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Distribution of Certain Chlorobenzene Isomers in Marine Sediments from the Southeastern Coastal Areas of Korea

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Eight chlorobenzenes (out of a total of 12 in the congener series) were measured in the sediments from 21 stations in the southeastern coastal areas of Korea. The levels of total chlorobenzene isomers varied between 2.08 and 12.45 ng/g dry weight. The highest contents of total chlorobenzenes (CBs) were found in the sediments from Pohang coast. Trichlorobenzenes (the sum of 1,3,5-, 1,2,4- and 1,2,3-trichlorobenzene) were dominant classes among four congeners, whereas tetrachlorobenzenes (the sum of 1,2,3,5-, 1,2,4,5- and 1,2,3,4-tetrachlorobenzene) and pentachlorobenzene were low levels. The contributions of total CBs showed similar patterns for all stations with positive significant correlation within CBs species. It means that CBs contamination in the southeastern coasts of Korea came from the similar source.

Key words: Chlorobenzenes (CBs), Sediment, Trichlorobenzenes, Tetrachlorobenzenes, Pentachlorobenzene, Contributions

Introduction

Chlorobenzenes (CBs) are ubiquitous hydrophobic chlorinated organic compounds in the environment. CBs are a group of benzene cyclic compound substituted with one to six chlorine atoms (Fig. 1). This results in 12 different isomers and congeners according to a position and number of chlorine. These compounds are used as de-ordants, solvents and pesticides, as well as byproducts of agro- or petro-chemical related manufacturing processes, such as PCBs and pentachlorophenol, or of biodegradation of lindane (Newhook and Meek, 1994). Unlike some organochlorine (OC) compounds, including polychlorinated biphenyls (PCBs) and various pesticides, CBs are not banned from production or use in any country until 1990 (Lane et al., 1992). Hexachlorobenzene (HCB) of CBs is widespread pollutants in various environmental compartments such as water, air, soil, birds, fish, human milk, adipose tissue, and blood (Taguchi et al., 1989; Tanabe et al., 1993; Umegaki et al., 1993; Kelly and Campbell, 1994; Hermanson et al., 1997; Lee et al., 1997). HCB residue in the environment is predominantly industrial byproducts, which originate from the manufacture of chlorobenzene, carbon tetrachloride, trichloroethylene, tetrachloroethylene, polyvinyl chloride, and nitroso rubber. HCB is bio-accumulated in species ranging from algae to catfish by

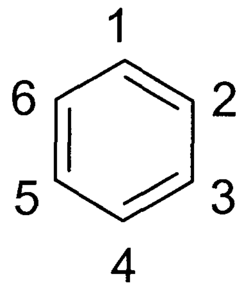

Fig. 1. Structure of chlorobenzenes and numbering system of chlorine.

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1,000~10,000 (Pastor et al., 1996). They are often synthetic precursors, byproducts or contaminants resulting from synthesis or degradation of other organic compounds. They also cover a wide range of volatility, some with vapor pressure comparable to various pesticides (Lane et al., 1992). They may be accumulated in tissue in local areas where they are discharged (Hermanson and Hites, 1990) or far from where they were used or produced (Bidleman et al., 1989). Their environmental persistence, which may produce adverse health effects in organisms, is an enough serious issue to the marine environment.

According to Williams et al. (1988), HCB was found in all of human tissue samples, as often as PCB or OC pesticides. Its high bioaccumulative potential has resulted in its identification as "probably carcinogenic to humans" (Newhook and Meek, 1994). Also, HCB have been implicated as endocrine disrupting chemical in human and other animals. In addition, Angélique (1999) suggested that HCB is also possible impact on the toxic equivalent (TEQ). Based on binding to the Ah-receptor, the dioxin-like effects, and the bioaccumulation in higher trophic levels, HCB should be classified as a dioxin-like compound. The mechanism of action resembles that of mono-ortho substituted PCBs, which have also phenobarbital-like properties and are included in the TEQ concept. Based on the limited information available it was estimated that HCB is about 10,000 times less potent than 2,3,7,8-tetrachloro dibenzo-*p*-dioxin (2,3,7,8-TCDD). The human occurrence of other isomers is low (<15% for pentachloro-, 0% for trichloro- and tetrachloro-isomers; Williams et al., 1988) in part because their half lives in important sources, such as fish, are a fraction of that for HCB. Human carcinogenic effect of trichloro-, tetrachloro- and pentachloro- isomers cannot be classified according to some studies (Giddings et al., 1994a, b). Thus, the traditional emphasis in investigations in environmental distribution has been only on HCB. However, other CBs are common waste products, and because of their short half-lives in birds and fish, they are observed on site and event-specific bases during periods of heavy contamination (Gebauer and Weseloh, 1993; Oliver, 1987). Previous investigations have seldom quantified their concentrations in environmental media despite the fact that many of these congeners are listed as priority pollutants owing to their toxicity and carcinogenicity (Harper et al., 1992). In particular, there are few data on level of chlorobenzene isomers available in marine environment. This investigation is the first effort for chlorobenzene isomers in Korean coastal ecosystem. The aim of this work was to investigate the levels of certain chlorobenzene isomers including HCB in marine sediments from southeastern coasts of Korea.

Material and Methods

Surface sediments (depth, 0~5 cm) were twice sampled from 21 stations in Pohang, Busan, Ulsan and Jinhae coasts of Korea in February and September 2000 (Fig. 2). Sediments were collected either with a Van Veen grab or box-corer sampler and then kept frozen at −20℃ until analysis. The freeze-dried sediments were sieved at 2 mm and then extracted in a Soxhlet apparatus with 200 mL of toluene (Ultra residue analysis, J.T. Baker, USA) for 20 hours. The volume was reduced to 1~2 mL in a rotary evaporator. The extract was transferred to *n*-hexane (Ultra residue analysis, J.T. Baker, USA) and adjusted to a volume of 10 mL after a spike of 4 species internal standard ($^{13}C_6$-1,2,3-TrCB, $^{13}C_6$-1,2,3,4-TeCB, $^{13}C_6$-PeCB and $^{13}C_6$-HCB; MCBS, Wellington Laboratories, Canada). Samples were cleaned up on a multi-layer slilica gel (Art No. 7734, 70~230 mesh, Merck) column chromatography containing $AgNO_3$-silica gel, H_2SO_4-silica gel and KOH-silica gel with *n*-hexane. The eluants were concentrated to less than 1 mL, and left at room temperature for one day to evaporate to 50~100 μL. The residues were dissolved with 50 μL of *n*-nonane (Pesticide residue analysis, Fluka, Switzerland) and then analyzed by gas chromatography coupled to mass spectrometery. An Agilent 6890 series II GC (Agilent, USA) equipped with a split/splitless injector was used (splitless time: 2 min; flow: 70 mL/min). Samples were injected splitlessly (2 μL portion of the total 50 μL) at the injector temperature of 250℃. The GC temperature program was from 50℃ (1.5 min), 3℃/min to 170℃, and then 10℃/min to 300℃ (5 min). The carrier gas was helium at a constant flow rate of 1.3 mL/min. A HP-1 (30 m, 0.25 mm ID, 0.25 μm film thickness, Hewlett Packard, USA) capillary column used for the separa-

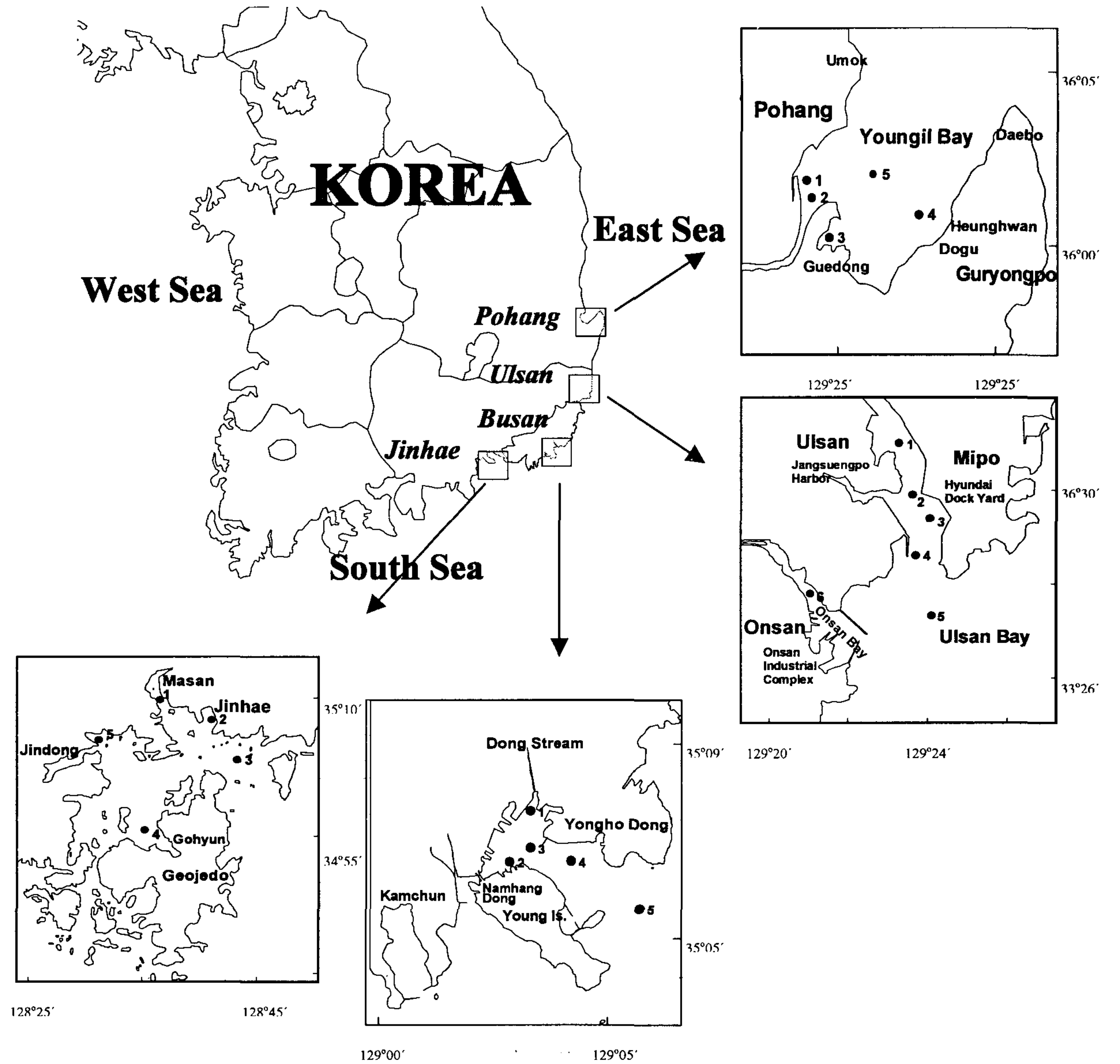

Fig. 2. Map showing and the corresponding sampling stations (enlarged map) in the southeastern coastal areas of Korea.

tion of chlorobenzene compounds. The GC was coupled to a 5973N mass selective detector (MSD). The mass spectrometer was operated under the selective ion monitoring (SIM) mode using two molecular ions ($M^{\cdot+}$ and $(M+2)^{\cdot+}$ for each degree of chlorination were monitored in the electron impact (EI) ionization mode at 70 eV. The interface temperature was 250℃.

The compounds of interest include 1,2,3-, 1,2,4- and 1,3,5-isomers of trichlorobenzene (1,2,3-TrCB, 1,2,4-TrCB and 1,3,5-TrCB), 1,2,4,5-, 1,2,3,4- and 1,2,3,5-isomers of tetrachlorobenzene (1,2,4,5-TeCB, 1,2,3,4-TeCB and 1,2,3,5-TeCB), pentachlorobenzene (PeCB) and hexachlorobenzene (HCB). We selected eight isomers ranging from tri- to hexachlorobenzene based on persistent ability to retain them on marine sediment materials, and analyzed them our chromatographic system. This method could not separately quantify the 1,2,3,5- and 1,2,3,4-tetrachlorobenzene and they were therefore quantified together as a compound. Blanks were run before and after standards to check for carryover. Sample recoveries were in the range of 83~97%. The data presented in this investigation were not corrected for recoveries.

The contents of total organic carbon (TOC) in marine sediments were obtained using a CHN analyzer (Perkin Elmer, 2400), after elimination of the calcium carbonate adding 1 N HCl. Grain size analyses were carried out by wet sieving, to separate sands, after a pretreatment with H_2O_2.

Results and Discussion

Sample characterization

TOC, grain size and water contents in surface sediments from the southeastern coastal areas of Korea is summarized in Table 1. The contents of TOC ranged from 1.02 to 4.64% for all sediment samples. Sediments were primarily muddy (about 84~94%) for all stations. Mud contents in sediments from Busan and Jinhae coasts were higher than those from Pohang and Ulsan coasts. Water contents in all sediments ranged from 28.0 to 65.4%.

Distributions of chlorobenzene isomers in surface sediments

Table 2 shows the concentrations of CB isomers in sediments from the southeastern coasts of Korea. Eight CB isomers were detected in all samples. The highest level of total CBs was found in Station P3 from Pohang coast. The levels from Pohang coast ranged from 2.61 to 12.45 ng/g dry weight with mean concentration of 5.09 ng/g dry weight. Total CBs concentrations in sediments from Ulsan coast were in the ranges of 3.75~9.13 ng/g dry weight and mean concentration was 5.06 ng/g dry weight. In Busan coastal areas, total concentrations of CBs ranged between 4.24 and 7.26 ng/g dry weight with mean concentration of 5.21 ng/g dry weight. Jinhae Bay showed the lowest levels of CBs, ranging from 2.08 to 3.45 ng/g dry weight. Pohang and Ulsan coasts were located close to a large number of industrial complex in Korea, so these stations were relatively likely to reveal the higher levels of CBs, which generated from various combustion processes, than stations from Busan and Jinhae coasts. In monitoring programs at the same stations, the similar local distribution patterns on the sediment chlorobenzenes were also observed for polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) (Moon et al., 2000a, b). These results indicate that persistent organic pollutants (POPs) such as PCDDs/DFs, PAHs and organochlorine compounds have a similar behavior in the marine environment.

In this study, HCB levels in sediments from the southeastern coasts of Korea were 0.42~1.91 ng/g dry weight. These results were similar or slightly low values compared to HCB levels in sediments from Ise Bay (2.2±1.8 ng/g dry weight), Lake Huron (2 ng/g dry weight) and Lake Erie (3 ng/g dry weight) (Masunaga et al., 1991). HCB levels in sediments from Mediterranean Sea ranged 0.01~39.4 ng/g dry weight (Tolosa et al., 1995), Kaohsiung coast in Taiwan (12.1~31.4 ng/g dry weight; Lee et al., 2000) and Lake Ontario (100±59 ng/g dry weight; Durrsma et al., 1989) with an order of about 10~50 magnitude greater than the study reported here. In the other hand, HCB levels in sediments from Western Baltic Sea (values between 0.01 and 0.23; Dannenberger, 1996) showed lower values than that in this investigation. Consequently, it means that CBs contamination in surface sediments from Korean southeastern coasts was relatively moderate in comparison to other marine environment in the world.

Contributions of chlorobenzene congener groups in surface sediments

Table 1. TOC, grain size and water content in sediments from the southeastern coasts of Korea

		POHANG		BUSAN		ULSAN		JINHAE	
		Range	Mean	Range	Mean	Range	Mean	Range	Mean
TOC (%)		1.02~3.68	1.7	1.37~2.34	2.0	1.11~4.64	1.9	1.05~3.33	2.4
Grain size (%)	Mud	72~92.5	82	80.6~99.2	90.5	38.6~98.3	81.8	77.6~99.7	94.3
	Sand	7.5~28	18	0.8~19.4	9.5	1.7~61.4	18.2	0.3~22.4	5.7
Water content (%)		28.0~50.5	41.2	47.5~55.4	50.9	31.3~57.1	49.6	54.1~65.4	58.9

Mud fraction <63 μm and Sand fraction >63 μm.

Table 2. The average concentrations (standard deviation) of chlorobenzene isomers in sediments from the southeastern coastal areas of Korea (ng/g dry weight)

	POHANG					BUSAN					ULSAN						JINHAE				
	P1	P2	P3	P4	P5	B1	B2	B3	B4	B5	U1	U2	U3	U4	U5	U6	J1	J2	J3	J4	J5
135-TrCB	0.14 (0.16)	0.04 (0.02)	0.07 (0.07)	0.03 (0.02)	0.04 (0.02)	0.10 (0.10)	0.06 (0.004)	0.08 (0.01)	0.09 (0.06)	0.07 (0.05)	0.05 (0.01)	0.05 (0.01)	0.05 (0.02)	0.08 (0.003)	0.03 (0.01)	0.04 (0.01)	0.03 (0.02)	0.11 (0.02)	0.05 (0.03)	0.04 (0.01)	0.02 (0.02)
124-TrCB	1.64 (0.80)	1.55 (1.25)	9.58 (7.12)	2.11 (1.05)	1.47 (0.35)	4.00 (4.31)	2.29 (0.88)	3.04 (1.05)	2.58 (0.22)	2.10 (1.89)	2.96 (0.93)	2.28 (1.83)	3.15 (0.79)	6.22 (2.58)	2.15 (1.34)	1.86 (0.41)	1.70 (1.27)	1.76 (1.51)	1.49 (1.12)	1.19 (0.29)	1.17 (1.08)
123-TrCB	0.74 (0.65)	0.29 (0.20)	1.20 (0.61)	0.32 (0.16)	0.26 (0.10)	0.75 (0.85)	0.35 (0.17)	0.63 (0.08)	0.39 (0.16)	0.53 (0.61)	0.43 (0.32)	0.24 (0.17)	0.42 (0.15)	0.77 (0.42)	0.29 (0.11)	0.26 (0.06)	0.37 (0.31)	0.39 (0.27)	0.24 (0.21)	0.21 (0.04)	0.22 (0.24)
TrCBs	2.52 (1.61)	1.88 (1.47)	10.85 (7.80)	2.46 (1.23)	1.77 (0.47)	4.84 (5.26)	2.70 (1.05)	3.74 (1.14)	3.05 (0.44)	2.70 (2.55)	3.45 (1.27)	2.57 (2.01)	3.62 (0.96)	7.07 (3.01)	2.47 (1.47)	2.16 (0.49)	2.10 (1.61)	2.25 (1.79)	1.78 (1.37)	1.45 (0.34)	1.40 (1.34)
1235+ 1245-TeCB	0.10 (0.06)	0.07 (0.07)	0.15 (0.14)	0.05 (0.02)	0.17 (0.10)	0.20 (0.21)	0.12 (0.07)	0.14 (0.02)	0.07 (0.02)	0.16 (0.19)	0.07 (0.04)	0.09 (0.08)	0.06 (0.01)	0.11 (0.01)	0.05 (0.03)	0.06 (0.03)	0.09 (0.06)	0.06 (0.03)	0.04 (0.03)	0.03 (0.01)	0.04 (0.03)
1234-TeCB	0.12 (0.08)	0.09 (0.07)	0.27 (0.21)	0.07 (0.04)	0.09 (0.04)	0.13 (0.10)	0.19 (0.10)	0.20 (0.02)	0.13 (0.03)	0.25 (0.29)	0.12 (0.08)	0.10 (0.05)	0.10 (0.02)	0.20 (0.02)	0.10 (0.05)	0.10 (0.04)	0.13 (0.09)	0.11 (0.04)	0.08 (0.05)	0.07 (0.02)	0.08 (0.07)
TeCBs	0.22 (0.14)	0.16 (0.14)	0.42 (0.35)	0.12 (0.06)	0.26 (0.15)	0.33 (0.30)	0.30 (0.17)	0.34 (0.04)	0.20 (0.05)	0.41 (0.47)	0.19 (0.12)	0.19 (0.13)	0.16 (0.03)	0.32 (0.03)	0.15 (0.08)	0.16 (0.07)	0.22 (0.14)	0.16 (0.07)	0.11 (0.08)	0.11 (0.03)	0.11 (0.10)
PeCB	0.22 (0.13)	0.17 (0.14)	0.38 (0.33)	0.13 (0.06)	0.16 (0.08)	0.18 (0.09)	0.34 (0.22)	0.31 (0.05)	0.17 (0.05)	0.44 (0.51)	0.31 (0.28)	0.22 (0.16)	0.21 (0.02)	0.32 (0.07)	0.17 (0.06)	0.16 (0.07)	0.22 (0.16)	0.14 (0.03)	0.11 (0.06)	0.09 (0.01)	0.12 (0.09)
HCB	1.17 (0.21)	0.80 (0.64)	0.79 (0.54)	0.55 (0.22)	0.42 (0.11)	1.91 (1.33)	1.27 (1.12)	1.04 (0.16)	0.81 (0.33)	0.95 (0.78)	1.54 (1.50)	0.89 (0.51)	0.70 (0.10)	1.42 (0.45)	0.95 (0.66)	0.73 (0.28)	0.92 (0.80)	0.54 (0.46)	0.53 (0.37)	0.44 (0.29)	0.59 (0.46)
Sum	4.13 (2.08)	3.01 (2.39)	12.45 (9.02)	3.26 (1.58)	2.61 (0.82)	7.26 (4.32)	4.62 (2.57)	5.43 (1.33)	4.24 (0.74)	4.49 (4.32)	5.49 (3.17)	3.86 (2.79)	4.68 (1.11)	9.13 (3.56)	3.75 (2.27)	3.20 (.091)	3.45 (2.71)	3.09 (2.35)	2.53 (1.88)	2.08 (0.59)	2.23 (1.99)

The contributions of CB congener groups to sum of eight isomers (tri- to hexachlorobenzene) in sediments from the southeastern coastal areas of Korea were illustrated in Fig. 3. Generally, the contribution of total CBs for each station showed similar patterns for all stations. This indicates that CBs contamination in analyzed sediments derived from the similar source. These patterns were comparable to a review by Lee et al. (2000) concerning contributions individual chlorobenzene to sum of chlorobenzene in surficial sediments from Kaohsiung coast, Taiwan. The predominant contributors for total CBs were trichlorobenzene groups (68.3±7.1%), whereas tetrachlorobenzenes (5.2±1.7%) and pentachlorobenzene (5.1±1.6%) showed low concentrations. HCB occupied 21.4±5.6% for total CBs. The contributions of trichlorobenzenes in sediments from Pohang and Ulsan coasts were greater than those of Busan and Jinhae coasts due to use as a primary solvent of trichlorobenzenes in a manufacturing process. In particular, Station P3 from Pohang coast was the highest level with 87.2% occupation, which was located close to the steel mill. Many authors reported that this plant were the main source of hazardous contaminants in marine environment (Hutzinger et al., 1985; Abad et al., 1996).

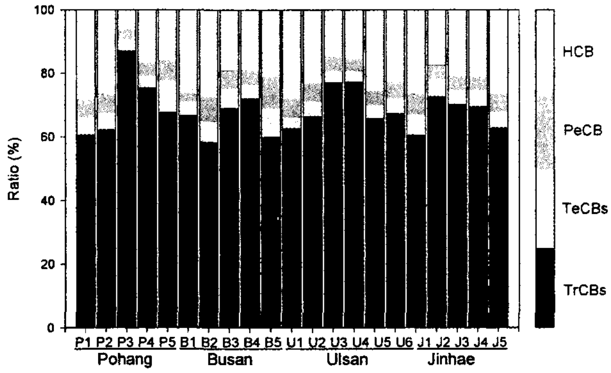

Fig. 3. Contributions of chlorobenzene congener groups in sediment from the southeastern coastal areas of Korea; TrCBs: the sum of 1,2,3-, 1,2,4- and 1,3,5-trichlorobenzene and TeCBs: the sum of 1,2,4,5-, 1,2,3,4- and 1,2,3,5-tetrachlorobenzene, PeCB: pentachlorobenzene, and HCB: hexachlorobenzene.

Correlation analysis

A correlation analysis was carried out to investigate the relationship within each congener group of CBs (Fig. 4). This statistical approach is based on the fact that each pollution source produces a cha-

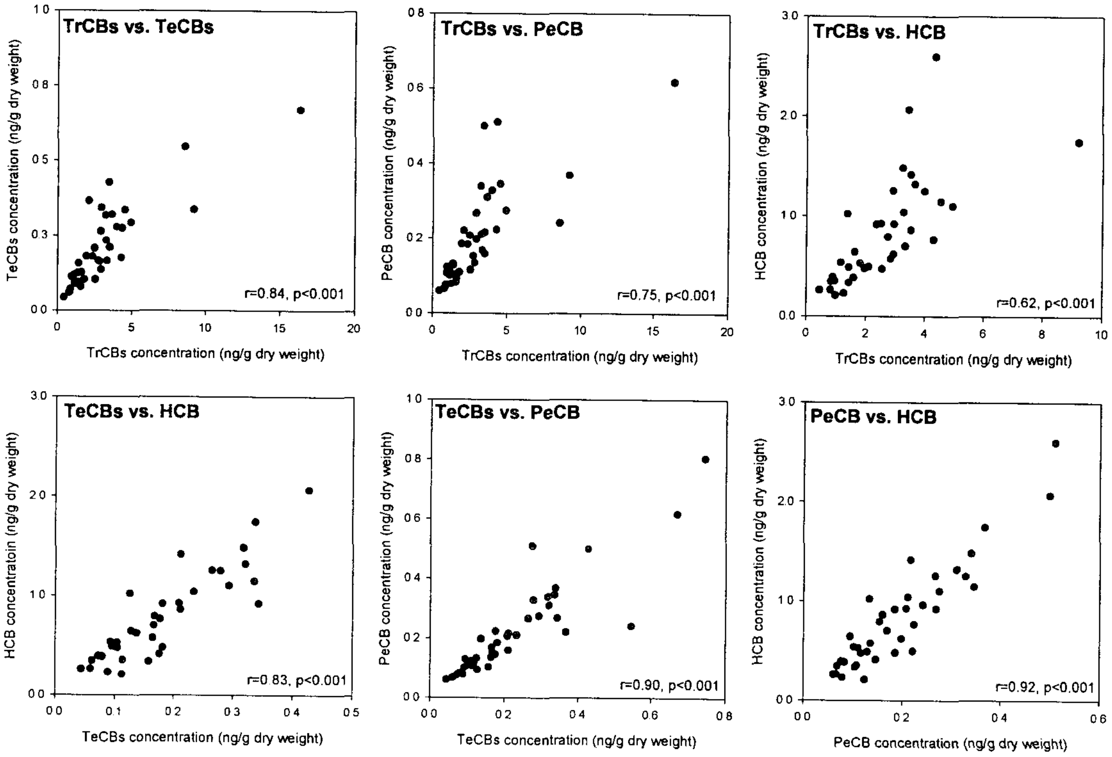

Fig. 4. Relationships within each congener group of chlorobenzenes in sediments from the southeastern coastal areas of Korea; TrCBs: the sum of 1,2,3-, 1,2,4- and 1,3,5- trichlorobenzene and TeCBs: the sum of 1,2,4,5-, 1,2,3,4- and 1,2,3,5- tetrachlorobenzene, PeCB: pentachlorobenzene, and HCB: hexachlorobenzene.

racteristic CBs pattern, so the correlation analysis can give an idea whether they all originate from the same source or not. In general, there was positive significant correlation within CBs species. This finding can be explained by the same contamination source by CBs for the southeastern coasts of Korea. Correlation coefficient for PeCB and HCB represented the highest value ($r=0.92$, $p<0.001$), followed by TeCBs and PeCB ($r=0.90$, $p<0.001$) and TrCBs and TeCBs ($r=0.84$, $p<0.001$). Correlation coefficinets for TrCBs and HCB slightly showed the lower value ($r=0.62$, $p<0.001$). These results seem to be based on various physical and chemical properties such as boiling point or half-lives of each congener group.

There was no apparent significant correlation between the concentration of CBs and the percentage of TOC.

Conclusions

Sediments have been sampled in several areas from Korean coasts to investigate the levels of chlorobenzenes (CBs) contamination. The levels of total chlorobenzene isomers ranged between 2.08 and 12.45 ng/g dry weight. Hexachlorobenzene (HCB) levels in sediments were low or moderate comparable to other countries in the world. The contribution of congener groups to sum of eight CB isomers in sediments showed similar patterns for all stations and a positive correlation within individual CB compound in marine sediments was observed. These results suggest that the CBs contamination of the southeastern coastal areas of Korea derived from the similar source.

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