	6.39	6.09	10.83	3.41	0.96	1.61	28.34	6.35	1.46	0.64	0.51	0.97	1.61	nd	0.42	0.38
PCB 170	0.13	nd	nd	nd	nd	1.22	1.85	3.44	0.90	0.22	0.30	8.43	1.59	0.26	0.20	nd	0.22	0.30	nd	nd	nd
PCB 180	0.36	nd	nd	nd	0.11	1.91	3.70	6.70	1.94	0.33	0.80	15.66	3.37	0.50	0.33	nd	0.56	0.83	nd	0.11	nd
PCB 187	0.40	0.18	nd	0.17	0.25	2.09	3.51	6.61	1.71	0.43	0.57	13.11	2.67	0.51	0.33	0.10	0.62	0.63	nd	0.26	0.23
PCB 194	0.12	nd	nd	nd	nd	nd	1.53	2.67	0.65	0.24	0.18	5.14	0.97	nd	0.15	nd	0.36	0.24	nd	nd	nd
PCB 195	nd	nd	nd	nd	nd	nd	0.71	1.43	0.41	0.10	nd	2.87	0.49	nd	nd	nd	nd	0.11	nd	nd	nd
PCB 205	nd	nd	nd	nd	nd	nd	nd	0.20	0.04	nd	nd	0.29	nd	nd	nd	nd	nd	nd	nd	nd	nd
PCB 206	nd	nd	nd	nd	nd	nd	nd	1.49	0.39	0.13	nd	1.11	0.21	nd	nd	nd	nd	0.11	nd	nd	nd
PCB 209	nd	nd	nd	nd	0.12	0.44	0.31	0.33	0.10	nd	0.27	0.39	nd	0.10	nd	nd	0.16	nd	nd	nd	nd
ΣPCBs	8.76	0.41	nd	0.52	1.67	47.57	31.88	56.08	16.70	3.85	7.78	138.60	28.74	5.05	2.40	1.16	11.18	9.53	0.11	1.31	2.56
OC	2.56	2.21	1.05	3.33	2.59	2.23	2.32	2.34	1.76	1.37	1.26	1.64	1.11	1.56	1.18		1.35	1.08	3.68	1.20	1.02
<63 um	98	96	77	99	98	95	81	96	80	99	85	98	86	93	88	38	71	80	92	86	79
Normalized ΣPCBs	342	19	nd	16	65	2133	1374	2397	949	281	618	8451	2589	324	203		828	882	3	109	251

nd: not detected
OC: organic carbon

ysis of PAHs to these bays revealed that the concentration of PAHs was the highest at station 3 of Yeongil Bay (Moon *et al.*, 2001). It seems that POSCO is suspected sources more responsible for PAHs than PCBs. The PCBs levels of southeastern coast sediments except station 1 of Ulsan coast seemed to be comparable with those from US-Mexico border region (0.6–36 ng/g: Gutierrez-Galindo *et al.*, 1998) and Liverpool Coast (0.082–37.88 ng/g: Camacho-Ibar and McEvoy, 1996), and lower than that from Corio Bay (14–810 ng/g: Prest *et al.*, 1995) and Osaka Bay (63–240 ng/g: Iwata *et al.*, 1994). According to Canadian sediment quality guideline (Smith *et al.*, 1996), stations 1, 2 and 3 of Busan Bay and 2 of Ulsan Bay were moderately contaminated sites as PCB concentrations were more than 21.5 ng/g. The levels of total PCBs were significantly different among bays (Kruskal-Wallis Test, p=0.036), showing the order of Busan>Ulsan>Yeongil>Jinhae Bay. The concentration of organic contaminants is often expressed as the carbon-normalized PCBs to compensate for the differences in grain sediment characteristics. Organic carbon contents of sediments in these study areas varied between 1.05 and 3.68% and silt-clay contents were in the range of 71–99% (Table 1). Although a strong correlation between PCB concentrations and the contents of fine particles in sediment was reported (Tolosa *et al.*, 1995; Camacho-Ibar and McEvoy, 1996), this feature was not observed in this study. It might result from the fact that the sediments of the bays (except St. U6) were predominantly silt-clay (<63 μm), accounting for 71–99% of the sediment. The comparison of the normalized PCBs concentrations among bays to organic carbon showed significant difference like total PCBs (Kruskal-Wallis Test, p=0.024). The contribution of individual chlorobiphenyl (CB) isomers and congeners to total PCB concentrations was not evenly distributed across 20 congeners. Penta-(101, 87,110,118), hexa-(153,138,128) and hepta-CBs (187,180,170) among PCB homologues in four bays contained higher proportions, accounting for 74–93% of total PCBs (Fig. 2). The octa- and deca-chlorinated con-

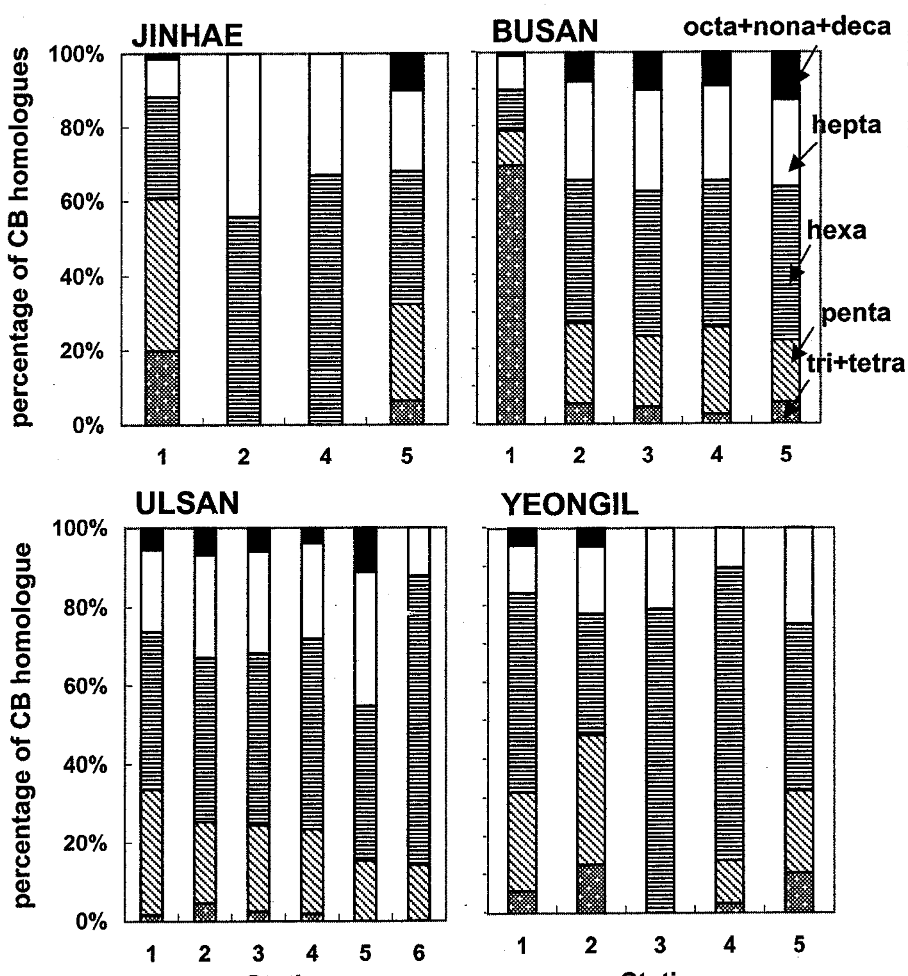

Fig. 2. Distribution of CB homologues in the marine sediments from the southeastern coastal areas of Korea.

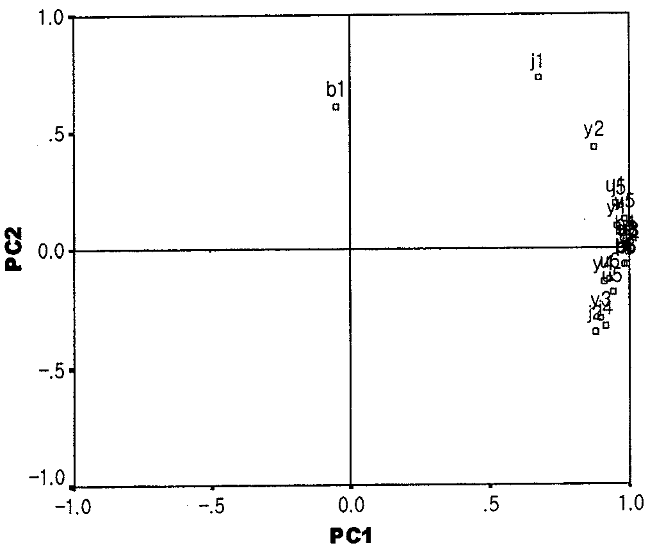

Fig. 3. Principal component plot for the southeastern coastal areas (j1-5: stations of Jinhae Bay, b1-5: stations of Busan Bay, u1-5: stations of Ulsan Bay, y1-5: stations of Yeongil Bay).

geners generally made small individual contributions to total loading. This pattern was similar to those observed sediments from several marine coastal locations in which penta-, hexa-, hepta-, or octa-CBs contributed greater proportions of the total PCBs (Pruell *et al.*, 1990; Kannan *et al.*, 1997). To evaluate the relative homologues distribution at the different sampling sites, PCA analysis was used. PCA revealed CB homologues were similarly distributed at 18 stations except station 1 of Busan Bay, station 1 of Jinhae Bay and station 2 of Yeongil Bay (Fig. 3). These three stations located in the inner sites of bays differed from the other stations in the contributions of tri- and tetra-CB homologues, which were slightly higher at those sites. Water column usually contained less chlorinated PCBs. Highly chlorinated PCBs exist in sediments than in water because of PCBs high hydrophobicity (Pruell *et al.*, 1990; Tolosa *et al.*, 1995; Kannan *et al.*, 1997). Therefore, it could be assumed that high percentages of less chlorinated PCBs reflected the recent input of PCB through the adsorption on the particles to those three sites.

Vertical profiles

The sedimentation rates were determined using ^{210}Pb and ^{137}Cs data. ^{210}Pb and ^{137}Cs activity profiles are presented in Table 2. Exponential decay of ^{210}Pb below the surface mixed layer yielded sedimentation rates of 0.12 and 1.39 cm/yr for Yeongil Bay and Ulsan Bay, respectively. Very close agreement on sedimentation rates was found between ^{210}Pb and ^{137}Cs. Core taken in Busan site showed vertical well-mixed activity profiles of ^{210}Pb, indicating a strong sediment mixing and/or deposition. The depth pro-

Table 2. Depth profiles of ^{210}Pb, ^{226}Ra and ^{137}Cs activities (dpm/g) in sediments from the southeastern coastal areas of Korea

Depth (cm)	Busan Bay (St.1)			Ulsan Bay (St. 3)			Yeongil Bay (St. 2)		
	^{210}Pb	^{226}Ra	^{137}Cs	^{210}Pb	^{226}Ra	^{137}Cs	^{210}Pb	^{226}Ra	^{137}Cs
0–2				10.02±1.27	1.72±0.21	0.3±0.11	7.80±0.94	1.54±0.19	0.17±0.10
2–4	8.25±1.10	1.70±0.22	0.18±0.10				14.34±1.94	3.01±0.35	0.38±0.14
6–8							8.15±1.08	2.31±0.23	0.28±0.13
8–10				9.14±1.25	1.96±0.22	0.18±0.10	4.96±0.91	1.62±0.19	0.20±0.10
10–12	8.01±1.06	1.62±0.20	0.18±0.09				3.52±0.68	1.59±0.13	0.12±0.06
12–14									
14–16				7.14±1.01	2.05±0.16	0.20±0.07			
18–20	8.71±1.16	1.38±0.22	0.39±0.13				3.89±0.75	1.81±0.16	0.16±0.07
20–22				7.37±1.18	1.88±0.22	0.16±0.10			
26–28				7.34±0.73	2.21±0.16	0.21±0.07	4.20±0.62	1.98±0.14	0.18±0.05
28–30									
34–36	8.47±1.21	1.73±0.24	0.27±0.14	6.30±0.92	2.00±0.19	0.40±0.08	4.02±0.83	1.84±0.17	
36–38									
42–44				5.05±0.97	2.29±0.21	0.33±0.10			
46–48				3.31±0.40	2.19±0.14	0.38±0.08			
50–52				5.98±0.92	1.64±0.18	0.51±0.10			
58–60				6.13±1.08	2.20±0.25	0.40±0.11			
64–66				5.64±1.13	1.67±0.24	0.30±0.12			

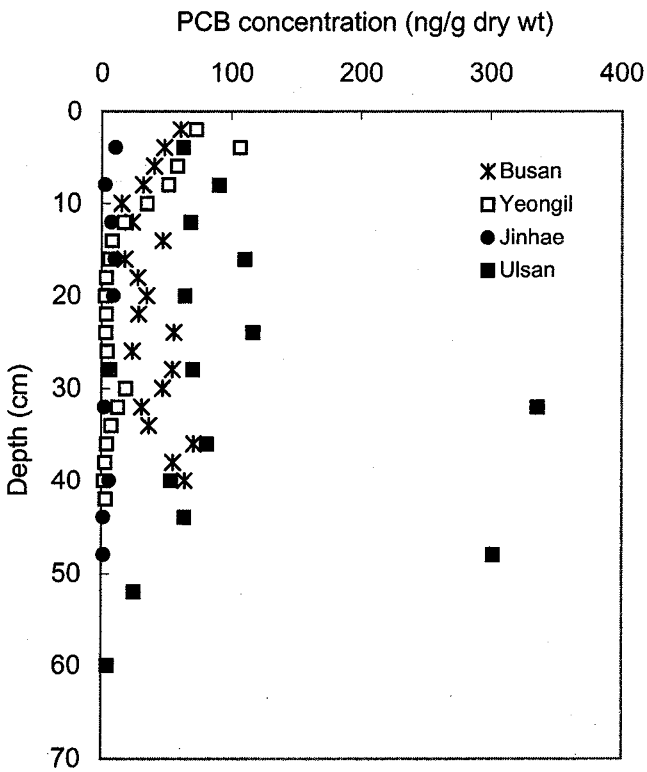

Fig. 4. Depth profiles of ΣPCBs in sediments from the southeastern coastal areas of Korea.

files in sediments might be disturbed by the activity of benthic organisms biologically or the physical mixing such as the transportation of ships, the violent hydrodynamical conditions or fishing activities using trawl nets. In Busan Bay, it could be assumed that the physical mixing caused by transportation of ships was likely to be the principal mechanism because the site was located in transportation route of large commercial harbors. The sedimentation rate of Jinhae site was used 0.40 cm/yr by Yang *et al.* (1995). The historical trends in PCBs in Jinhae, Ulsan and Yeongil cores were dominated by distribution of subsurface maximum with decreased contaminant levels both to the surface and with depth in the cores (Fig. 4). Maximum concentrations of total PCBs were detected at depths of 34–36 cm for Busan site, 18–22 cm for Jinhae site, 30–32/46–48 cm for Ulsan site and 2–4 cm for Yeongil site. These sections of the core approximately corresponded to the early 1960s in Jinhae Bay, in the late 1970s/late 1960s in Ulsan Bay and in the late 1960s in Yeongil Bay (Table 3). PCB concentrations in surface sediments corresponding to the 1999 or 2000 were 1.5–5 fold less than the maximum value observed for sediments deposited in the late 1960s. In 1960, Korea entered a period of rapid industrial development and major industrial activities have occurred in Masan, Busan, Ulsan and Pohang. Development resulted in the release of pollutants, many of which were transported by river to ocean. Internationally, large-scale production of PCB commenced in the 1930s for use in a variety of industrial applications. PCBs were never manufactured in Korea, but have been imported and used extensively in the electricity industry as insulating fluids or resins in transformers and capacitors until 1984 when imports were banned (Kim & Lee, 1996). From these finding, it could be assumed that inputs were diffusely associated with development even though information relating to the PCBs imported into Korea is extremely limited. Venkatesan *et al.* (1999) also reported that maximum concentrations in sediment cores of Richardson Bay correspond to the period when PCB usage was at a maximum. However, Yamashita *et al.* (2000) observed that concentration of PCBs increased dramatically from the late 1970s to the early 1980s by a time lag between peak periods of production and usage and the deposition to coastal environment. The PCB congener patterns in sediment cores were dominated by penta-, hexa- and hepta-CBs, collectively accounting for 42–95% of the total PCBs in Busan and Yeongil Bay (Fig. 5). The uniformity on patterns profiles of PCB congeners in the sediment cores suggested that the PCB contaminants sources have not changed over time barely since the beginning period of input of PCBs into the southeastern coastal areas.

Accumulation rate

The PCB accumulation rates were obtained using calculated sedimentation rates and sediments densities as well as the concentrations of PCBs. Table 3 shows the estimated PCB accumulation rates into sediments. The accumulation rates of Ulsan Bay were 1–2 order of magnitude higher than that of Jinhae and Yeongil Bay. However, the rates were lower than those of Narragansett Bay (Latimer & Quinn, 1996) and similar to those of Tokyo Bay (Yamashita *et al.*, 2000). The PCB accumulation rates in Jinhae and Yeongil Bay were comparable to those reported in northwest Mediterranean sediments (Tolosa *et al.*, 1995). The wide range of accumulation rates among areas seemed to derive from the difference of concentrations, sediment characteristics and sedimentation rates.

Table 3. Sediment accumulation rates (ng/cm^2/yr) and PCB concentrations (ng/g dry wt) in sediment cores of the southeastern coastal areas of Korea

Depth (cm)	Jinhae Bay (St. 1')			Ulsan Bay (St. 3)			Yeongil Bay (St. 2)		
	Approx. time period of core	Conc	Rate	Approx. time period of core	Conc	Rate	Approx. time period of core	Conc	Rate
0–2	1995–1999			1998–1999			1983–2000	72.75	4.37
2–4	1991–1995	10.56	2.53	1996–1998	62.48	43.42	1967–1983	106.34	6.38
4–6	1986–1991			1995–1996			1950–1967	57.82	3.47
6–8	1982–1986	2.75	0.66	1993–1995	90.55	62.93	1933–1950	51.17	3.07
8–10	1978–1986			1992–1993			1917–1933	34.71	2.08
10–12	1974–1978	7.48	1.80	1990–1992	68.51	47.61	1900–1917	17.13	1.03
12–14	1970–1974			1989–1990					
14–16	1966–1970	10.29	2.47	1987–1989	110.34	76.69			
16–18	1961–1966			1986–1987					
18–20	1957–1961	9.17	2.20	1985–1986	64.23	44.64			
20–22	1953–1957			1983–1985					
22–24	1949–1953			1982–1983	116.65	81.07			
24–26	1945–1949			1980–1982					
26–28	1941–1945	4.79	1.15	1979–1980	70.37	48.91			
28–30	1936–1941			1977–1979					
30–32	1932–1936	2.48	0.60	1976–1977	335.80	233.38			
32–34	1928–1932			1975–1976					
34–36				1973–1975	81.31	56.51			
36–38				1972–1973					
42–44				1967–1969	63.97	44.46			
46–48				1964–1966	301.75	209.72			
50–52				1962–1963	25.01	17.38			
58–60				1956–1957	4.51	3.13			
64–66				1952–1953					

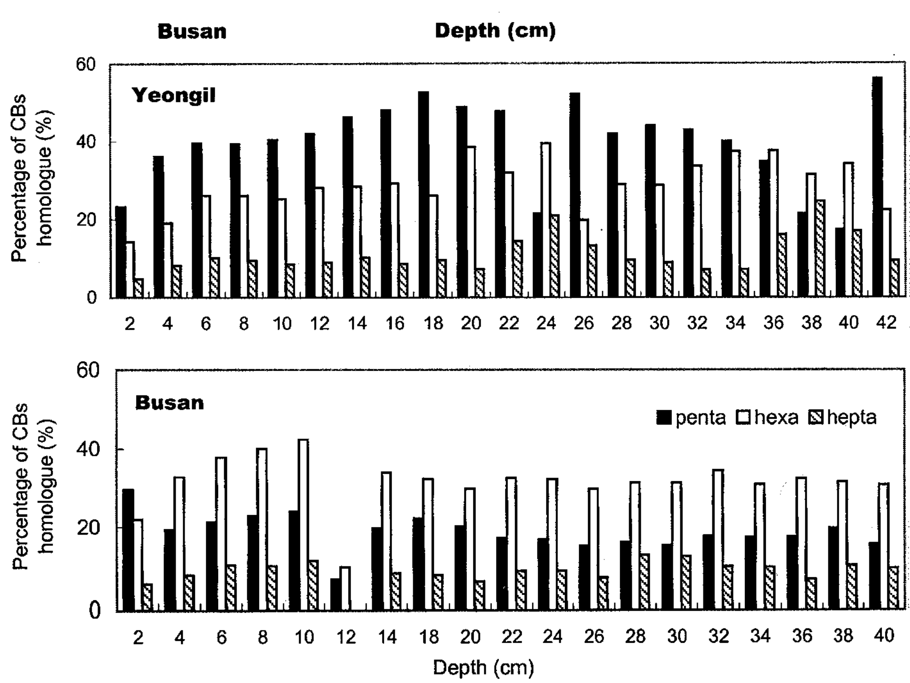

Fig. 5. Percentage of penta-, hexa, and hepta-CBs to total PCBs in sediment cores from Yeongil and Busan Bay.

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