

## Analysis of Polybrominated Diphenyl Ethers (PBDEs) in Sediments from the Southeastern Coastal Areas of Korea

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Polybrominated diphenylether (PBDEs) congeners, 2,4,4'-TrBDE, 2,2',4,4'-TeBDE, 2,2',4,4',5-PeBDE, 2,2',4,4',5,5'-HxBDE, and 2,2',4,4',5,6-HxBDE, were measured in sediments from 52 stations in the southeastern coastal areas of Korea. Sediment samples were analyzed using gas chromatography coupled to mass spectrometer detector (GC/MSD) with positive electron impact (PEI) mode. New analytical methodology for PBDEs by the isotope dilution method was established using a multi-layer silica gel column chromatography. Total PBDEs levels in sediments for Pohang coast ranged from 1.1 to 33.8 ng/g dry weight (mean 5.3 ng/g dry), from 1.6 to 36.4 ng/g dry weight (mean 5.7 ng/g dry) for Ulsan coast, from 0.8 to 20.3 ng/g dry weight (mean 4.9 ng/g dry) for Busan coast, and from 0.8 to 10.3 ng/g dry weight (mean 4.4 ng/g dry) for Jinhae Bay. PBDEs contamination in surface sediments from Korean southeastern coasts was relatively moderate in comparison to that of other marine environment in the world. The predominant PBDE congeners were 2,2',4,4'-TeBDE and 2,2',4,4',5-PeBDE.

### INTRODUCTION

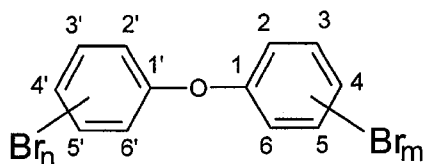
Polybrominated diphenylethers (PBDEs) have been widely used in large quantities as flame-retardant additives for many applications such as television sets, computers, radios, textiles, new synthetic building materials and automobiles (Sellström *et al.*, 1993; Pijnenburg *et al.*, 1995). PBDEs are structurally similar to polychlorinated biphenyls (PCBs) and polybrominated biphenyls (PBBs) (Fig. 1). They are a class of dicyclic aromatic compounds substituted with one to ten bromine atoms. This results in 209 possible congeners depending on the numbers and positions of the bromine atoms and has similar properties with highly lipophilic and hydrophobic characters (WHO, 1994; Alcock *et al.*, 1999). In addition, based on its chemical structure and biological experiments, it is concerned that PBDE may have endocrine disrupting effects (Hoque *et al.*, 1998; Eriksson *et al.*, 1999). Indeed, Safe (1992) developed a value of relative toxic equivalent of individual halogenated aromatic relative to 2,3,7,8-TCDD based on the common receptor-mediated mechanism of action of toxic halogenated aromatic compounds and their structural activity relationship. The derived toxic equivalent proposed

that the brominated aromatic compounds have similar values as PCBs, polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs).

The use of flame-retardants is increasing on a global scale, in 1992, 600,000 tonnes were consumed worldwide with 150,000 tonnes of these being brominated compounds of which 40,000 tonnes were PBDEs, including 30,000 tonnes of the decabromodiphenyl ether (DeBDE) (WHO, 1994). Hence, there are large amounts of PBDEs in environment, which was released from their manufacturing or operating processes into environment (Haglund *et al.*, 1997; De Boer *et al.*, 1998; Lindstrom *et al.*, 1999). Furthermore, when the products containing flame-retardants were incinerated, it has estimated to be able to act as precursors for polybrominated dibenzo-*p*-dioxins (PBDDs) and dibenzofurans (PBDFs).

Since PBDEs were first detected in pike, eel and sea trout samples of Sweden in 1981 (Andersson and Blomkvist, 1981), many researchers have been confirmed the existence of PBDEs in the various samples from marine environment of many countries (Watanabe *et al.*, 1987; Nylund *et al.*, 1992; Sellström and Jansson, 1995; Allchin *et al.*, 1999). In particular, Norén and Meironyté (2000) investigated organohalogen compounds such as PCBs, polychlorinated naphthalenes (PCNs), PCDDs, PCDFs, PBDEs, and pesticides (DDT,

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**Fig. 1.** Structure of polybrominated diphenylethers (PBDEs) and numbering system.

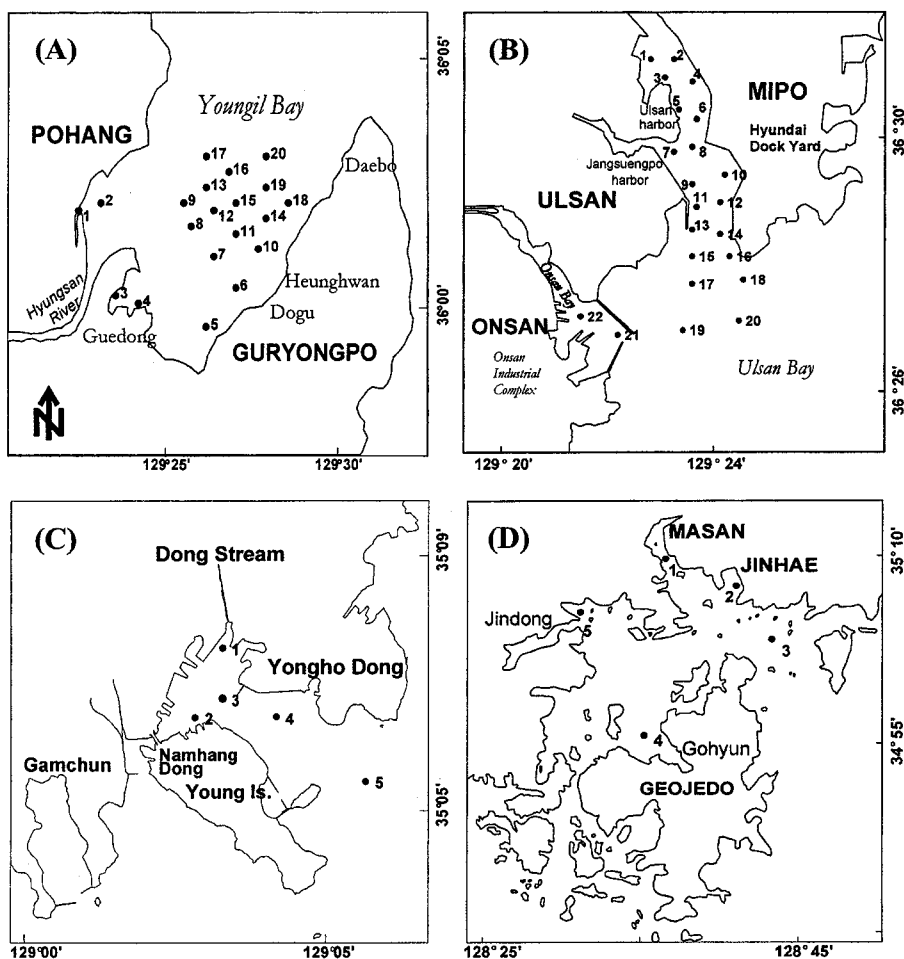
DDE, hexachlorobenzene, dieldrin) as well as methylsulfonyl metabolites of PCBs and DDE in breast milk from women living in the Stockholm region during different periods up to 1997. During the course of 20–30 year the levels of organochlorine compounds in human milk have decreased to various extent. On the contrary to the organochlorine compounds, the concentrations of PBDEs have increased during the period of 1972–1997, indicating a doubling of the levels by 5 year. In addition, considering now a world-wide important environmental problems reaching even remote areas, it is surprising that there is no report on concentrations of PBDEs in envi-

ronment including biological samples in Korea. Thus, prior to risk assessment of PBDEs, PBDEs contamination in marine environment of Korea needs to be investigated. The objective of this study is to develop a routine analytical methodology for PBDEs in marine sediment samples and to investigate the levels of PBDEs in sediments collected from the southeastern coastal areas of Korea using developed method.

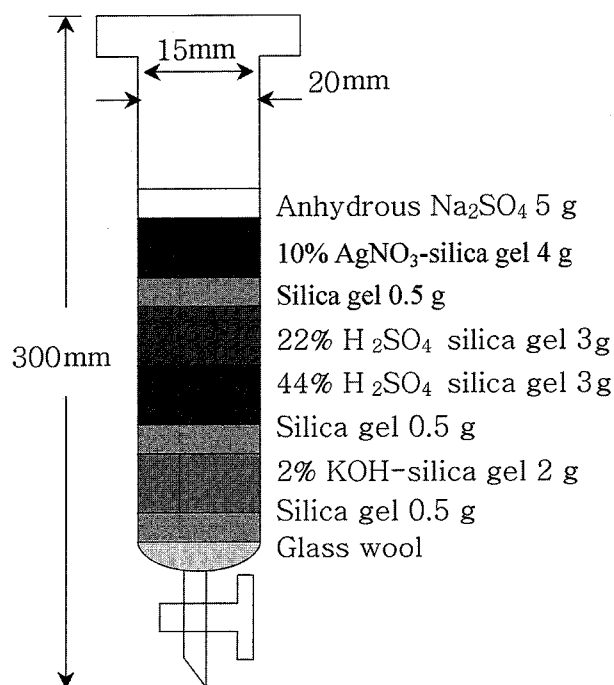
## MATERIALS AND METHODS

### Sampling and sample preparations

Surface sediments (0–5 cm) were sampled at 52 stations from Pohang, Ulsan, Busan and Jinhae coasts, which were located in the southeastern parts of Korea, in November 2000 (Fig. 2). Sediment samples were collected with box-core sampler and then kept frozen at  $-20^{\circ}\text{C}$  until extraction. They were freeze-dried and sieved through 2 mm. Twenty grams of sediments were extracted in a Soxhlet apparatus with



**Fig. 2.** Map showing sampling stations in four coasts. (A): Pohang coast, (B): Ulsan coast (C): Busan coast, (D): Jinhae coast.



**Fig. 3.** Clean-up column chromatography of multi-layer silica gel for PBDEs.

200 mL of toluene (Ultra residue analysis, J. T. Baker) for 24 hours, after being spiking with 5 species as internal standards (MBDE-MXA and MBDE-MXB, Wellington Laboratories). The extracts were reduced to 1–2 mL in a rotary evaporator and then were transferred to *n*-hexane (Ultra residue analysis, J. T. Baker).

Samples were cleaned up on a multi-layer silica-based adsorbents (70–230 mesh, Neutral, Merck) column (15 mm i.d. ×300 mm long) with 160 mL of *n*-hexane. The elution flow was set at 10 mL/min. The extract was passed through adsorbents in the following order; anhydrous sodium sulfate 5 g, silver nitrate impregnated silica gel 4 g, silica gel 0.5 g, 22%

sulfuric acid impregnated silica gel 3 g, 44% sulfuric acid impregnated silica gel 3 g, silica gel 0.5 g, 2% potassium hydroxide impregnated silica gel 2 g, and finally silica gel 0.5 g (Fig. 3).

The purified samples were concentrated to less than 1 mL, and left at a room temperature for one or two days to evaporate to dryness. The residues were dissolved with 20  $\mu$ L of *n*-nonane (Pesticide residue analysis, Fluka) and determined for PBDEs.

### Identification and quantification

Target compounds of PBDEs used in this study were as follows; 2,4,4'-TrBDE (BDE #28), 2,2',4,4'-TeBDE (BDE #47), 2,2',4,4',5-PeBDE (BDE #99), 2,2',4,4',5,5'-HxBDE (BDE #153) and 2,2',4,4',5,6-HxBDE (BDE #154) (Wellington Laboratories). Isotope labelled compounds ( $^{13}\text{C}_{12}$ -2,4,4'-TrBDE,  $^{13}\text{C}_{12}$ -2,2',4,4'-TeBDE,  $^{13}\text{C}_{12}$ -2,2',4,4',5-PeBDE,  $^{13}\text{C}_{12}$ -2,2',4,4',5,5'-HxBDE, and  $^{13}\text{C}_{12}$ -2,2',4,4',5,6-HxBDE, Wellington Laboratories) were used as internal standards. Quantification was performed by the method of relative calibration curves using both native and internal standards.

An Agilent 6890 series gas chromatography (GC) coupled to a 5973N mass selective detector (MSD) was used for the determinations of PBDEs in this study. The mass spectrometer was operated under the selective ion monitoring (SIM) method using tow ion molecular ions for each degree of bromination with the positive electron impact (PEI) mode. A DB 5MS capillary column was used for the separation of PBDE compounds. Samples were injected splitlessly (2  $\mu$ L portion of the total 20  $\mu$ L). Detailed instrumental conditions for PBDEs by GC/MSD used were summarized in Table 1.

Procedural blanks were processed in the same manner as real samples, and they were below 10% of analytes abundance. Blanks were run before and after

**Table 1.** Analytical conditions of PBDEs by GC/MSD.

Instrument	Agilent 6890 GC/MSD 5973N
Column	DB 5 MS, 30 m, 0.25 mm ID, 0.25 $\mu$ m film thickness, J&W Scientific
Injector type	Splitless
Carrier gas	Helium, 1.2 mL/min
Injector temperature	250°C
Column temperature	120°C (2 min) –10°C/min –215°C –3°C/min –270°C –10°C/min –310°C (10 min)
Interface temperature	280°C
Ionization mode	EI+, 70 eV
Injection volume	2 $\mu$ L
Selective ion monitoring	$M^+$ and $(M+2)^+$ , $(M+2)^+$ and $(M+4)^+$

the injection of standards to check for carryover.

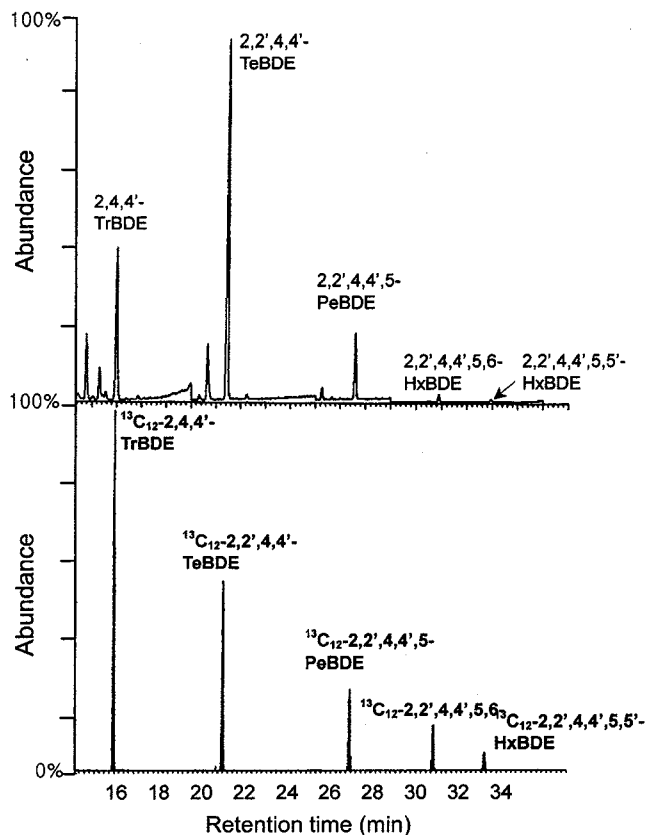
## RESULTS AND DISCUSSION

### *Development of analytical method for the determination of PBDEs*

Due to the similarities of PBDEs with organochlorine aromatic compounds such as PCBs, the analytical method for the determination of organohalogen compounds are also applicable to PBDEs. In most analyses of PBDEs in environmental samples, treatments with concentrated sulfuric acid (Andersson and Blomkvist, 1981; Watanabe *et al.*, 1987) and/or gel permeation chromatography (GPC) using *n*-hexane/methylene dichloride (Haglund *et al.*, 1997; Akutsu *et al.*, 2001) were used for removal of the bulk of organic material or lipid in samples. These procedures are unsuitable for routine analysis for PBDEs in marine sediments because of the use of large volumes of toxic reagents and requirements of long pre-treatment time. In this study, we have established the new analytical methodology for PBDEs by using the isotope dilution method, as same as the analytical method for dioxin analogues. So, we modified a multi-layer silica gel column chromatography used as pre-cleanup method for PCDDs and PCDFs (Moon *et al.*, 2001).

All the spiked isotope compounds were detected with no interfering peak. The average recoveries were in the range of 79±8%, 83±7%, 89±5%, 92±4% for tri-, tetra-, penta- and hexa-brominated diphenylethers, respectively. The calculated detection limit (S/N ratio =3) for individual PBDE in sediment was estimated at 0.01 ng/g dry weight.

Most researchers have used gas chromatography (GC) with electron capture detector (ECD) or gas chromatography coupled to mass spectrometry (GC/MS) with negative chemical ionization (NCI) mode, which is considered to be capable of highly sensitive analysis of organohalogen compounds, to identify and quantify the PBDEs in various environmental samples (Jansson *et al.*, 1987; Nylund *et al.*, 1992; Hartonen *et al.*, 1997; Sjodin *et al.*, 1998; De Boer, 2000; Hori *et al.*, 2000). Recently, some researchers used high resolution gas chromatography/high resolution mass spectrometer (HRGC/HRMS) for PCDDs/DFs analysis because of the great difficulties in rigorous identification of individual isomer of PBDEs (Alaee *et al.*, 2001; Burreau *et al.*, 2001). However, in the present study, the GC/MSD with PEI mode, which



**Fig. 4.** Example of SIM chromatogram of PBDEs in sediments analysed in this study (Station P2). Upper chromatogram is native peak and lower chromatogram is internal standards added into the same sediment samples.

has been used for analysis of as normal organohalogen compounds such as PAHs, PCBs, and pesticides etc., was investigated for determination of PBDEs in marine sediments. Fig. 4 presents the typical chromatograms of the PBDEs in sediments monitored by GC/MSD, showing good separations of target PBDEs.

### *PBDEs in sediments*

Fifty-two marine sediments were analyzed using developed analytical method. Table 2 shows the concentrations of PBDE congeners in sediments collected from the southeastern coasts of Korea. Five PBDE congeners were detected in all sediment samples. The highest level of total PBDEs was found in Station U2 from Ulsan coast and the lowest level was Station B3 from Busan coast. Total levels in sediments from Pohang coast ranged from 1.1 to 33.8 ng/g dry weight with a mean concentration of 5.3 ng/g dry weight. Total PBDEs concentrations in sed-

**Table 2.** The levels of PBDE congeners in sediments from Pohang, Ulsan, Busan and Jinhae coasts in Korea (ng/g dry weight)

St. No.	BDE #28	BDE #47	BDE #99	BDE #153	BDE #154	SUM
P1	0.21	0.39	1.4	0.80	0.63	3.4
P2	0.54	0.57	0.47	0.15	0.18	1.9
P3	0.25	0.35	0.34	0.10	0.16	1.2
P4	0.41	0.44	0.25	0.12	0.18	1.4
P5	0.36	0.52	0.38	0.20	0.13	1.6
P6	0.43	0.55	0.38	0.24	0.19	1.8
P7	0.30	0.43	0.28	0.11	0.14	1.3
P8	0.21	0.39	0.22	0.15	0.15	1.1
P9	2.5	4.4	3.7	1.0	1.2	12.8
P10	0.51	0.67	0.42	0.23	0.36	2.2
P11	2.5	1.9	1.3	0.87	0.55	7.1
P12	0.53	0.88	0.70	0.27	0.15	2.5
P13	0.26	0.51	0.34	0.15	0.14	1.4
P14	1.5	0.68	0.42	0.44	0.35	3.4
P15	0.31	0.38	0.23	0.19	0.17	1.3
P16	4.1	5.4	4.9	1.8	1.6	17.8
P17	4.8	18	7.4	1.7	2.3	33.8
P18	0.52	0.75	0.44	0.18	0.13	2.0
P19	1.8	1.7	1.0	0.50	0.29	5.3
P20	1.3	0.60	0.35	0.35	0.28	2.8
U1	2.0	2.5	3.0	0.86	0.87	9.2
U2	3.3	11	16	2.5	3.8	36.4
U3	2.8	5.3	6.2	1.5	1.4	17.2
U4	0.57	0.70	0.32	0.47	0.43	2.5
U5	1.4	0.95	0.93	0.72	0.63	4.7
U6	2.1	3.1	3.7	1.5	1.7	12.1
U7	2.5	2.5	2.0	1.1	1.3	9.4
U8	0.05	0.66	0.51	2.6	1.1	4.9
U9	0.51	0.68	0.67	0.42	0.22	2.5
U10	0.37	0.64	0.45	0.33	0.33	2.1
U11	0.45	0.65	0.49	0.33	0.24	2.1
U12	0.46	0.76	0.61	0.39	0.37	2.6
U13	0.40	0.61	0.48	0.24	0.22	2.0
U14	0.70	0.82	0.61	0.56	0.52	3.2
U15	0.37	0.66	0.48	0.36	0.42	2.3
U16	0.38	0.80	0.72	0.48	0.41	2.8
U17	0.29	0.48	0.37	0.35	0.41	1.9
U18	0.31	0.36	0.38	0.29	0.29	1.6
U19	0.33	0.50	0.43	0.36	0.42	2.0
U20	0.33	0.43	0.41	0.44	0.50	2.1
U21	1.1	0.82	0.58	0.45	0.40	3.4
U22	0.50	0.59	0.58	0.39	0.32	2.4
B1	0.64	7.7	9.9	1.1	0.98	20.3
B2	0.05	0.85	0.32	0.07	0.03	1.3
B3	0.08	0.27	0.36	0.04	0.03	0.8
B4	0.16	0.31	0.40	0.07	0.01	1.0
B5	0.01	0.81	0.06	0.05	0.06	1.0

**Table 2.** (Continued).

St. No.	BDE #28	BDE #47	BDE #99	BDE #153	BDE #154	SUM
J1	0.14	0.07	0.27	0.33	0.28	1.1
J2	2.5	2.0	1.4	1.2	0.83	7.9
J3	0.15	0.16	0.27	0.14	0.10	0.8
J4	0.09	0.11	0.75	0.10	0.95	2.0
J5	3.1	2.4	2.0	1.9	0.93	10.3

**Table 3.** Comparison of PBDE levels in sediments from other studies (ng/g dry weight)

Locations	2,2',4,4'-TeBDE (mean)	2,2',4,4',5-PeBDE (mean)	References
Pohang coast, Korea	0.35–18 (2.0)	0.22–7.4 (1.3)	This study
Ulsan coast, Korea	0.36–11 (1.5)	0.32–16 (1.8)	This study
Busan coast, Korea	0.27–7.7 (2.0)	0.06–9.9 (2.2)	This study
Jinhae coast, Korea	0.07–2.4 (1.0)	0.27–2.0 (0.9)	This study
Drammenfjord	0.18	0.26	Zegers <i>et al.</i> (2000)
Western Wadden Sea	0.05	0.05	Zegers <i>et al.</i> (2000)
Lake Woserin	0.45	0.75	Zegers <i>et al.</i> (2000)
Haringvliet-east, Netherlands	11	12	De Boer and Dao (1993)
River Meuse, Netherlands	11	13	De Boer and Dao (1993)
River Waal, Netherlands	36	33	De Boer and Dao (1993)
River Humber, UK	21	36	Allchin <i>et al.</i> (1999)
River Nith, UK	0.3–1.7 (0.9)	0.6–1.0 (1.7)	Allchin <i>et al.</i> (1999)
Avonmouth, UK	2.4–3.6 (3.0)	2.9–4.7 (4.0)	Allchin <i>et al.</i> (1999)
Tees estuary, UK	8.9–368 (132)	16–898 (304)	Allchin <i>et al.</i> (1999)
River Skerne, UK	51–239 (134)	85–319 (188)	Allchin <i>et al.</i> (1999)

iments from Ulsan coast were in the range of 1.6–36.4 ng/g dry weight and a mean concentration was 5.7 ng/g dry weight. In Busan coastal areas, total concentrations of PBDEs ranged between 0.8 and 20.3 ng/g dry weight with mean concentration of 4.9 ng/g dry weight. Jinhae Bay showed the range of 0.8–10.3 ng/g dry weight with a mean 4.4 ng/g dry weight.

Until recent, most survey on PBDEs distribution were focused on the compounds of 2,2',4,4'-TeBDE (BDE #47) and 2,2',4,4',5-PeBDE (BDE #99) (Nylund *et al.*, 1992). PBDE levels in sediments measured in this study with those in other countries are summarized in Table 3. In this study, the mean levels of 2,2',4,4'-TeBDE (BDE #47) and 2,2',4,4',5-PeBDE (BDE #99) in sediments from the southeastern coasts of Korea were in the range of 1.0–2.0 ng/g dry weight and 0.9–2.2 ng/g dry weight, respectively. These results were comparable to the PBDE levels in sediments from River Nith and Avonmouth (Allchin *et al.*, 1999). PBDE levels in sediment from Drammenfjord, Western Wadden Sea and Lake Woserin (Zegers *et al.*, 2000) showed the lower values than those in this investigation. However, PBDE levels in

sediments collected from Haringvliet-east, River Meuse, Waal (De Boer and Dao, 1993), Humber, Tees estuary, and River Skerne, which were located near suspected sources including a manufactures of PeBDE and OcBDE, several industries using PeBDE, several landfill receiving wastewaters suspected to contain PBDEs (Allchin *et al.*, 1999), revealed an order of about 10–50 fold greater than those reported here. Consequently, it means that PBDEs contamination in surface sediments from several southeastern coasts of Korea, which are surrounded by industrial complex such as Pohang, Ulsan, Busan and Masan cities, was relatively moderate in comparison to other marine environment in the world.

### **PBDEs compositions**

To compare PBDE homologue patterns of each coast, all data were normalized to the total sum of five PBDE species. The homologue compositions of PBDEs for each coast showed slightly different patterns (Fig. 5). 2,2',4,4'-TeBDE and 2,2',4,4',5-PeBDE were predominant congeners in sediments collected from Pohang,

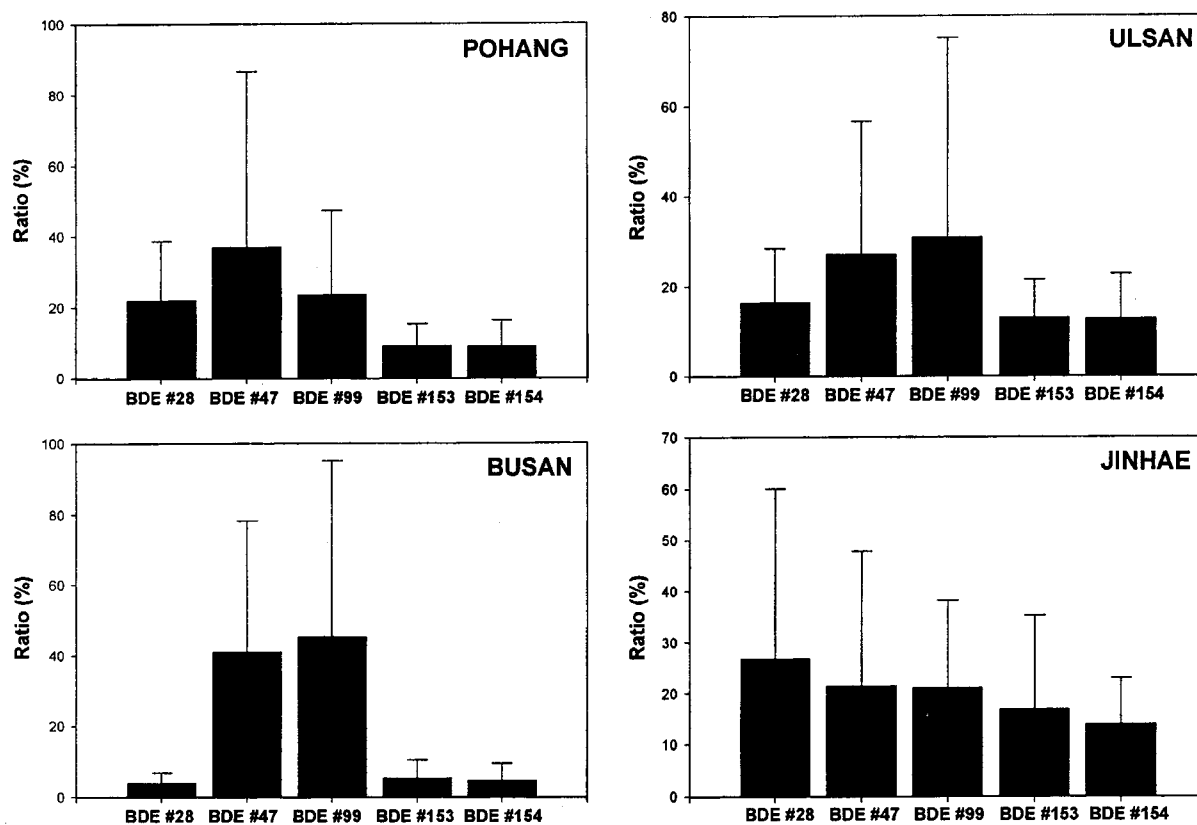


Fig. 5. Average normalized compositions of PBDEs congeners in sediments from Pohang, Ulsan, Busan and Jinhae coasts in Korea. Vertical lines represent standard deviation.

Ulsan and Busan coasts, whereas 2,4,4'-TtBDE (BDE #28) occupied high proportion followed by TeBDE, PeBDE and HxBDE congeners in Jinhae Bay.

Allchin *et al.* (1999) investigated the levels of PBDEs in sediments and biota samples collected from some locations in the UK where brominated flame-retardants are known to be manufactured or handled in large quantities. From these results, the tetra-, penta- and deca-BDE were detected in most sediment samples and 2,2',4,4',5-PeBDE generally occurred at a higher concentrations than TeBDE in sediments from downstream in the UK.

In this study, preliminary contributors in sediment were 2,2',4,4'-TeBDE and 2,2',4,4',5-PeBDE, showing similar findings to Allchin *et al.* (1999).

The main source of tetra- and penta-BDE in the marine environment was known as the debromination by photo- and bio-degradation of DeBDE which occupies approximately 75% of PBDE congeners used as flame-retardants. On the basis of chemical structure, DeBDE is fully brominated and there is only one congener. Commercial DeBDE is typically composed of 97–98% DeBDE (WHO, 1994). Despite DeBDE having

the highest consumption, the majority of the PBDEs found in the environment are lower brominated compounds. This discrepancy can be explained if the higher compounds degrade to tetra, penta and hexa congeners (Rahman *et al.*, 2001). Indeed, DeBDE has been identified as serious pollutants in river sediments from Osaka in Japan, which was located near manufacturing factories of flame-retardants. However, DeBDE was not detected in marine sediments from Osaka Bay, Japan (Watanabe *et al.*, 1987). It indicates that DeBDE was metabolized toward lower brominated compounds such as tetra, penta and hexa congeners (Renner, 2000). Therefore, TeBDE and PeBDE were likely to be dominant congeners in marine ecosystem.

## CONCLUSIONS

Sediments have been sampled in several coastal areas from the southeastern part of Korea, to develop the routine analytical methodology and investigate the levels of polybrominated dipenylethers (PBDEs) contamination using developed method. The levels

of total five species PBDE congeners in sediments from the southeastern coastal areas of Korea ranged between 0.8 and 36.4 ng/g dry weight. These levels in this investigation were relatively moderate in comparison to other marine environment in the world. New analytical methodology for PBDEs by the isotope dilution method was established using a multi-layer silica gel column chromatography and GC/MSD with positive electron impact (PEI) mode. The predominant PBDE congeners were 2,2',4,4'-TeBDE and 2,2',4,4',5-PeBDE.

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